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Original Research

Operating Cost Analysis and Treatment of Domestic Wastewater by Electrocoagulation Using Aluminum Electrodes

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Abstract

In this study, treatment of domestic wastewater (DWW) by electrocoagulation (EC) was investigated. The samples were supplied from the Sivas domestic wastewater pretreatment plant. The experimental studies were conducted to determine the optimum operating conditions such as electrode type, initial pH, current density, and EC time. Aluminum and iron electrodes were used, and aluminum electrodes were preferred to iron since it had a higher removal rate of COD, turbidity, and phosphate than the iron electrodes. At the end of these studies, the optimum operating conditions were found for original pH (7.8), 100 A/m^2, and 10 min EC time and obtained removal efficiencies of COD, turbidity, and phosphorus (72%, 98%, and 98%), respectively. Besides the operating costs investigated in the present study were the energy cost of EC and the material cost because of the consumption of aluminum electrodes. Operating costs varied from 0.03 to 1.67 \$/m^3 and 0.44 to 3.85 \$/m^3 wastewater treatment at 10-150 A/m^2 and 5-40 min, respectively.

Keywords: domestic wastewater, electrocoagulation, energy and electrode consumption, COD removal, phosphorus removal

Introduction

Discharging wastewater without treatment into surface water resources can affect aquatic life negatively. Especially the amount and concentration of wastewater determine how they harm the intake habitat. In general, domestic wastewater is one of the largest pollution resources due to its volume and concentration. Traditionally, biological treatment methods are used in treatment of domestic wastewater. The most commonly used among these methods is the classical actived sludge system, which is an aerobic treatment method. This biological process exploits the diverse metabolic reactions of microorganisms in the degradation of organic matter [1, 2]. Also in this process, organic matters and suspended solid matters can be removed at high rates [3], but nitrogenous compounds and phosphate are insufficiently removed. However, these compounds can be removed through advanced treatment systems. The processes in which aerobic and anaerobic systems are combined (nitrification and denitrification) are generally used for this purpose. The actived sludge process has some disadvantages like continuous need for requiring continuous air supply, high operational and investment costs, sensitivity against shock toxic loadings, longer treatment time, and ultimate sensitivity of microorganisms against pH and temperature.

In recent years, new processes for efficient and adequate treatment of various wastewaters with relatively low costs have been needed due to strict environmental regulations. At this point, the process has attracted a great deal of attention in treating various wastewaters because of its ver-

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satility and environmental compatibility. The EC process has such advantages as simple equipment, easy operation, a shortened reactive retention time, no chemical additions, and decreased amount of precipitate or sludge, which sediments rapidly. Advantages of EC are simple equipment, ease of operation, a shorter retention time, high sedimentation velocities, and reduced amount of sludge. Therefore, in this study, EC process treatment of domestic wastewater by EC was studied for treatment in shorter time and more cost-effective ways.

Electrocoagulation (EC) has been practiced in various types of wastewater in recent years and high efficiencies have been achieved. Researchers have achieved successful results in treatment of landfill leachate wastewater [4], restaurant wastewater [5], food and protein wastewater [6], pulp and paper mill wastewater [7, 8], textile wastewater [9-11], saline wastewater [12], fluoride-bearing wastewater [13], dye stuff-bearing wastewater [14-18], nitrate- and arsenic-bearing wastewater [19, 20], and phosphate-bearing wastewater [21]. When the literature is scanned of studies of treatment by EC of DWW, a few studies were found. Similarly, one publication by Kurt et al. [22] investigated treatment of DWW by EC using Fe-Fe electrodes. According to this study, the EC process for the treatment of DWW can be put forward as an advanced treatment method because of its efficiency, low energy requirement, and lower and more stable sludge production compared with conventional treatment methods [22]. In another study, in France, an electrocoagulation and flotation system were joined to study the membrane process. This study showed that a combination of electrocoagulation and a flotation system with microfiltration could increase removal efficiency. In this process, it is possible to increase by more than turbidity removal, COD removal, and SS removal in comparison with electrocoagulation alone [23]. In another study, anaerobic stabilization of sludge produced during DWW treatment by electrocoagulation was investigated, with COD removal efficiency in the range 67-91%. But anaerobic digestion of sludge was not recommended as an appropriate stabilization method [24]. But these studies did not investigate in detail the effects of electrode type, initial pH, current density, and EC time on COD, turbidity, and phosphorus on removal. Therefore, we undertook the current study.

The EC process consists of occurring metallic hydroxide flocs within the effluent to be cleaned, by electrodissolution of soluble anodes. The characteristics of EC are simple equipment and easy operation, a brief reactive retention period, and negligible equipment for adding chemical and decreasing the amount of sludge [25]. Usually, electrode materials for EC are aluminum and iron. EC has three main processes:

1. electrolytic reaction at the surface of electrodes,
2. formation of coagulants in the aqueous phase,
3. adsorption of soluble or colloidal pollutants on coagulants, and removal by sedimentation or floatation [26].

When iron and aluminum electrodes are used, the generated Fe^{3+} or $Al_{(ag)}^{3+}$ ions will immediately undergo further spontaneous reactions to produce corresponding hydroxides and/or polyhydroxides. For example, $Al_{(ag)}^{3+}$ ions on hydrolysis may generate $Al(H_2O)_6^{3+}$, $Al(H_2O)_5\ OH^{2+}$, $Al(H_2O)_4\ (OH)^{2+}$, and the hydrolysis products may form many monomeric species such as $Al(OH)^{2+}$, $Al(OH)_2^{+}$, $Al_2(OH)_2^{4+}$, and $Al(OH)_4^{-}$, and polymeric species such as $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$, and $Al_{13}(OH)_{34}^{5+}$ in a wide pH range. Similarly, ferric ions generated by electrochemical oxidation of iron electrode may form monomeric ions, $Fe(OH)_3$ and hydroxil complexes, namely $Fe(H_2O)_6^{2+}$, $Fe(H_2O)^{2+}$, $Fe(H_2O)_4(OH)^{2+}$, $Fe_2(H_2O)_8(OH)_2^{4+}$, and $Fe_2(H_2O)_6(OH)_4^{4+}$. The formation of these complexes depends strongly on solution pH [27-29]. Above pH>9, $Al(OH)^{4-}$ and $Fe(OH)^{4-}$ are the dominant species.

When aluminum is used as an electrode material, the reactions are as follows:

At the Anode;

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

At the Cathode;

$$3H_2O + 3e^- \rightarrow \frac{3}{2}H_{2(g)} + 3OH^- \quad (2)$$

In the Solution;

$$Al_{(aq)}^{+3} + 3H_2O \rightarrow Al(OH)_3 + 3H^+(aq) \quad (3)$$

$$nAl(OH)_3 \rightarrow Al_n(OH)_{3n} \quad (4)$$

These insoluble Aluminum hydroxides react with the suspended and/or colloid solids and precipitate. Coagulation, adsorption, precipitation, and flotation are the removal mechanism of the EC process.

In this research, treatment of DWW by EC process was investigated. For this purpose, a reactor was designed in which mono-polar aluminum and iron electrodes connected in parallel were used. The effect of electrode type, initial pH, density of current, and EC time on COD, turbidity and phosphorus removal efficiency were investigated. Also, operating cost (energy and electrode consumption) of the EC process was calculated.

Experimental Methods

Wastewater Source and Characteristics

Domestic wastewater used in this study has been provided from the Sivas DWW pretreatment plant that serves some 300,000 people in Sivas by producing approximately 50,000 m^3 of wastewater per day. Wastewater samples were taken from the outlet of the grit chamber. The characteristics of the wastewater used in this study are given in Table 1.

Experimental Setup

The experimental set up of the electrocoagulation study was given in Fig. 1. The dimensions of the reactor are

Table 1. General characteristics of domestic wastewater.

Parameter	Value
pH	7.8
COD (mg/L)	350
BOD (mg/L)	210
Suspended Solids (mg/L)	150
Phosphate (PO_4-P) (mg/L)	12.9
Conductivity (μS/cm)	1330
Turbidity (NTU)	98
Alkalinity (mg/L)	435

100x100x130 mm, and the reactor was produced from plexiglass with double sides. The reactor was designed with a jacket to keep the temperature stable during the experiments and continuous water circulation was provided. Four electrode materials were used and the electrodes were placed as monopolar and parallel in the reactor. The spaces between the electrodes were adjusted as 20 mm, and the electrodes were immersed in the wastewater completely. Electrode materials were chosen as 48x72x3 mm with the 216 cm^2 active surface area aluminum plates and 50x72x2 mm with the 210 cm^2 active surface area of iron. Aluminum plates consist of 99.53% Al and the iron plates are made of 99.32% Fe content. Current and voltage were controlled by a digital power supplier (GPC 6030D).

All the experiments are conducted with 1,000 ml wastewater under 25ºC temperature with a mixing speed of 250 rpm. After each set of experiments, electrodes were washed for the removal of the residuals on the surface and weighed. The sampled wastewater was filtered in preparation for chemical analysis.

Analytical Procedures

COD, turbidity, pH, phosphate, and SS determinations were carried out as proposed by standard methods [30].

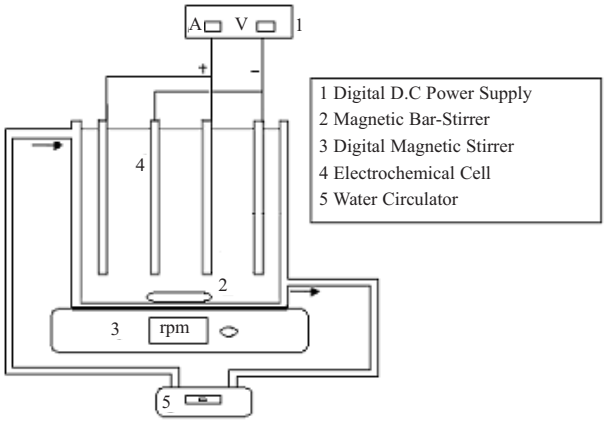

Fig. 1. Schematic diagram of experimental setup.

The COD of samples were analyzed using a Chebios Model UV-VIS optimum one double beam spectrophotometer. The turbidity of samples was analyzed using an HF Model Micro TPI. The phosphate of samples were analyzed using a Merck NOVA60 spectrophotometer. pH was measured by a pH meter (Consort Model C931). Conductivity was determined by a conductivity meter (WTW Model 340İ). The pH was adjusted to a desirable value using NaOH or H_2SO_4 (Merck).

Results and Discussion

This study is mainly focused on the treatment of DWW for determining effects of the basic operating parameters on system performance. Therefore, COD, turbidity, phosphorus, and operating cost (electrodes and energy consumption) were investigated in terms of selection of electrode material, initial pH, current density, and EC time in order to determine the optimum operating conditions for maximum removal of COD, turbidity, and phosphate.

Operating cost is one of the most important parameters in the EC process because it effects the application of any method of wastewater treatment. The operating cost includes material (mainly electrodes) cost, electrical energy cost, labor, maintenance, and other costs. The latter costs items are largely independent of the electrode material [31-34]. Thus, in this study the operating cost was calculated with electrodes and electrical energy costs. So both energy and electrode consumption costs are taken into account as major cost items. Calculation of operating cost is expressed as:

$$\text{Operating Cost} = X\,\text{Energy}_{\text{consumption}} + Y\,\text{Electrode}_{\text{consumption}}$$

...where $\text{Energy}_{\text{consumption}}$ and $\text{Electrode}_{\text{consumption}}$ are consumption quantities per m^3 of treated wastewater. Unit prices, *X* and *Y*, given for the Turkish Market, September 2009, are: electrical energy price 0.06 US $/kWh, electrode material price 1.80 US $/kg for aluminum.

Selection of Electrode Material

The selection of electrode material is important. The most common electrode materials for EC are aluminum and iron. They are cheap, readily, available and effective [5, 28]. Metal electrodes are dissolved during the EC process, which occurs with coagulant species and metal hydroxides. Metal anode dissolution is accompanied by hydrogen gas evolution at cathodes, the bubbles capturing and floating the suspended solids formed and thus removing contaminants [10, 25, 28].

The investigation of treatment efficiency for aluminum and iron electrodes was tested for removal efficiency of COD, turbidity, and phosphate. All experimental studies were carried out under the same conditions, which was initial pH 7.8 (orj.), current density 75 A/m^2, and EC time 10 minutes. Fig. 2 compares treatment efficiency for these two kinds of electrodes under the same conditions. The EC

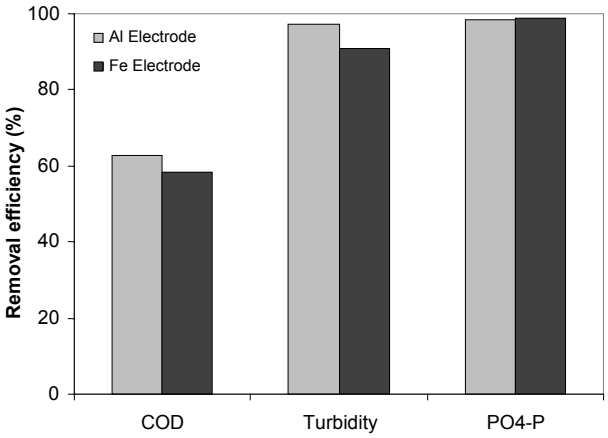

Fig. 2. The comparison of electrode material on the treatment efficiency of the domestic wastewater (pH 7.8, current density 75 A/m^2 and EC time 10 min).

process using aluminum electrodes was obtained to be more effective than for iron electrodes. The results for COD (63%), turbidity (97%), and phosphorus (98%) were removed.

The effluent treatment of aluminum electrodes was found to be more clear and stable than iron electrodes. The effluent EC process using iron electrodes appeared greenish first, and then turned yellow and turbid. The green and yellow colors must have resulted from Fe^{2+} and Fe^{3+}. Fe^{2+} is the common ion generated in situ of electrolysis of iron electrode. It has relatively high solubility at acidic or neutral conditions and can be oxidized easily into Fe^{3+} by dissolved oxygen in water. Furthermore, Fe^{3+} creatives in yellow fine particles of $Fe(OH)_3$ and is difficult to settle [5, 29, 30], and iron electrodes corrode. Hence, it is clear that aluminum electrodes are better than iron electrodes. Consequently, all experiments were carried out with aluminum electrodes.

Effect of Initial pH

It has been established in previous studies that pH is an important factor influencing the treatment performance of the EC process [5, 16, 25, 29]. The pH was adjusted to a desirable value using NaOH or H_2SO_4 and varied in the range 3-7.8 (pH 7.8 is original pH domestic wastewater). Fig. 3 shows the removal efficiencies of COD, turbidity, and phosphorus as a function of initial pH.

As seen clearly from the figure, the removal of COD and turbidity increases with initial pH in the range 3-7.8. The maximum removal of COD and turbidity were obtained at original pH (7.8) of domestic wastewater. As the pH increased from 3 to 7.8, COD removal reached 63%. The removal of phosphate showed which pH effect is not significant in the range 3-7.8. When all parameters are investigated, pH 7.8 is obtains removal of COD, turbidity, and phosphate more than low pH. And this pH was an economical and effective treatment for DWW.

After electrocoagulation, Fig. 3 shows the pH change of the wastewater. When the initial pH is low, the final pH increases [5, 31]. Initial pH is increased from 3 to 7.8, the final pH increased approximately from 4.7 to 8.5 since OH^- ion assembles in aqueous solution during the EC process. Other investigators attribute the pH increase to hydrogen evolution at cathodes [6, 34]. In fact, besides hydrogen evolution, the formation of $Al(OH)_3$ near the anode would release hydrogen ions, leading to a decrease of pH. Furthermore, oxygen evolution reaction also leads to pH decrease.

Effect of Current Density

The effect of current density is another important parameter for pollutant removal in the electrocoagulation process that effects the metal hydroxide concentration formed during the process. High current density especially causes both decomposition of the electrode material and an increase in pollutant removal [31]. The effect of current density on the treatment of the DWW shown in Fig. 4 was investigated by varying the applied current to the wastewater in the same conditions (pH 7.8, EC time 10 min). The density of the current was applied between the range of 10 to150 A/m^2 in order to assess the effect of current density

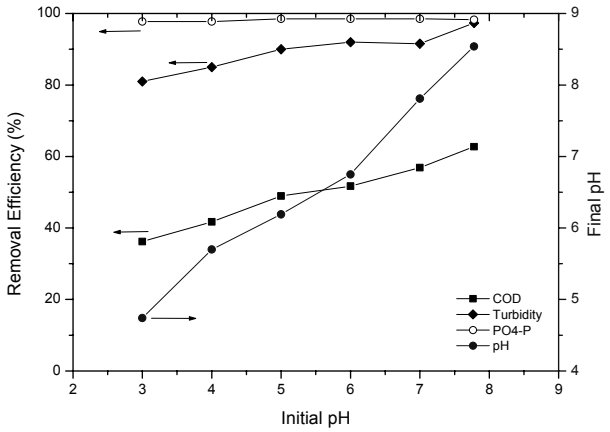

Fig. 3. Effect of initial pH on COD, turbidity, and phosphate removal (Current density 75 A/m^2 and EC time 10 min).

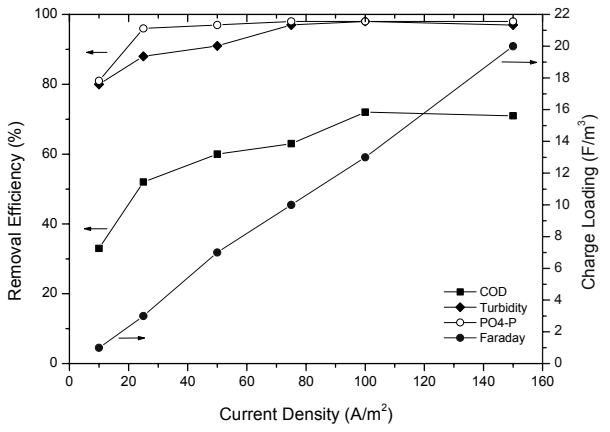

Fig. 4. Effect of current density on COD, turbidity, and phosphate removal (pH 7.8 and EC time 10 min).

on COD, turbidity, and phosphate. Charge loading is increased from 1.3 to 20 Faradays/m^3 under these conditions. The removal of pollutants increased rapidly from 33 to 71% COD, from 80 to 98% turbidity, and from 81 to 98% phosphorus, respectively.

For the same operating conditions, consumptions of energy and electrode material are presented in Fig. 5 [31, 32].

Calculation of energy consumption is expressed as (Eq. 5):

$$\text{Energy}_{\text{consumption}} = (V \text{ x } I \text{ x } t) / v \quad (5)$$

...where $\text{Energy}_{\text{consumption}}$, V, I, t, and v are energy consumption (kWh/m^3), V is voltage (Volt), I is current (Ampere), t is Ec time (s) and v is volume of the treated wastewater (m^3). It is also seen that there is a direct relationship between current density for both energy and electrode consumption. They increase with increasing current densities (Fig. 5). Coagulant Al^{3+} is produced by electrochemically sacrificing the aluminum anode. Al^{3+} dosage is determined by charge loading. According to Faraday's law, electrode material consumption and charge loading are calculated in the following equations:

$$\text{Faraday/m}^3 = (I \text{ x } t) / (F \text{ x } v) \quad (2)$$

$$\text{Electrode}_{\text{consumption}} = (I \text{ x } t \text{ x } M_w) / (z \text{ x } F \text{ x } v) \quad (3)$$

...where F is Faraday's constant (96,485 C/mol), M_w is the molar mass of aluminum (26.98 g/mol), and z is the number of electron transfer (z_{Al}:3). When current density was increased from 10 to 150 A/m^2, Energy consumption increased from 0.1 to 18.5 kWh/m^3 and electrode consumption increased from 0.01 to 0.31 kgAl/m^3. Charge loading and electrolysis voltages effect energy consumption. Besides, high Charge loading increases energy and electrode consumption. EC requires the charge loading, which depends on both the influent wastewater property and desirable effluent water quality.

Electrode consumption effects depend on wastewater characteristics and operating conditions. In fact, total electrode material consumption can be greater than the theoretical value. Consumption of the four electrode materials was 0.15 kg in the EC process at 1.62 A. According to Faraday's law, theoretical consumption electrode was 0.09 kg of aluminum at the same current. It is clear that electrode consumption, besides electrochemical and chemical reactions, contributes significantly to the dissolution of aluminum electrodes to generate coagulant Al^{3+} in the EC [31].

Fig. 6 was presented as the operating cost changed with the increased current density. When the current density was changed 10-150 A/m^2, operating costs were determined as 0.03-1.67 $/m^3.

Effect of EC Time

The EC time is another significant parameter that is influential on the electrocoagulation process. Because the formation and concentrations of metal hydroxides play an important role on pollutant (COD, turbidity and phosphorus) removal, this depends on operation time. The effect of EC time on treatment efficiency was carried out by varying the charge loading from 7 to 54 Faradays/m^3. In the EC

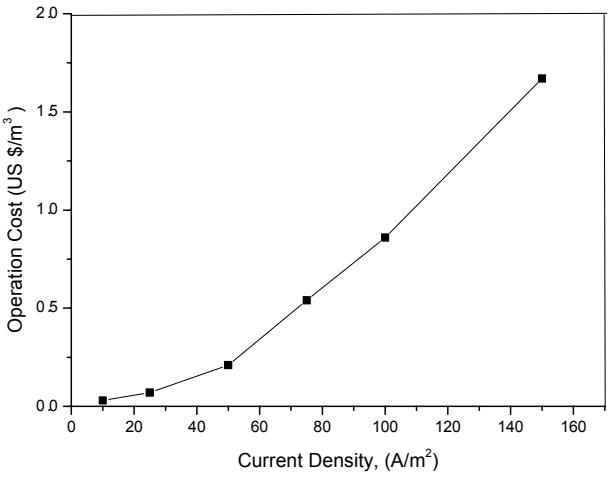

Fig. 6. Effect of current density on operating cost (pH 7.8 and EC time 10 min).

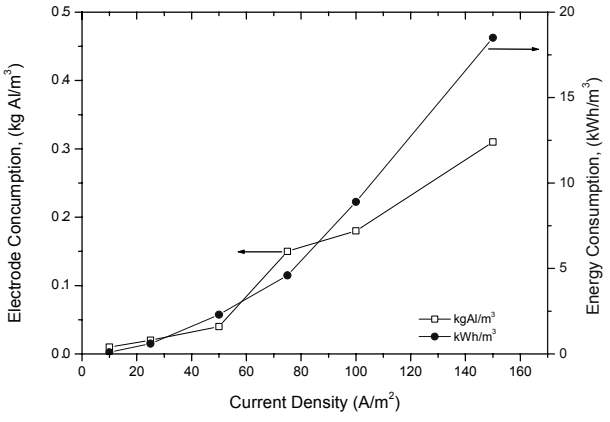

Fig. 5. Effect of current density on electrode and energy consumption (pH 7.8 and EC time 10 min).

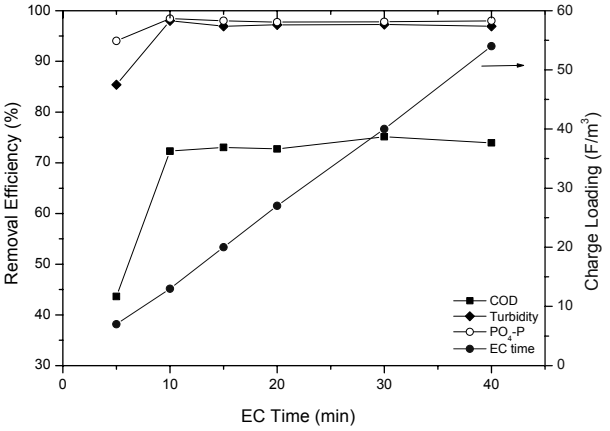

Fig. 7. Effect of EC time on COD, turbidity, and phosphate removal (pH 7.8 and current density 100 A/m^2).

process, anode produces metal ions during electrochemical reaction. Metal ions are a destabilization agent. If the charge loading were low, the metal ion dosage was not sufficient to destabilize all colloidal and suspended particles, so pollutant removal was not high. Fig. 7 shows that the EC time has an important effect on pollutant removal efficiency. When EC time was changed from 4 to 40 min (similarly, charge loading was changed from 7 to 54 Faradays/m^3), the removal efficiencies of COD from 44 to 75%, turbidity from 85 to 97%, and phosphorus from 94 from 98% were obtained.

Fig. 8 showed that the EC time affected consumptions of energy and electrode. When the EC time was increased, both the energy and electrode consumption increased during the EC process were shown. The EC time increase from 5 to 40 min causes an increase in energy consumption from 4.5 to 40.5 kWh/m^3, and an increase in electrode consumption from 0.09 to 0.79 kgAl/m^3. It is clear that EC time is an important parameter for the EC process because it affects the economic applicability in the treatment of the DWW.

Fig. 9 was presented as operating cost that changed with the increased EC time. When EC time was changed to 5-40 min, operating costs were determined as 0.44-3.85 \$/m^3.

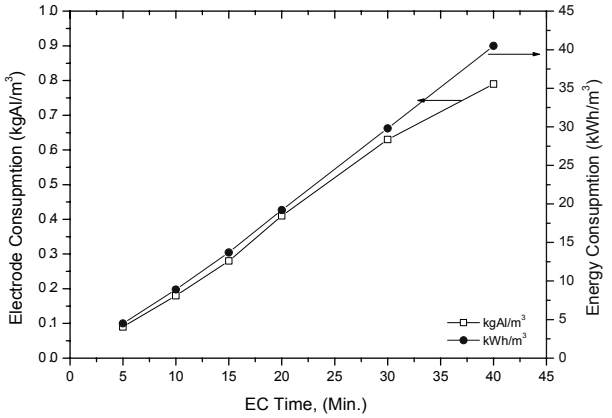

Fig. 8. Effect of EC time on electrode and energy consumption (pH 7.8 and current density 100 A/m^2).

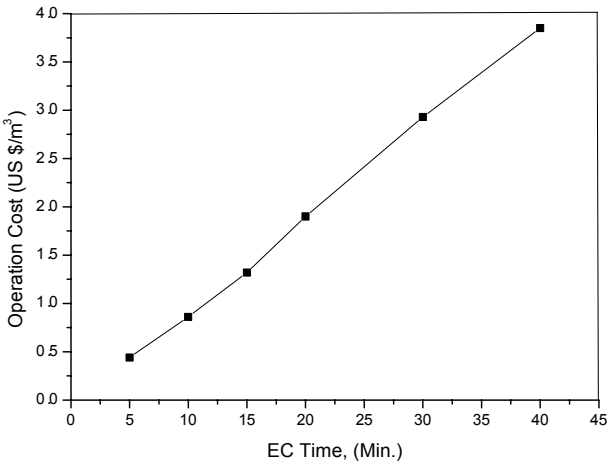

Fig. 9. Effect of EC time on operating cost (pH 7.8 and current density 100 A/m^2).

Conclusions

In this study, the EC process was found to be an effective method for the treatment of DWW. The effects of operational conditions such as electrode type, initial pH, current density, and EC time on performance were examined. Besides, operating cost of the EC process was evaluated. DWW was successfully treated by EC process in a very short time (5-10 min). The removal of efficiencies of COD, turbidity, and phosphorus were high at 75%, 98%, and 98%, respectively. The optimum operating conditions were pH 7.8, current density 100 A/m^2, and EC time 10 min. The power requirement and electrode consumption were 8.9 kWh/m^3, and 0.18 kgAl/m^3 wastewater treated under the optimum operating conditions. In addition, operating costs were determinated with respect to different values of current density and EC time. Operating cost as 0.86 \$/m^3 under the optimum operating conditions was evaluated. According to our results, the EC process in the optimum conditions was shown to be economical treatment of DWW.

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