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# Treatment of textile wastewaters by electrocoagulation using iron and aluminum electrodes

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#### Abstract

Treatment of textile wastewaters by electrocoagulation using iron and of aluminum electrode materials has been investigated in this paper. The effects of relevant wastewater characteristics such as conductivity and pH, and important process variables such as current density and operating time on the chemical oxygen demand (COD) and turbidity removal efficiencies have been explored. Furthermore, the electrode and energy consumptions for each electrode have been calculated. The results show that iron is superior to aluminum as sacrificial electrode material, from COD removal efficiency and energy consumption points.

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# 1. Introduction

Wastewater from dyeing and finishing processes, with a chemical oxygen demand (COD) concentration exceeding 1600 mg/l and a strong dark color, is categorized as high strength wastewater [1-3]. It is a significant source of environmental pollution.

The combination of strong color and highly suspended solid content results in high turbidity of the waste effluent. Due to the characteristics of textile wastewater, COD and turbidity removals exhibit similar trends. The total volume of wastewater originating from

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textile dyeing and finishing factories in Turkey is around 150 million metric tons per year, two-third of which is waste in dye and rinse baths [4].

Conventional methods for dealing with textile wastewater consist of various combinations of biological, chemical and physical methods [5,6]. Because of the large variability of the composition of textile wastewaters, most of these conventional methods are becoming inadequate and insufficient. Furthermore, treatment cost of textile waste effluents has been escalating fairly rapidly in recent years. On the other hand, due to the scarcity of space, extremely high land cost and the complexity of handling chemicals in some countries, a simple and efficient treatment process for the textile wastewater is essentially necessary. It should require minimum chemical consumption and space.

One of promising methods for treating hard-to-treat wastewater streams is the electrochemically based. Electrochemical processes (electrolysis and electrocoagulation) have been successfully demonstrated for removing pollutants in various industrial wastewaters [7–9]. Removal mechanisms reported in the electrolysis process generally include oxidation, reduction, decomposition, whereas the mechanisms in the electrocoagulation process include coagulation, adsorption, precipitation and flotation [10–12]. Electrocoagulation utilizes aluminum or iron anodes to produce aluminum or iron hydroxide flocs by reaction at the anodes followed by hydrolysis.

The electrocoagulation is a simple and efficient method for the treatment of many water and wastewaters. It has not been widely accepted because of high initial capital costs as compared to other treatment technologies. In recent years, many investigations have been especially focused on the use of electrocoagulation owing to the increase in environmental restrictions on effluent wastewater [13–15]. Electrocoagulation has been applied successfully to treat potable water [16,17], food and protein wastewater [18], yeast wastewater [19], urban wastewater [20], restaurant wastewater [21,14], tar sand and oil shale wastewater [22], nitrate containing wastewater solutions [23] and arsenic containing smelter wastewater [24]. Electrocoagulation has been proposed in recent years as an effective method to treat wastewater streams from dyeing factories. The electrogenerated flocs separate rapidly and remove color and turbidity from dyeing wastewaters [2,25]. Electrocoagulation treatments of textile dye-containing solutions or wastewater samples have been conducted on a laboratory scale and good removal of COD, color, turbidity and dissolved solids at varying operating conditions have been obtained [2,26-28]. Electrocoagulation has been explored in combination with the addition of polymer solution [29]. The process has been found to be very efficient in COD removal and decoloration with low-energy consumption [30].

Since iron and aluminum electrodes have not been compared in detail for the treatment of textile wastewater, it is the purpose of this study is to compare the treatment of textile wastewater by electrocoagulation using aluminum and iron electrode materials. In addition, the effects of relevant wastewater characteristics, pH and conductivity, and operational variables, current density and treatment time, on the process performance is explored. Four criteria have been selected for this purpose. The two technical criteria which are of primordial importance are COD and turbidity removal efficiencies, and the two criteria which may directly influence the process economy, especially operational costs, are energy and electrode consumptions.

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Table 1
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Characteristics	Value
Chemical oxygen demand (COD) (mg/l)	3422
Total suspended solids (TSS) (mg/l)	1112
Total organic carbon (TOC) (mg/l)	900
Conductivity (µS/cm)	3990
Turbidity (NTU)	5700
pH	6.95

#### 2. Experimental

Wastewater was obtained from a tank containing a mixture of exhaust dyeing solutions at a textile factory in Turkey (Gebze) producing approximately  $1000 \text{ m}^3$  of wastewater per day. The composition of the wastewater is shown in Table 1. The wastewater was first filtered using a screen filter to remove large suspended solids before it was used for the subsequent studies.

The experimental setup is shown in Fig. 1. The thermostated electrocoagulator was made of Plexiglas with the dimensions  $65 \text{ mm} \times 65 \text{ mm} \times 110 \text{ mm}$  at constant stirring speed (200 rpm). There are four monopolar electrodes, two anodes and two cathodes of the same dimensions. Both aluminum (99.53%) or iron (99.50%) cathodes and anodes were made from plates with dimensions of  $46 \text{ mm} \times 55 \text{ mm} \times 3 \text{ mm}$ . The total effective electrode area was  $78 \text{ cm}^2$  and the spacing between electrodes was 11 mm. The electrodes were connected to a digital dc power supply (Topward 6306D; 30 V, 6 A) with potentiostatic or galvanostatic operational options.

All the runs were performed at constant temperature of  $25 \,^{\circ}$ C. In each run,  $250 \,\text{cm}^3$  of the 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 the filtrate was centrifugated at 2000 rpm, 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



Fig. 1. Schematic diagram of experimental setup.

in a solution freshly prepared by mixing  $100 \text{ cm}^3$  HCl solution (35%) and  $200 \text{ cm}^3$  of hexamethylenetetramine aqueous solution (2.80%) [27]. At the end of the run, the electrodes were washed thoroughly with water to remove any solid residues on the surfaces, dried and re-weighted.

COD, total suspended solids (TSS), total organic carbon (TOC) and turbidity were carried out according to the Standard Methods for Examination of Water and Wastewater [31]. The turbidity (NTU) and COD of samples were analyzed using a Shimadzu Model UV-160 double beam spectrophotometer. The pH was measurement by a pH meter (AZ 8601). Conductivity was determined by a conductivity meter (Lutron CD-4303). The pH and conductivity were adjusted to a desirable value using NaOH or H<sub>2</sub>SO<sub>4</sub>, and NaCl (Merck), respectively.

#### 2.1. A brief description of electrocoagulation

Generally, three main processes occur serially during electrocoagulation:

- (a) electrolytic reactions at electrode surfaces,
- (b) formation of coagulants in aqueous phase,
- (c) adsorption of soluble or colloidal pollutants on coagulants, and removal by sedimentation or floatation.

The inherent complexities of the above processes, and also the presence of secondary processes, makes electrocoagulation quite complex in nature.

Main reactions at the electrodes are:

$$Al \rightarrow Al_{(aq)}^{3+} + 3e \quad (anode)$$
 (1)

$$3H_2O + 3e \rightarrow \frac{3}{2}H_2 + 3OH^-$$
 (cathode) (2)

Meanwhile, if anode potential is sufficiently high, secondary reactions may occur also, such as direct oxidation of organic compounds and of  $Cl^-$  ions present in wastewater [10]:

$$2Cl^{-} \rightarrow Cl_{2} + 2e \tag{3}$$

The chlorine produced is a strong oxidant that can oxidize same organic compounds and promote electrode reactions. In addition, the cathode, may be chemically attacked by  $OH^-$  ions generated during H<sub>2</sub> evolution at high pH values [25]:

$$2\text{Al} + 6\text{H}_2\text{O} + 2\text{OH}^- \rightarrow 2\text{Al}(\text{OH})_4^- + 3\text{H}_2 \tag{4}$$

 $Al_{(aq)}^{3+}$  and  $OH^-$  ions generated by electrode reactions (1) and (2) react to form various monomeric species such as  $Al(OH)^{2+}$ ,  $Al(OH)_2^+$ ,  $Al_2(OH)_2^{4+}$ ,  $Al(OH)_4^-$ , and polymeric species such as  $Al_6(OH)_{15}^{3+}$ ,  $Al_7(OH)_{17}^{4+}$ ,  $Al_8(OH)_{20}^{4+}$ ,  $Al_{13}O_4(OH)_{24}^{7+}$ ,  $Al_{13}(OH)_{34}^{5+}$ , which transform finally into  $Al(OH)_3$  according to complex precipitation kinetics [26,32–34].

On the other hand, electrogenerated ferric ions may form monomeric ions, ferric hydroxo complexes with hydroxide ions and polymeric species, depending on the pH range. These are:  $FeOH^{2+}$ ,  $Fe(OH)_2^+$ ,  $Fe_2(OH)_2^{4+}$ ,  $Fe(OH)_4^-$ ,  $Fe(H_2O)_2^+$ ,  $Fe(H_2O)_5OH^{2+}$ ,  $Fe(H_2O)_4(OH)_2^+$ ,  $Fe(H_2O)_8(OH)_2^{4+}$ ,  $Fe_2(H_2O)_6(OH)_4^{2+}$ , which transform finally into  $Fe(OH)_3$  [33–35].

Formation rates of the different species play an important role in the decolorization process. Several interaction mechanisms are possible between dye molecules and hydrolysis products and the rates of these depend on pH of the medium and types of ions present. Two major interaction mechanisms have been considered in recent years: precipitation and adsorption, each one being proposed for a separate pH range. Flocculation in the low pH range is explained as precipitation while it is explained as adsorption in the higher pH range (>6.5) [26,32].

Precipitation:

$$DYE + monomeric Al \rightarrow [DYE-monomeric Al]_{(s)} \quad (pH = 4.0-5.0)$$
(5)

DYE + polimeric Al 
$$\rightarrow$$
 [DYE-polimeric Al]<sub>(s)</sub> (pH = 5.0-6.0) (6)

Adsorption:

$$DYE + Al(OH)_{3(s)} \rightarrow \rightarrow [particle]$$
(7)

$$[DYE-polimeric Al]_{(s)} + Al(OH)_{3(s)} \rightarrow \rightarrow \rightarrow [particle]$$
(8)

Freshly formed amorphous Al(OH)<sub>3</sub> "sweep flocs" have large surface areas which are beneficial for a rapid adsorption of soluble organic compounds and trapping of colloidal particles. These flocs polymerize as [32]

$$n\mathrm{Al}(\mathrm{OH})_3 \to \mathrm{Al}_n(\mathrm{OH})_{3n}$$
 (9)

and are removed easily from aqueous medium by sedimentation and  $H_2$  flotation. The same mechanism is also valid for iron.

# 3. Results and discussion

This study is primarily focused on some crucial questions which must find suitable answers before process the design step:

- What is the best sacrificial electrode material for a given textile wastewater?
- Is there any need for the adjustment of pH and conductivity of the wastewater before the electrocoagulation stage?
- What are the optimum values of the operational variables such as current density, operating time?

For comparative purpose, the same experiments have been run in the same setup with both materials. Based on preliminary experimental results, the effects of pH and conductivity have been explored at constant current density  $100 \text{ A/m}^2$  and operating time of 10 min.

### 3.1. Effect of initial pH

It has been established that pH is an important operating factor influencing the performance of electrocoagulation process [1,21,27]. Generally, the pH of the medium changes during the process, as observed also by other investigators [14,17]. This change depends on



Fig. 2. (a) Effect of initial pH on COD and turbidity removal by aluminum electrodes. (b) Effect of initial pH on COD and turbidity removal by iron electrodes.

the type of electrode material and on initial pH. As seen in Fig. 2(a), in the case of aluminum, the final pH is higher for initial pH < 8, and above this point the final pH is lower. Furthermore, for iron, the final pH is always higher than initial pH. The difference between initial and final pH values diminishes for initial pH > 8 (see Fig. 2(b)). These results suggest that electrocoagulation exhibits some pH buffering capacity, especially in an alkaline medium [14].

The effect of initial pH on the COD and turbidity removal efficiencies is presented in Fig. 2(a) for the aluminum electrode. As seen, for pH < 6, the turbidity removal is as high as 98% and between 65 and 61% for COD removal. Turbidity and COD removals drop dramatically at pH > 6. On the other hand, for iron electrodes, as seen in Fig. 2(b),



Fig. 3. Effect of initial pH on energy consumption.

in the range 3–7 initial pH, the turbidity and COD removals reach 98–75 and 77–47%, respectively. They then drop to 87 and 26% at pH > 10.

For both materials, it is clear that COD and turbidity removals show the same trend. The highest removal efficiencies have been obtained with aluminum in acidic medium with pH < 6, while iron is more efficient in neutral and alkaline medium, especially between 6 < pH < 9.

Fig. 3 shows the specific energy demand in relation for aluminum and iron electrodes during the electrocoagulation, measured in kWh consumed per kg COD removed. Except



Fig. 4. Effect of initial pH on electrode consumption.

for highly acidic and basic mediums, iron electrodes are more energetically efficient, as high as 90% lower than aluminum. Between pH 5 and 9, the energy consumption is almost constant at 0.65 kWh/kg COD, for the iron case.

Fig. 4 shows the electrode consumption per kg of COD removed, in relation to initial pH in electrocoagulation. This economically important process parameter is higher for the iron electrode. Meanwhile, the difference is not very dramatic if market prices of iron and aluminum are taken into account. On the other hand, if the electrode consumption is recalculated on atom gram basis, by considering atomic weights (Fe 56.8, Al 26.9), it is seen that the situation is reversed; for example, for 5 < pH < 9, a nearly constant



Fig. 5. (a) Effect of wastewater conductivity on COD and turbidity removal by aluminum electrodes. (b) Effect of wastewater conductivity on COD and turbidity removal by iron electrodes.



Fig. 6. Effect of wastewater conductivity on energy consumption.

consumption value of 3.5 at. g/kg COD is obtained for iron, which is approximately 15–20% lower than those of aluminum in the same pH range. This result indicates that iron is more efficient than aluminum, for COD removal. This probably result from the differences in the mechanisms of COD removal for the iron and aluminum electrodes. The COD from textile wastewater using aluminum electrodes are mainly removed by electrocoagulation, while the COD removal by iron electrodes is due to the collective effect of electrocoagulation and electrooxidation.



Fig. 7. Effect of wastewater conductivity on electrode consumption.

# 3.2. Effect of conductivity

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Due to the chemical substances added at a high concentration from dyeing and finishing processes in the textile industry, the textile wastewaters have a broad variation in ionic strength. The greater ionic strength will generally cause an increase in current density at the same cell voltage, or the cell voltage decreases with increasing wastewater conductivity at constant current density. Therefore, it is necessary to investigate the effect of wastewater conductivity on electrocoagulation in terms of COD and turbidity removals, electrode and energy consumptions.



Fig. 8. (a) Effect of current density on COD and turbidity removal by aluminum electrodes. (b) Effect of current density on COD and turbidity removal by iron electrodes.



Fig. 9. Effect of current density on energy consumption.

The conductivity of the wastewater is adjusted to the desired levels by adding an appropriate amount of NaCl or deionized water. This adjustment has shown negligible effect on the initial pH of the wastewater, approximately 0.3 pH units, with mean pH value of 6.8.

For the aluminum and iron electrodes, Fig. 5(a) and (b) shows the effect of wastewater conductivity on the performance of the electrocoagulation process. As seen, for both electrode materials, the turbidity removal efficiency remains almost unchanged in the conductivity



Fig. 10. Effect of current density on electrode consumption.

range of  $1000-4000 \,\mu$ S/cm. The COD removal efficiency with the aluminum electrode is slightly reduced, whereas for the iron electrode, it is slightly enhanced with increasing conductivity, as also reported by Lin and Peng [2].

The energy and electrode consumptions for aluminum and iron electrodes are shown in Figs. 6 and 7, respectively, as a function of conductivity. It is clear that, for aluminum, the energy consumption is higher and electrode consumption is lower. For both electrodes, the energy and electrode consumptions decrease with increasing wastewater conductivity. In conclusion, it may be said that higher conductivity is more desirable for high process performance.



Fig. 11. (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.

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According to the experimental results, it may be concluded that no adjustments are needed for the pH and conductivity of the wastewater, and the effects of operational variables on process performance may be explored with wastewater as discharged from the textile dyeing plant.

#### 3.3. Effect of current density

Fig. 8(a) and (b) depicts the effect of current density on COD and turbidity removal efficiencies, for iron and aluminum electrode materials with operating time constant at 10 min. In the case of aluminum, minimum  $150 \text{ A/m}^2$  is required for good efficiencies, with a charge loading approximately equal to  $28 \text{ F/m}^3$ . In the case of iron,  $80-100 \text{ A/m}^2$  is sufficient with a charge loading  $17 \text{ F/m}^3$ . On the other hand, as seen in Fig. 9, for a current density  $100 \text{ A/m}^2$ , aluminum electrode consume 67% more energy then iron electrode; this value reaching 130% for a current density  $150 \text{ A/m}^2$ . Finally, the electrode material consumption is given in Fig. 10. The consumption is nearly equal for iron and aluminum at a current density of  $80 \text{ A/m}^2$ . These efficiencies and consumption values show the superior performance of iron over aluminum as electrode material, at a current density between 80 and  $100 \text{ A/m}^2$  as optimal value, for the wastewater studied in this paper.

#### 3.4. Effect of operating time

To explore the effect of operating time, the current density is hold constant at  $100 \text{ A/m}^2$ . As seen in Fig. 11(a), the aluminum electrode requires 15 min for good removal efficiencies. This time is 50% more than for the iron electrode, according to Fig. 11(b). Finally, the energy and electrode material consumptions are given comparatively for two materials, in Figs. 12 and 13, respectively.



Fig. 12. Effect of electrocoagulation time on energy consumption.



Fig. 13. Effect of electrocoagulation time on electrode consumption.

When Figs. 8–10 on one hand, and Figs. 11–13 on the other hand, are compared, respectively, it is seen that the effect of current density and operating time on performance criteria are very similar, this means that two variables may be combined as a single variable, charge loading expressed as Faraday per unit volume (or mass) of wastewater, and this may simplify greatly the process optimization studies.

#### 4. Conclusion

The use of iron and aluminum as sacrificial electrode materials in the treatment of textile wastewater by electrocoagulation has been found to be pH dependent. According to the results, in acidic medium, pH < 6, COD and turbidity removal efficiencies of aluminum are higher than those of iron, while in neutral and alkaline medium iron is preferable. High conductivity favors high process performances. On the other hand, for the same turbidity or COD removal efficiencies, iron requires a current density of 80–100 A/m<sup>2</sup>, while aluminum requires  $150 \text{ A/m}^2$  for a operating times of 10 min. The experiments show that operating time and current density exhibit similar effects on the process performances, on electrical energy and electrode consumption values. Thus, charge loading the product of these two process variables, may be used more conveniently in process design and optimization tasks. Finally, the energy consumption kWh per kg COD removed is lower with iron, while the electrode consumption per kg COD removed is lower generally with aluminum. It is clear that these two important operating costs will strongly influence the decision about the type sacrificial electrode material for given wastewater characteristics, and COD and turbidity removal levels imposed by environmental restrictions about process effluents.

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