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Electrocoagulation in a Plugflow Reactor: The Treatment of Cattle Abattoir Wastewater by Iron Rod Anodes

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ABSTRACT: The performance of a plug flow electrocoagulation reactor for the treatment of cattle abattoir (slaughterhouse) wastewater was investigated. A cylindrical iron reactor was operated as a cathode at a recycling batch mode while three iron rods located at the center of the reactor were used as an anode. The effects of different parameters, including the current density, the supporting electrolyte dosage (Na_2SO_4), the wastewater recirculation rate, the initial pH and the polyelectrolyte addition were also evaluated. In order to achieve a high removal efficiency, the Electro-Fenton process was also evaluated by adding H_2O_2 of various concentrations. When the abattoir wastewater was subjected to the Electro-Fenton process, the best result regarding the removal of COD was obtained as 90% with an electrical energy consumption of 0.2 kWh/L. The experimental results reveal that the direct dischargeable effluent, according to Turkish legislation, was obtained in this study. In conclusion, the electrochemical reactor designed for this purpose is effective for the removal of COD from wastewater.

Key words: Electrocoagulation, Iron, Slaughterhouse wastewater, Electro-Fenton, COD

INTRODUCTION

An abattoir is a facility where animals are killed for the production of food products and, as a result, a large amount of wastewater is generated from the slaughtering step, the washing of the equipment and the production of by-products (skins, fats, and bones). Various amounts of wastewater are produced, from 1 to 8.3m^3 per slaughtered animal, and most of this amount is discarded as wastewater (Saddoud and Sayadi 2007). Because of its high organic content, this wastewater causes important environmental problems.

In the treatment of abattoir wastewater, aerobic (Li *et al.*, 2008; Al-Mutairi 2009) and anaerobic (Saddoud and Sayadi 2007; Cao and Mehrvar 2011; Debik and Coskun 2009) processes are mainly used for the removal of organic pollution. Due to limited oxygen transfer and high energy requirement in aeration and generation of high amounts of sludge, aerobic processes may not be convenient for the treatment of the wastewater. Also, anaerobic processes may not be suitable because of the accumulation of organic acids in the intermediate steps resulting in process failure, especially under high loading conditions.

Electrocoagulation (EC) is an alternative technology for wastewater treatment systems and is very effective in removing inorganic and organic contaminants and also pathogens. Electrocoagulation has certain advantages over chemical coagulation, such as requiring only basic equipment, easy operation and automation, a short retention time, low sludge production and usually requiring no chemicals (Merzouk *et al.*, 2011; Harif *et al.*, 2012). Electrocoagulation is the in situ generation of M^{3+} ions into wastewater using sacrificial anodes while the hydrogen gas and $(OH)^-$ ions are produced at the cathode. A further reaction between $M^{3+}_{(aq)}$ and OH^- ions produce amorphous $M(OH)_3$ flocs which are capable of adsorbing soluble organic compounds and trapping colloidal particles. The further polymerization of these flocs to $M_n(OH)_{3n}$ allows for easy settling of the flocs from the wastewater by sedimentation or/ and flotation by hydrogen gas.

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The electrocoagulation (EC) of abattoir wastewater has been studied by several researchers. The electrocoagulation of abattoir wastewater in an acrylic reactor was tested using eight pieces of mild steel and aluminium electrodes arranged in a bipolar (BP) or monopolar configuration system by Asselin et al. (2008). The monopolar configuration system, with four parallel aluminum and iron electrodes in the plexiglass reactor, was also used by Kobya et al. (2006) and Bayramoglu et al. (2006) for the EC of poultry abbattoir wastewater. The EC of poultry abattoir wastewater, using ten monopolar aluminum electrodes in a plexiglass reactor, was studied by Bayar et al. (2011). Eight aluminum and iron electrodes were used for the electrolysis of fowl-abattoir wastewater to minimize odors and organic matter by Marconato et al. (1998). Among these studies, only Tezcan Un et al. (2009) used a dissimilar design, which consisted of cylindrical iron and aluminum anodes and stirrer cathodes with two blades for the EC of the cattle abattoir wastewater. Although the electrocoagulation of abattoir wastewater has been utilized by various researchers as mentioned above, they have mainly focused on two parameters: (1) the electrode materials and (2) the configuration of the parallel plate electrodes. Electrocoagulation studies up to date have failed to examine the usage of monotype reactors.

In order to meet this shortfall in the current literature three iron rod anodes and a cylindrical iron cathode was used for the EC of cattle abattoir wastewater. The purpose of the work was to investigate the performance of this reactor designed differently from those in the literature to obtain direct dischargeable effluent. Experiments were conducted to examine the effects of the operating parameters, such as the current density, the supporting electrolyte concentration, the initial pH, the wastewater recirculation rate, the hydrogen peroxide (H_2O_2) concentration, the wastewater flow regime and the addition of coagulant-aid on COD removal efficiency. Energy consumption was also analyzed.

MATERIALS & METHODS

The wastewater used in this study was provided from a cattle abattoir plant located in the city of Eskisehir, Turkey. The wastewater was mixed with washing water at the plant and samples were taken just before admission to the conventional treatment facility. Although the quality of the effluent was variable, it was highly colored and typically had a pH of 7.1 and a COD of 840 mg/L.

The treatment of the cattle abattoir wastewater by electrocoagulation was performed in a recycling batch mode operation. The plug flow electrochemical reactor was made of iron with a height of 75 cm and a diameter of 3.5 cm, and was operated as a cathode. Three iron rods (outside diameter = 1.1 cm; height=72cm) were used as anodes and located in the center of the cathode triangularly as seen in Fig.1. The anode and cathode sets were respectively connected to the positive and negative outlets of a DC power source. In each run, 800 mL of wastewater was fed into the bottom of the reactor from a reservoir via a peristaltic pump and was continuously recycled. Prior to each test, the pH was adjusted to a desired value using 1N H_2SO_4 or 1N NaOH. A specific amount of supporting electrolyte (Na_2SO_4) was added to the wastewater to increase the conductivity. A sample was taken to determine the initial COD concentration and then current was applied to the circuit by a power supply (Statron T-25) for a the period of 90 minutes. The current was held constant for each run. The pH and the conductivity of the

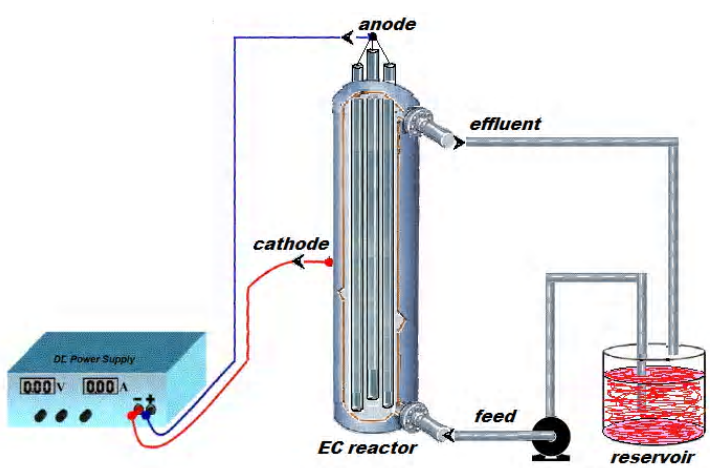

Fig. 1. Experimental setup

solution were not controlled, but they were monitored during the experiments using a pH-meter (Hanna Ins. 301) and a conductivity meter (Radiometer Pioneer 30). Samples were withdrawn at periodic time intervals, centrifuged and were analyzed to determine the concentration of COD by the closed reflux method. All the analyses were repeated twice. The electrodes were polished, washed with dilute H_2SO_4 and rinsed with distilled water before each run.

RESULTS & DISCUSSIONS

The effect of the current density was investigated applying 10, 20, 30 and 40 mA/cm^2 to the anode at a pH of 7.1 and a wastewater recirculation rate of 600mL/min. COD removal efficiencies of 68.0, 72.0, 76.0 and 81.0% were obtained at the current densities of 10, 20, 30 and 40 mA/cm^2, respectively as seen in Fig. 2-a. The COD removal efficiency increased significantly with an increase of current density due to an increase in the amount of iron ions dissolved in the wastewater and in the formation rate of $M(OH)_3$. The initial COD concentration of 840 mg/L was reduced to 160 mg/L at 40mA/cm^2 after 90 min electrocoagulation. According to Turkish legislation the legal discharge limit to the receiving body for the slaughtering industry is 250 mg/L for a composite sample of two hours. As a result of the experiments, COD concentrations under 250 mg/L were obtained after 90 minutes EC at 10 and 20 mA/cm^2, after 75 minutes EC at 30 mA/cm^2 and after 60 minutes EC at 40 mA/cm^2. Consequently, the direct dischargeable effluent was obtained by reducing the initial COD concentrations under 250 mg/L at all the current densities applied. Increasing the current density reduces the time required to treat a specific quantity of effluent to obtain the desired quality.

Although, higher current density favors COD removal efficiency, it impacts the cell voltage by means of various over potentials which results in an increase in energy consumption. Electrical energy of 0.127, 0.463, 0.933 and 1.732 kWh/L were consumed after 90 minutes EC at current densities of 10, 20, 30 and 40 mA/cm^2, respectively. As seen from Fig. 2-b the electrical energy consumption increased more rapidly with time as the current density increased. As acceptable wastewater quality, according to the legislation, was obtained in less than 90 minutes at 30 and 40 mA/cm^2, the energy consumptions will be reduced to 0.748 (75 min EC time) and 1.001 kWh/L (60 min EC time) at 30 and 40 mA/cm^2, respectively.

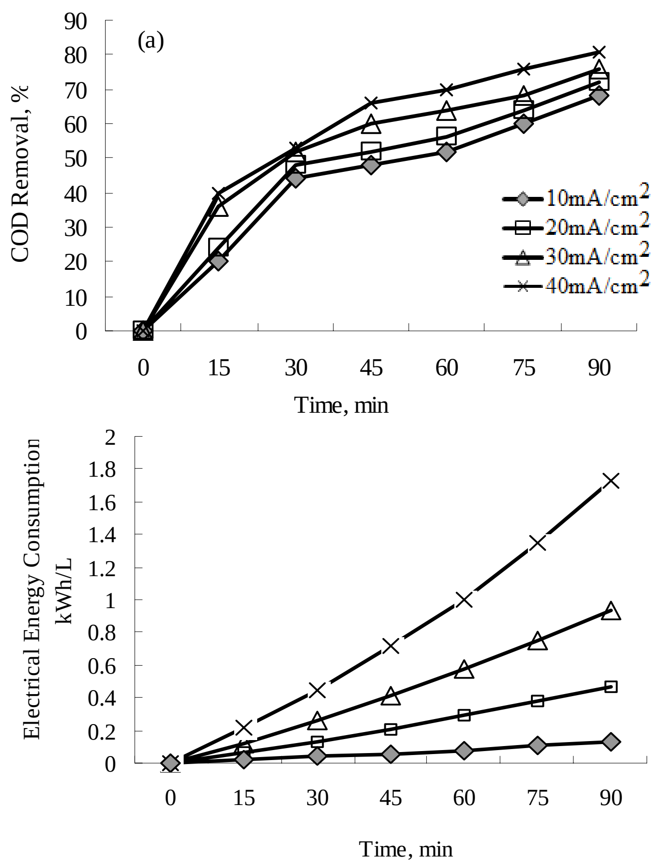

Fig. 2. Effect of the current density on COD removal efficiency (a), and on electrical energy consumption (b). $v_{circulate}$: 600mL/min, pH: 7.1

Using supporting electrolyte is the most common method to overcome high ohmic resistance between the anode and the cathode which results in lower cell voltage, and also to save electrical energy consumption. In this study Na_2SO_4 was used as the supporting electrolyte to obtain higher electrical conductivity, thus reducing electrical energy consumption. To clarify the effect of supporting electrolyte concentration, experiments were performed at a current density of 40 mA/cm^2, a wastewater recirculation rate of 600mL/min and a pH of 7.1. Fig.3 illustrates the results in terms of (a) COD removal efficiency and (b) electrical energy consumption at various Na_2SO_4 concentrations ranging from 0.05 to 0.2 M. As shown in Fig. 3(a), the rates of removal decreased with an increase in Na_2SO_4 concentration. The removal efficiency of the COD was 72, 66 and 54% with 0.05, 0.1 and 0.2 M Na_2SO_4 dosages, respectively, after 90 min electrocoagulation. This adverse effect has been explained with the passivation of the electrode by Lakshmipathiraj et al. (2010). Anodic passivity occurs with the competition of the sulphate ions with the hydroxyl ions for adsorption, and thus the active surface area of the electrode reduces thereby limiting the dissolution of the iron electrode. Similar behavior of the Na_2SO_4 was observed by Lakshmipathiraj et al. (2008), Mansouri et al.(2011) and Tezcan Un et al. (2009).

Fig. 3(b) shows the influence of the supporting electrolyte on electrical energy consumption. It can be concluded from this data that in the presence of 0.2 M Na_2SO_4, electrical energy of 0.348 kWh/L was consumed, whereas about a five fold amount of electrical energy was required (1.7323 kWh/L) in the electrocoagulation without Na_2SO_4. Although the addition of Na_2SO_4 is beneficial for the electrical energy requirements, it affects the electrocoagulation efficiency adversely. Therefore, 0.1 M Na_2SO_4 concentration should be preferred to avoid the adverse effects of an excess supporting electrolyte concentration as well as to avoid the additional cost of Na_2SO_4 while it reduces electrical energy consumption. A typical electrolysis reaction involves the transfer of charge between an electrode and a species in solution. The entire process, due to the interfacial nature of the electron transfer reactions, constitutes a series of steps. Reactants are transported to the electrode surface, electrochemical transformation occurs and the product is formed and transported from the electrode to perform coagulation in the bulk. The reaction can be controlled by two factors: (1) the rate constant and (2) the surface concentration of the reactant. With increasing polarization of the electrode, as current density increases, the electrode can become starved of reactant and the rate of mass transport governs, and limits, the overall reaction. Thus, movement of the reactant in and out of the interface so-called mass transport, is important. The transfer of a solute through a liquid can be affected by molecular diffusion under the influence of a concentration gradient, by migration of a charged species under the influence of a potential or an electrical field and by convection due to the bulk motion of the medium. In this study, we assume that the electrolysis reaction is not significantly affected by migration because of the addition of a sufficient quantity of the electrolyte and because mass transport due to forced convection in the bulk is considered to be faster compared to molecular diffusion because the wastewater is continuously recirculated through the reactor. To determine the effect of the wastewater recirculation rate, wastewater flow rates of 240, 600, 1000 and 1400 mL/min were examined under conditions

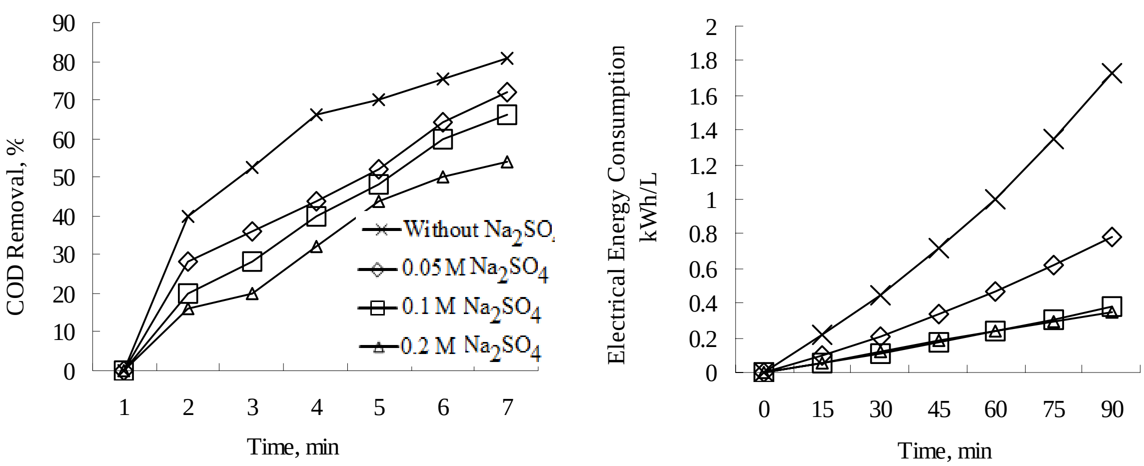

Fig. 3. The effect of supporting electrolyte (a) on COD concentrations and (b) on electrical energy consumption. $v_{circulate}$: 600mL/min, pH 7.1, Current Density: 40 mA/cm^2

of pH:7.1, a current density of 40mA/cm^2 and the presence of 0.1M Na_2SO_4. As shown in Fig. 4, as the flow rate increased, the COD removal efficiency also increased. The efficiencies of the COD removal were 64, 66, 68 and 70% at 240, 600, 1000 and 1400 mL/min, respectively. This effect resulted from an increased amount of contact at the interface with an increase in forced convection rate of the solution. Furthermore, the corrosion of iron increased because of the number of pores for electrogeneration increased. Therefore, high removal efficiency was obtained with the production of a sufficiently high amount of coagulated iron. On the other hand, a very high wastewater recirculation rate should not be applied, to support the flocculation of coagulated iron and to avoid breaking the flocs into smaller pieces.

The effect of the pH on the electrocoagulation process is important because the type of iron hydroxide species formed in the solution and the surface charge of the particle depend on the pH. The effects of the initial pH on COD removal from the wastewater using iron rods electrodes by electrocoagulation was investigated using a current density of 40mA/cm^2, a wastewater recirculation rate of 600mL/min and the presence of 0.1MNa_2SO_4. The removal efficiency of COD versus time in different initial pH's is shown in Fig. 5. The efficiencies of COD removal were 28, 40, 66 and 52% at pH 3, 5, 7 and 9 at the end of 90 min of electrocoagulation, respectively. This clearly indicates that removal efficiency increased with pH up to 7 and then decreased with a further increase in pH to 9. At low and high pH's, soluble iron hydroxide species are formed according to the Pourbaix diagram of iron. There was minor removal at an acidic pH because of collapsing hydroxide ions generated at the cathode by protons which caused an insufficient formation of iron hydroxide. At a high pH, the removal efficiency decreased because of the dominant formation of soluble $Fe(OH)^-_4$ which is not suitable for the formation of floc. At a neutral pH, the majority of iron complexes were formed and a higher removal efficiency achieved. Therefore, the addition of chemicals to adjust the initial pH was not necessary because the initial pH of the abattoir wastewater was 7.1 being within a neutral range. This behavior has been observed by many investigators(Kumar *et al.*,2009;Daneshvar *et al.*, 2007). Fenton's reaction is a reaction in which hydrogen peroxide catalysis by iron producing a strong catalytic power to generate highly reactive hydroxyl radicals ($OH^·$). The reaction between iron and hydrogen peroxide generates certain hydroxyl radicals, which are powerful non-selective oxidants, according to Eq.1-5 (Nidheesh and Gandhimathi 2012).

$$Fe^{2+} + H_2O_2 \longrightarrow Fe^{3+} + OH^· + OH^- \quad (1)$$
$$Fe^{3+} + H_2O_2 \longrightarrow Fe^{2+} + OOH^· + H^+ \quad (2)$$
$$RH + HO\cdot \longrightarrow R\cdot + H_2O \quad (3)$$

where, RH denotes organic pollutants

$$R\cdot + Fe^{3+} \longrightarrow R^+ + Fe^{2+} \quad (4)$$
$$Fe^{2+} + HO\cdot \longrightarrow Fe^{3+} + OH^- \quad (5)$$

These reactions are very practical due to their high organic removal efficiency without the need for special equipment. Recently, the Electo-Fenton (EF) process, a modification of the traditional Fenton oxidation process, has been widely-used for the removal of various organic compounds such as phenols (Babuponnusami and Muthukumar 2012), pesticides and herbicides (Boye *et al.*, 2002; . Abdessalem *et al.*, 2010), dyes (Rosales *et al.*, 2012) and so on. In this part of the study, the Electro-Fenton process was carried out at pH 3 using a reaction between electrogenerated Fe^{2+} ions and H_2O_2 which was added externally to the reactor. In the EF process, the pH of the solution is very important. A pH around 3 has been described as the optimum pH and it has been found that efficiency decreases by increasing the pH for the

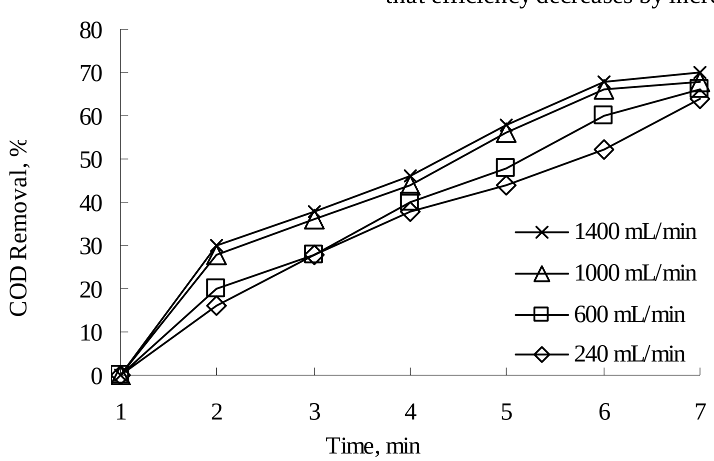

Fig. 4. Effect of the flow rate on COD concentrations: Current density: 40 mA/cm^2, pH:7.1, 0.1M Na_2SO_4.

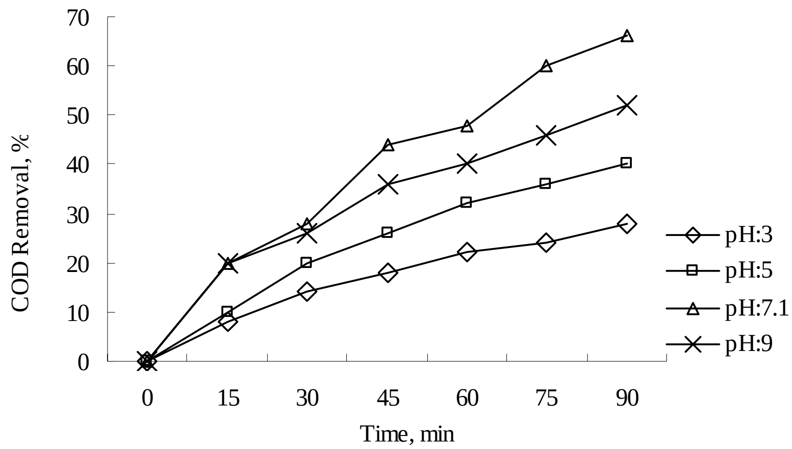

Fig. 5. Effect of the pH on COD removal efficiency. $v_{circulate}$: 600mL/min, 0.1M Na_2SO_4, Current Density: 40 mA/cm^2

EF by several researchers (Rosales *et al.*, 2009, Rosales *et al.*, 2012, Zhao *et al.*, 2012). It was reported that H_2O_2 becomes unstable and decomposes to oxygen and water at a neutral to basic region (Eq. 6,7) (Nidheesh and Gandhimathi 2012). Therefore, by increasing pH during the EF process, the EF process becomes ineffective and electrocoagulation, characterized by electrostatic attraction and complexation using $Fe(OH)_3$, will be the dominant mechanism in the solution.

$$H_2O_2 + 2H^+ + 2e^- \longrightarrow 2H_2O \quad (6)$$

$$H_2O_2 + H^+ \longrightarrow H_3O_2^+ \quad (7)$$

To perform the EF process, various concentrations of H_2O_2 were added to the reactor since the concentration of the H_2O_2 plays an important role in the removal of organics from the wastewater. The results obtained from experiments at pH 3 and a current density of 40 mA/cm^2 can be seen in Fig.6. The removal of pollutants increased with an increase in the H_2O_2 concentration. The removal efficiencies were 68, 70 and 72% with the addition of 20mM, 40mM and 80mM H_2O_2. This increase resulted from the increase in the hydroxyl radical concentration as a result of the addition of H_2O_2.

The application of synthetic polyelectrolytes in water and wastewater treatment, accompanied by the metallic coagulants, has been commonly used for many years. A lot of synthetic polyelectrolytes under different trade names are available commercially. The selection of the polyelectrolyte type among cationic, anionic, and nonionic polyelectrolytes depends on both the type of particle being removed and the chemical characteristics of the wastewater. Polyelectrolytes are used to enhance aggregation in the coagulation and flocculation process. It works with two mechanisms: (a) it affects the surface charge on the particle to be neutralized and (2) it provides bridging between the particles which creates a network from them. Consequently, large, fast-settling flocs with a very high molecular weight are formed by bridging between many small primary flocs. In this study, the anionic polyelectrolyte, which is the most suitable flocculant for negatively charged colloids, was added to the wastewater in order to achieve particle instability and to increase the particle size, consequently achieving effective removal of organic substances represented as COD. This was provided by the publicly-owned wastewater treatment plant of the city (Eskisehir) and a dosage of 0.24 g/L was used as suggested by the supplier.

The results obtained from the experiment can be seen in Fig.7. The removal efficiency of 66% was increased to 72% using polyelectrolyte as seen in Fig.7(a) with 0.1 M Na_2SO_4 at pH 7.1, $v_{circulate}$: 600mL/min at a current density of 40 mA/cm^2. The removal efficiency of 72% obtained corresponded to the COD concentration of 235 mg/L in the effluent, which meets the legal requirement of 250 mg/L for the COD discharge by the abattoir industry in Turkey. Additionally, the energy consumption was reduced from 0.38 to 0.29 kWh/L as expected using polyelectrolyte.

To determine the highest performance of the reactor, one more experiment was performed under the best operating conditions. The best conditions were determined at the end of the study as a current density of 40 mA/cm^2, a recirculation rate of 1400 mL/min, a pH of 7.1, a Na_2SO_4 concentration of 0.1 M, a H_2O_2 concentration of 0.08 M and a polyelectrolyte dosage of 0.24 g/L. As shown in Fig.7, the highest removal efficiency of 90% was obtained under these conditions. The initial COD concentration of 840 mg/L was reduced to 84 mg/L after 90 minutes electrocoagulation. Additionally, the electrical energy consumption under

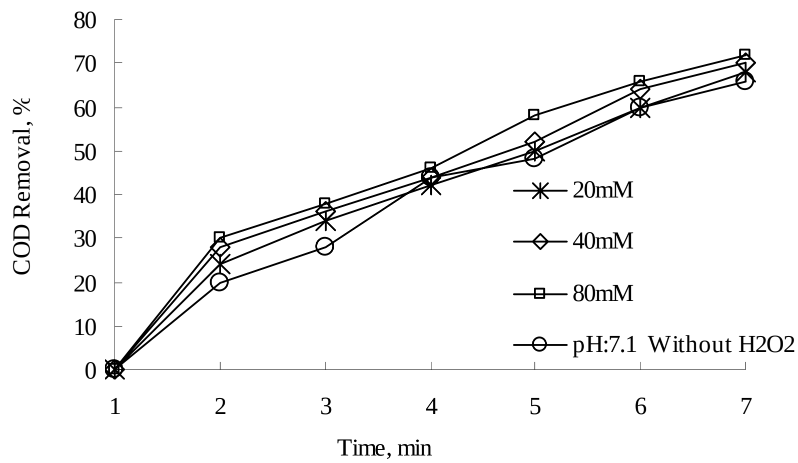

Fig. 6. The effect of H_2O_2 on COD removal efficiency. pH:3, $v_{circulate}$: 600mL/min, 0.1M Na_2SO_4, Current Density: 40 mA/cm^2

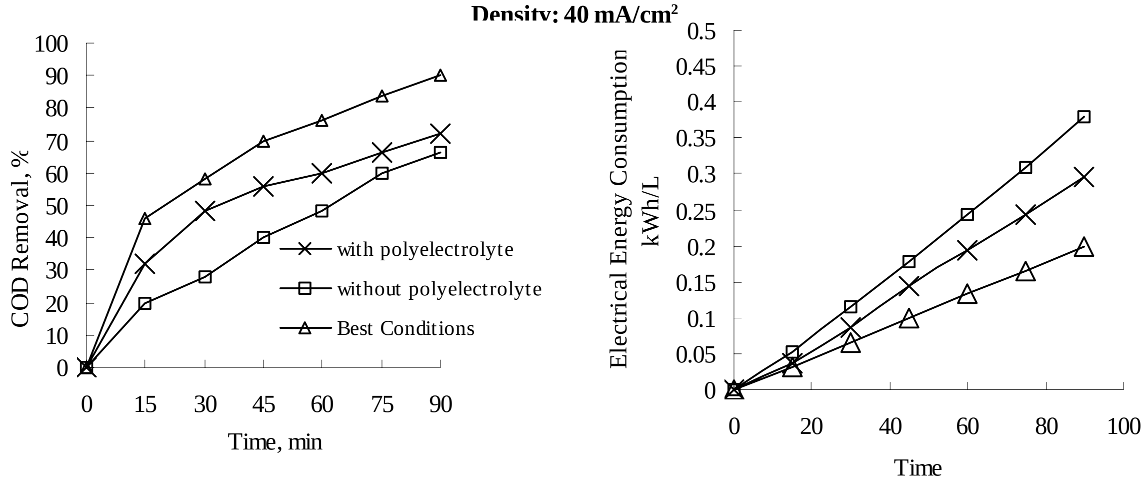

Fig. 7. Effect of the polyelectrolyte and operation under best conditions (a) on COD concentrations and (b) on electrical energy consumption.

these conditions was the smallest obtained in the study at 0.2kWh/L after 90 minutes electrocoagulation. In the industrial application, the expected performance of electrocoagulation systems is limited by legislation. In this study, effluent COD decreased below 250 mg/L within 45 minutes with a reactor operation at the higher wastewater recirculation rate and the presence of polyelectrolyte and H_2O_2. Consequently, according to the legislation, no further EC treatment after that point was necessary. As a result, the electrical energy consumption reduced to 0.1 kWh/L.

CONCLUSION

The performance of a plug flow electrocoagulation reactor operating at a recirculating batch mode for the treatment of cattle abattoir wastewater was investigated. It can be concluded that the treatment of wastewater by EC is effective based on:

- The enhanced COD removal efficiency by increasing current density and wastewater recirculation rate. For a higher performance, the initial pH must be close to a neutral value and the concentration of **Na_2SO_4** used as a supporting electrolyte must be kept to a minimum, although it affects electrical energy consumption positively.
- The pollutant removal by the electro-Fenton process improved when the reactor was operated by adding H_2O_2. Polyelectrolyte addition also helped to remove the COD very efficiently.
- The electro-Fenton process operated under the following conditions: a current density of 40 mA/cm^2, an initial pH of 7.1, a recirculation rate of 1400 mL/min, and the presence of 0.1 M Na_2SO_4, 0.08 M H_2O_2, and 0.24 g/L polyelectrolyte. The best result regarding the removal of COD attained was 90%. The initial concentration of 840 mg/L was reduced to 84 mg/L with an electrical energy consumption of 0.2 kWh/L. Since the allowable COD concentration for abattoirs according to Turkish legislation is 250 mg/L, direct dischargeable effluent was obtained in this study.

Considering the results, it can be said that electrocoagulation with a plug flow reactor and the electro-Fenton process can eventually represent suitable alternative processes for the complete treatment of abattoir wastewater.

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