Treatment of Domestic Wastewater by Electrocoagulation in a Cell with Fe–Fe Electrodes

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ABSTRACT

In this study, the potential to treat domestic wastewater (DWW) using an electrocoagulation (EC) process was tested on two different samples, one of which was heavily loaded [380 mg chemical oxygen demand (COD)/L]; the other was weakly loaded [260 mg COD/L]. The samples were supplied from the Istanbul-Yenikapi DWW pretreatment plant. The experimental studies were conducted within an electrolysis cell with Fe–Fe electrodes. In this cell, under optimum operational conditions in the form of electrical power and electrolysis time, pollutant removal efficiency (as COD), suspended solids (SS) and characteristics of formed sludge were tested. At the end of these studies, the optimum operational conditions found for electrical power was 0.6 W for both DWW samples, and electrolysis times were determined as 15 min for heavily loaded DWW (HLDWW) and 8 min for weakly loaded DWW (WLDWW). Energy consumption in optimum electrical charge conditions was 0.4 kWh/m^3 HLDWW and 0.2 kWh/m^3 WLDWW. The removal efficiency of COD and SS were obtained and shown to be over 60 and 70%, respectively. The level of sludge production was 1.5–2% during the optimal reaction time, and sludge volume was decreased 50% using H_2O_2.

Key words: COD; domestic wastewater; electrocoagulation; electricity consumption; sludge reduction

INTRODUCTION

WASTEWATER CAN CREATE SEVERE WATER POLLUTION problems for aquatic life because of its various contents. Although wastewaters can be composed of various sources, it is most commonly composed of domestic wastewaters (DWWs). Currently, DWW is generally treated by aerated biological methods. Activated sludge, being the most popular method, produces high quality effluent—90% biological oxygen demand (BOD) and suspended solids (SS) removal (Metcalf and Eddy, 2003). But this commonly used method has some disadvantages, such as requiring continuous air supply, high op-

erating costs (skilled labor, energy, etc.), sensitivity against shock toxic loads, longer treatment time, and necessary sludge disposal. Some lab-based studies have been carried out to treat DWW chemically by coagulation. Chemical oxygen demand (COD) treatment between the ratios of 55 and 75%, especially if it includes SS substances in the wastewater, can be realized by this method (Debik, 1999). The chemicals that are consumed in this type of treatment and the chemical characteristics of the sludge produced are the most significant problems encountered using this method.

From an environmental point of view, the sewage treatment process is still far from being environmentally sus-

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tainable. Based on a desk study, a structural approach is given on how to achieve a more sustainable treatment process. Some of the possibilities are the improvement of the mitigation of toxic pollutants, high-temperature sludge treatment processes, and membrane separation processes (Rulkens, 2006). Electrochemical treatment methods can be mentioned in this context also.

Electrochemical treatment seems to be a promising treatment method due to its high effectiveness, its lower maintenance cost, less need for labor and rapid achievement of results (Feng *et al.*, 2003). Electrocoagulation-flotation treatment is investigated, as it has greater ability for the removal of COD and SS from effluents in comparison with treatment by conventional coagulation (Jiang *et al.*, 2002).

At the end of the studies identifying the most appropriate treatment alternatives, electrocoagulation (EC) is shown to be one of the best methods for the wastewater. Reusing of wastewater has become an absolute necessity, so there is an urgent need to develop inventive, more effective, and low-cost techniques for treatment of wastewater (Feng *et al.*, 2003). Electrochemical treatment techniques are one of these. A host of very promising techniques based on electrochemical technology are being developed, and existing ones are being improved that do not require chemical additions (Mollah *et al.*, 2001). EC can be used to remove irons, silicates, humus, dissolved oxygen (Chen, 2004), reduce copper (Comninellis and Pulgarin, 1993) and decolorize (Bechtold *et al.*, 2002). EC has also been applied successfully to treat potable water, food, and protein wastewater, yeast wastewater, urban wastewater, restaurant wastewater, tar sand, and oil shale wastewater, nitrate containing wastewater, heavy metals, textile dyes, fluorine, polymeric wastes, organic matter from landfill leachate, suspended particles, chemical and mechanical polishing wastes, aqueous suspensions of ultrafine particles, and phenolic waste (Mollah *et al.*, 2004). Presently, EC technologies are more efficient and more compact. Removal mechanisms of the EC process include coagulation, adsorption, precipitation, and flotation (Kobya *et al.*, 2003). Similarly, the effect of DC electric fields on COD in aerobic mixed sludge processes has been investigated, and optimum operational conditions were determined (Alshawabkeh *et al.*, 2004). In another study, removal of nitrogen compounds has been successfully realized using a rotating electrobiological contactor; an interesting study, in which over 83% efficiency of the denitrification was observed at a nitrification efficiency of 68.9 % (Krzemieniewski and Rodzievicz, 2005). When the literature is scanned for studies of treatment by electrocoagulation of DWW, it is almost an untouched area. That is to say, there is a research requirement on the matter.

In the EC process, coagulation is generated *in situ* by electrolytic oxidation of an appropriate anode material. During this process, charged ionic species are removed from wastewater by allowing it to react with an ion having opposite charge, or with flock metallic hydroxides generated within the effluent.
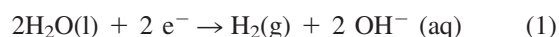
EC treatment methods offer an alternative to the use of metal salts or polymers and polyelectrolyte addition for breaking stable emulsions and suspensions. The EC treatment method gives highly charged polymeric metal hydroxide to aqueous media, so the electrostatic charges on suspended solids and oil droplets are neutralized to facilitate coagulation and their resultant separation from the aqueous phase. The treatment prompts the precipitation of certain metals and salts.

Treatment performances of the system are studied via optimization of electrical current and of electrode types, which are the two most important parameters for the method (Chen *et al.*, 2002; Chen, 2004).
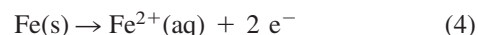
For the EC treatment method, soluble electrodes (such as Fe or Al) must be selected as anode material. In this research, the use of iron sheets as anodes and cathodes in the treatment of DWW as an electrochemical treatment method is explored.

Iron in an electrolytic system produces ferrous and ferric hydroxide upon oxidation. When iron electrodes are used, the reactions occurring during the electrochemical treatment process are as follows:
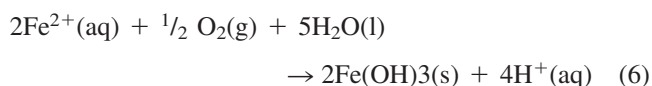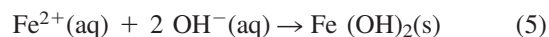
Rxns in a cathode environment,

$$2H_2O(l) + 2\ e^- \rightarrow H_2(g) + 2\ OH^-\ (aq) \qquad (1)$$

$$8H^+(aq) + 8\ e^- \rightarrow 4H2(g) \qquad (2)$$

Rxns in an anode environment,

$$2H_2O(l) \rightarrow O_2(g) + 4H^+(aq) + 4e^- \qquad (3)$$

$$Fe(s) \rightarrow Fe^{2+}(aq) + 2\ e^- \qquad (4)$$

Due to OH^- ion concentrations increasing near the cathode, the pH of the medium begins rising. Meanwhile, the anode melts and dissolves ferrous ions into solution as shown in the following equation:

$$Fe^{2+}(aq) + 2\ OH^-(aq) \rightarrow Fe\ (OH)_2(s) \qquad (5)$$

$$2Fe^{2+}(aq) + {}^1\!/_2\ O_2(g) + 5H_2O(l)$$
$$\rightarrow 2Fe(OH)3(s) + 4H^+(aq) \quad (6)$$

Table 1. Characteristics of the samples (after grit chamber).

Parameters	Unit	*1st Sample*	*2nd Sample*
pH	—	7.2	7.4
Conductivity	mS	1.68	1.91
COD	mg/L	260	380
Suspended solids	mg/L	95	120
BOD$_5$	mg/L	145	210
Chloride	mg/L	295	330

BOD, biological oxygen demand; COD, chemical oxygen demand.

Equation (1) is a cathodic reaction occurring during the electrolysis of water. As hydroxyl ions (OH^-) are released during this reaction, the pH increases. In addition, the observation of vigorous bubbling on the cathode indicates hydrogen production, according to Equation (1) and (2).

Our study showed that if an iron plate is used as anode material, oxidation gasses, such as oxygen, carbon dioxide, and chlorine are not bubbled on the surface of the anodic electrode. Instead, there is formation of green flocks in the aquatic media, indicating its conversion to $Fe(OH)_2$. These results show that Equation (4) forms despite Equation (3) at the anode area.

After melting of the iron in the presence of hydroxyl alkalinity, $Fe(OH)_2$ and $Fe(OH)_3$ flocks develop in the medium [given with Equation 5)]. Oxidation developing over time affects the formation of $Fe(OH)_3$ flocks [given with Equation (6)]. Besides, if oxidant forms such as H_2O_2 add into the electrolysis cell, Fe^{2+} ions convert to Fe^{3+} ions [Equation (7)], and following brownish $Fe(OH)_3$ flocks form [Equation (8)]. Hydroxyl radical in Equation (7) does not put forward an apparent Fenton's oxidation due to dominating high pH.
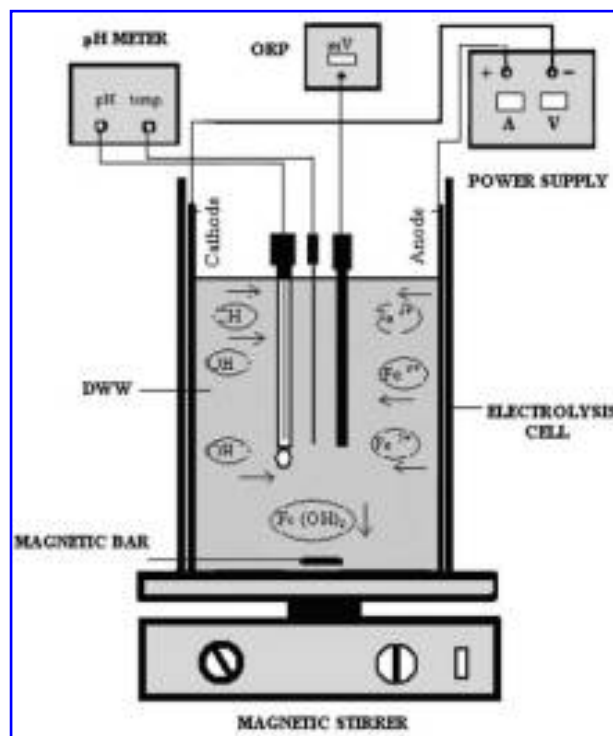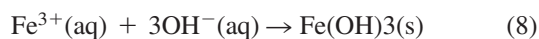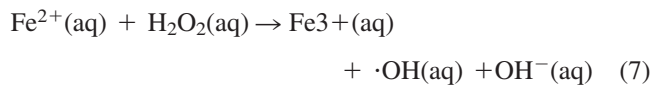
$$Fe^{2+}(aq) + H_2O_2(aq) \rightarrow Fe3+(aq)$$
$$+ \cdot OH(aq) + OH^-(aq) \quad (7)$$

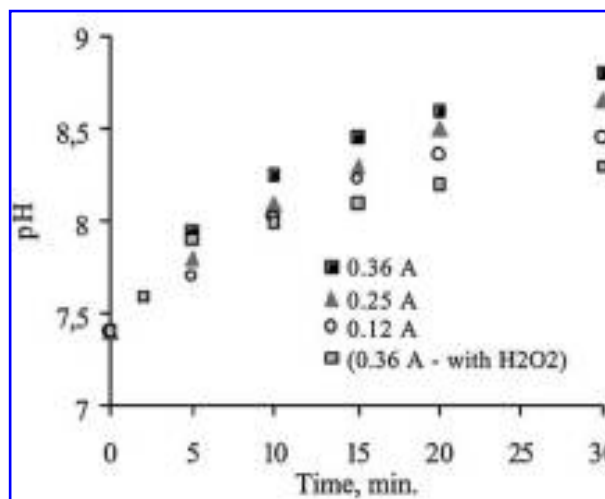$$Fe^{3+}(aq) + 3OH^-(aq) \rightarrow Fe(OH)3(s) \quad (8)$$

Figure 2. pH variation during experimental runs.

Sweeping of the flocks removes particles and dissolved pollutants that are adsorbed onto them (Mollah *et al*., 2001). High treatment efficiency is achieved for the wastewater to which EC treatment is applied, as a result of the above reactions. Therefore, this advanced treatment method has recently become widely preferred.

In this research, the treatability of DWWs by the EC method was tested, and the energy requirements were determined.

MATERIALS AND METHODS

The samples

Two different DWW samples were taken from the outlet of the grit chamber (where particles larger than 1 mm diameter can settle) of the Istanbul-Yenikapi DWW Pretreatment Plant, which provides services to 3 million inhabitants in a 10,000 ha area located on the European Side of Istanbul. The characteristics of DWWs were given in Table 1.

Experimental setup

The studies were exercised in a lab-scale batch system, which was composed of an electrolysis cell, a power supply, a magnetic stirrer, a pH meter, and an ORP meter. An electrolysis cell was made of plexiglas (W 60 mm × L 64 mm × H 160 mm) and contained inside one iron anode and one iron cathode with a separation distance of 60 mm. Sizes of the electrodes were 2 mm (thick) × 60 mm (width) × 120 mm (as wet height). Wet area of the electrodes was 72 cm^2.

Apparatus

- *A digital power supply* named GW Instek (GPS 3030 DD type) having 0–30 V and 0–3 A range.
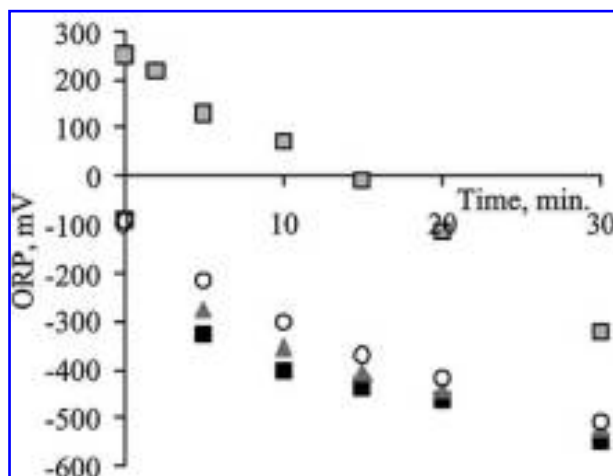

Figure 1. Experimental setup.

Figure 3. ORP variation during experimental runs.

- A Jenway 3040 type pH meter was used for pH measurements.
- A WTW ORP meter with platinum electrodes that determines ORP as mV.

The experimental runs were conducted in the set up illustrated in Fig. 1.

Experimental method

Performance of the EC process was tested on two DWW samples, relatively loaded (380 mg COD/L) and weakly loaded (260 mg COD/L) with organic pollutions, using three electrical current conditions (0.36 A–15 V, 0.25 A–10 V, 0.12 A–5 V, and 0.36 A–10 V, for pH: 3.5) and different treatment times.

Experimental steps were applied as sampling, transportation, preserving, and analysis. For the analysis methods, Merck analytical quality chemicals were used in the preparation of reagents, and all experiments were performed in accordance with Standard Methods (APHA, 2005).

Each experimental application was carried out with 400 mL of sample at room temperature.

In the beginning, the experiments commenced for the unadjusted raw sample (pH = 7.4). After electrolysis, settling ensued for 1 h, then the samples taken from the limpid phase and were analyzed for COD (as total), SS, turbidity, pH, and ORP. Meanwhile, the present sludge percentage was recorded.

Consequently, the performance of the EC process on DWW samples was analyzed based on current intensity and electrolysis time. In addition, electrical consumptions were calculated to evaluate viability of this process.

In a parallel work, to reduce formed sludge volume, EC runs with 0.36 A are repeated using 200 mg H_2O_2/L. This concentration is sufficient to convert Fe^{2+} to Fe^{3+} for 20 min of reaction time according to Faraday's law. To determine the contribution on COD, SS, and turbidity removals of the oxidant addition, an additional search was realized.

EXPERIMENTAL RESULTS AND EVALUATION

pH and ORP changes

During the electrochemical processes, determination of effective reactions [from Equations (1)–(8)] at the anode and cathode electrodes is essential. pH and ORP measurements are very important in showing the type of dominate reactions, such as reduction or oxidation. The experimental findings related to the pH of the solution are illustrated in Fig.
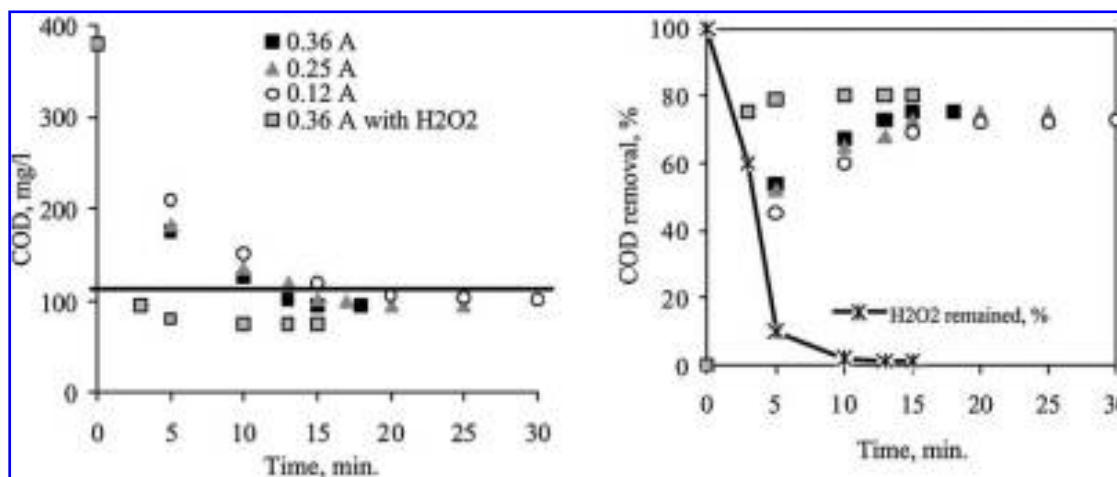

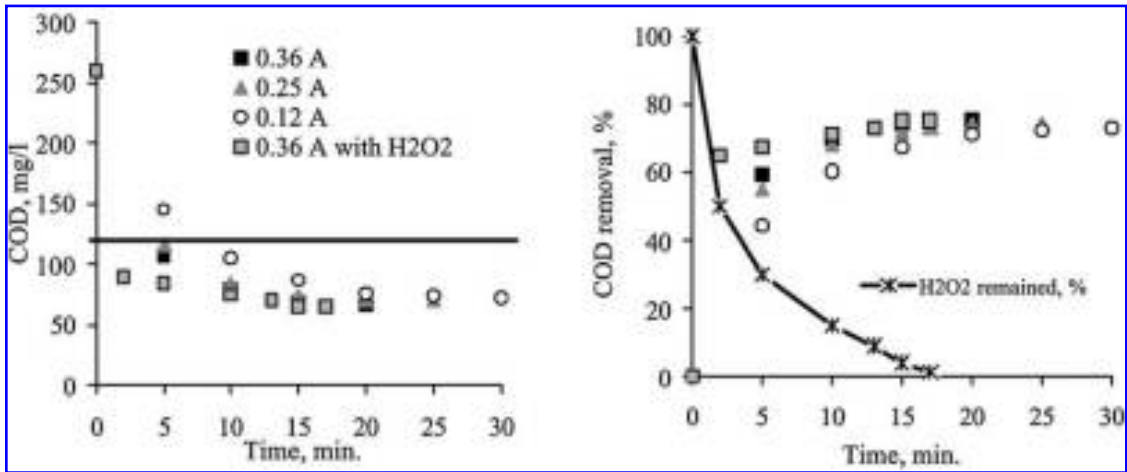**Figure 4.** Changes of COD in experimental runs for COD_i: 380 mg/L.

Figure 5. Changes of COD in experimental runs for COD_i: 260 mg/L.

2. Different from the pH data, an entirely reverse tendency is shown in Fig. 3, illustrated with obtained ORP values.

Figure 2 shows that pH value increases over time. The fact that this case illustrates that cathodic activity is more dominant in this type of electrochemical treatment can be explained quite well by Equation (1). But, when used H_2O_2, pH was increased in a slower rate, as different from no added oxidant cases. This case can be possibly based on increasing Fe^{3+} ions that makes bond with one more hydroxyl ion than Fe^{2+}. Therefore, oxidant addition pulls out alkalinity in the solution.

ORP data verifying the redox potential of the solution in mV has been shown in Fig. 3, for the DWW electrocoagulation process. ORP decreased during the treatment time in the figure means that the treated-effluent became more and more reductive owing to the presence of dissolved ferrous ions in DWW.

Pollutant removal results

To determine the performance of EC treatment for two different DWW samples, using three different current intensities (A), we performed the measurements of COD, an essential organic polluting parameter, and the removal results we found are illustrated in Fig. 4 (for initial COD: 380 mg/L) and Fig. 5 (for initial COD: 260 mg/L). By taking into consideration the limit established by the Local Authority in Istanbul (ISKI) regarding aquatic medium discharge (for over 100,000 inhabitants), which is 120 mg COD/L, Fig. 4 shows that the optimum value is reached in
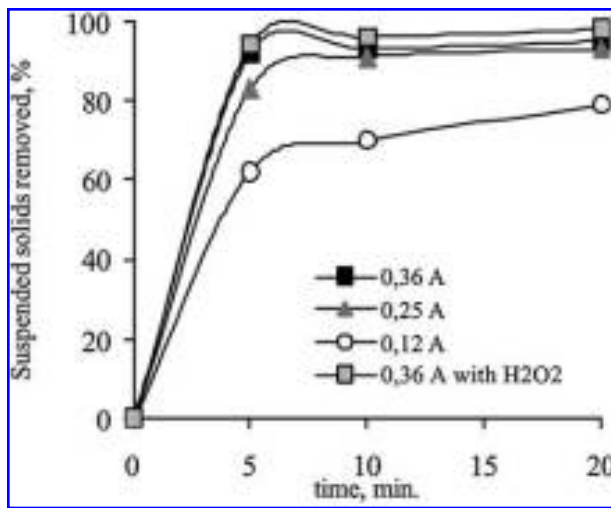

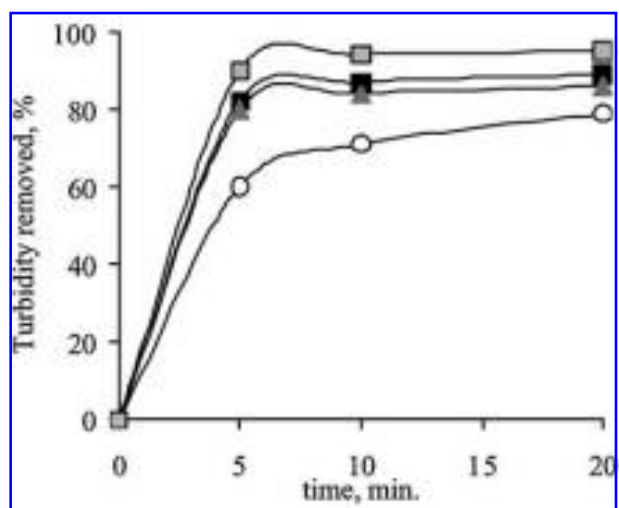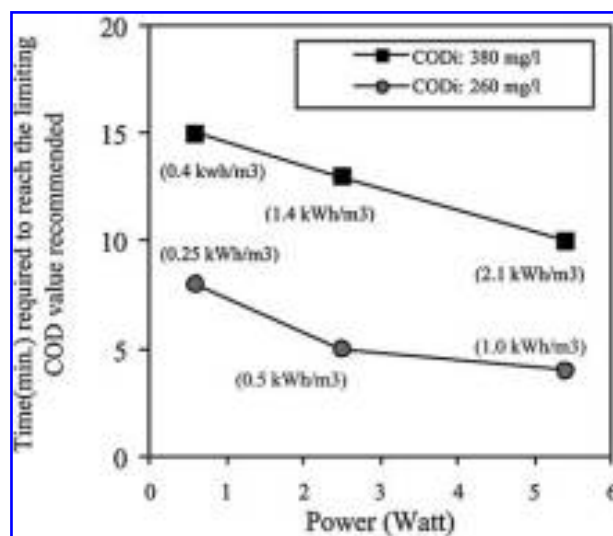**Figure 6.** Removal efficiency of SS at the EC process.

Figure 7. Turbidity values at the EC process.

10 min using a 0.36 A current, in 13 min with 0.25 A current and in 15 min with 0.12 A, for DWW samples with 380 mg CODi/L polluting strength. The other DWW sample, with 260 mg CODi/L strength, could be treated by the discharge limit in 5 min by applying 0.36-A and 0.25-A current. In cases with current intensity as low as 0.12 A, the limit was met in about 8 min. It is noted from Fig. 4 and Fig. 5 that 60–70% COD removal percentages are obtained by the EC process. In fact, a coagulation process sweeps and precipitates the suspended solids by flocks formed. Turbidity and suspended solids were effectively removed from DWW owing to aggregating colloidal particles by means of Fe^{2+}/Fe^{3+} produced during electrolysis. These ions react with colloids to form settable flocks that can be easily separated from solution. Figures 6 and 7 confirm this phenomena that SS and turbidity decrease by negligible values of beginning concentrations. But 60–70% COD removal percentages do not originate only from SS removal, on the other hand, during electrocoagulation, many processes such as precipitation, coprecipitation, sweep coagulation, cathodic reduction, electrostatic attraction on the surface of metallic hydroxides, and adsorption can take place.
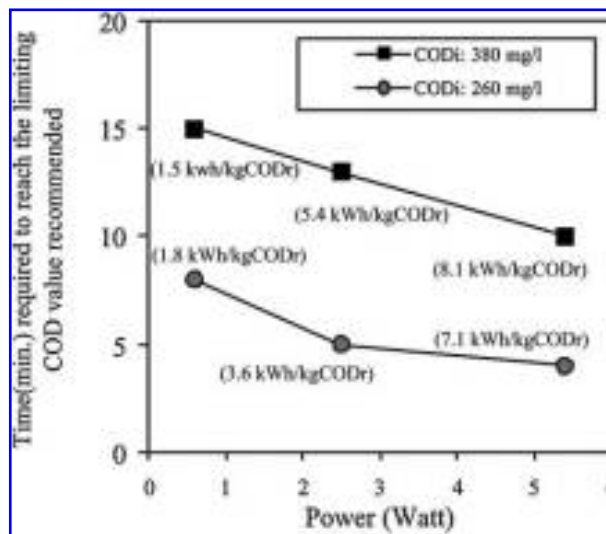
When H_2O_2 was added, the efficiency of COD reduction for treated DWWs was prominently increased, as given in Figs. 4 and 5. For RLDWW and WLDWW, treated COD values were analyzed as 95 mg/L in 3 min and 90 mg/L in 2 min, respectively. Oxidation potential of H_2O_2 and higher precipitation capability of ferric hydroxides have taken a role in the formation of these results.

Energy consumptions

Energy consumption data calculated for COD removal are given in Figs. 8 and 9. Pollutant load is sufficiently reduced

Figure 8.   Energy consumption for m^3 DWW treated up to discharge limit (120 mg COD/L).

Figure 9.   Energy consumption for kg COD removed.

using a moderately low electrical current in an electrolysis cell. The EC mechanism is effective in a few minutes on DWW samples. To maintain the discharge limit of the local authority, electrolysis time was 15 min for RLDWW and 8 min for WLDWW, applying 0.6 Watts of electrical power. In the case of using a higher electrical power, like 2.5, 3.6, and 5.4 Watts, the process causes energy cost increases. Contrarily, the efficiency of COD removal is not considerably enhanced. Optimum energy costs were obtained with 0.4 kW-h/m³ DWW for 380 mg COD/L loaded DWW and 0.25 kW-h/m³ DWW for 260 mg COD/L loaded DWW. As can be seen from Figs. 8 and 9, if electrical power decreases to a 1/9 ratio (from 5.4 W to 0.6 W), the reaction time is about 1.5–2 times longer (from 10 min to 15 min, and from 4 to 8 min) but energy consumption decreases four to five times (from 2.1 kWh to 0.4 kWh and from 1.0 kWh to 0.25 kWh for m³ DWW for relatively loaded and weakly loaded DWWs, respectively). For both effluents (RLDWW and WLDWW), it can be seen than the energy consumed (kWh/m³) increased while increasing the electrical power (watts). It means that the treatment time is the predominant factor in the electrocoagulation process used to remove COD from DWW.

Sludge produced

At the process, the activity of EC mechanisms on the removal of SS and turbidity was investigated and data connected with this subject were given before in Figs. 6 and 7, respectively. In addition to this, the volume and the inorganic fraction of precipitated sludge, as well as the amount of dissolved iron from the anode material, were measured and the results are shown in Figs. 10 and 11. All mentioned parameters are vital for the practical viability of the EC process.
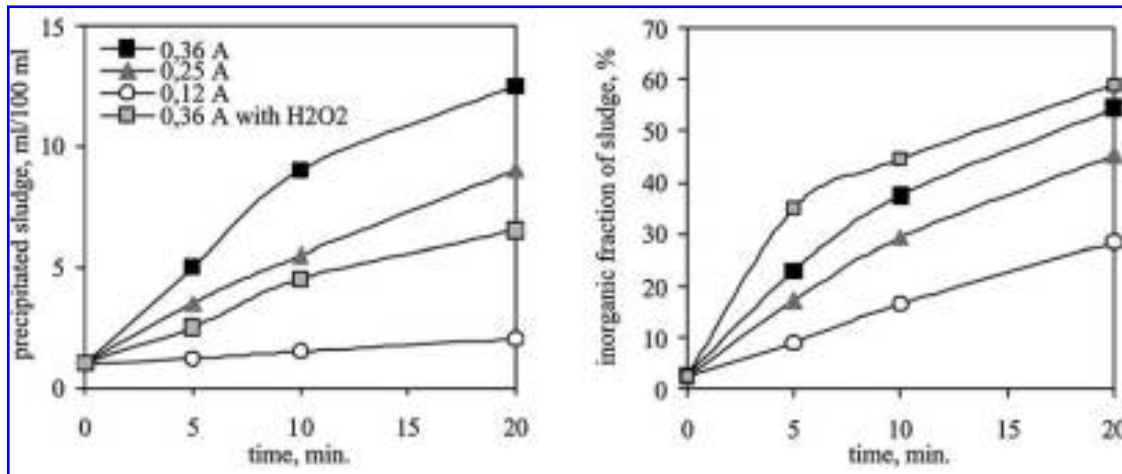
Figure 10. Features of precipitated sludge.

According to the results in Fig. 10, the levels of sludge production remained between 1.5 and 9% in 5–15-min reaction time. The inorganic fraction of obtained sludge was increased gradually from 13 to 37% in 5–15 min as a function of current intensity.

The chemical structure of the sludge formed determines sludge volume. By the EC reaction [Equation (5)], green-colored $Fe(OH)_2$ sludge is formed. If oxidant species, such as H_2O_2, are present in the media, according to Equations (7) and (8), Fe(II) ions convert to Fe(III) ions and brownish colored $Fe(OH)_3$ sludge is formed despite the presence of $Fe(OH)_2$ sludge.

In a review, Liu (2003) declares that the excess sludge production was reduced by 50% when ozone applied in an aeration tank. To reduce the sludge deposition, EC runs with 0.36 A are repeated by using 200 mg H_2O_2/L. This concentration is sufficient to convert Fe^{2+} to Fe^{3+} for 20 min of reaction time according to Faraday's law. At the end of these studies, it was seen that the sludge deposition levels decreased 50% as rounded. While precipitated sludge volume was 12.5% using a 0.36-A current without H_2O_2, this value was reduced to 6.5% at the same electrical current and with H_2O_2.

To understand the difference and get a comparison, some chemical coagulation (CC) works in parallel with the above, a given sludge volume reduction work was performed for the same iron concentration (300 mg Fe/L) and pH value (around 8.7). The experimental works were made for two different iron ion forms ($FeSO_4$ for Fe^{2+} and $FeCl_3$ for Fe^{3+}) and hydrogen peroxide additions. Precise experimental work results for a 1-h setting following CC is presented in Table 2. Also, the table clearly indicates that EC causes 30% lower sludge volume compare with CC. In addition, EC with oxidant addition supplied 40% more improvement in sludge minimization in comparison with CC with an oxidant addition. If EC with H_2O_2 is compared with only Fe^{2+} CC or Fe^{3+} CC treatments, at least 60% volume reduction is determined. In fact, the characteristics of the EC-treated flocks differ noticeably from those generated by CC, by containing less bound water, more shear resistance, and more filterability (Mollah *et al.*, 2001). In addition that, the extra upgrading effect on the sludge minimization of oxidant addition during the EC process was possibly due to stabilization of the organics supplying a
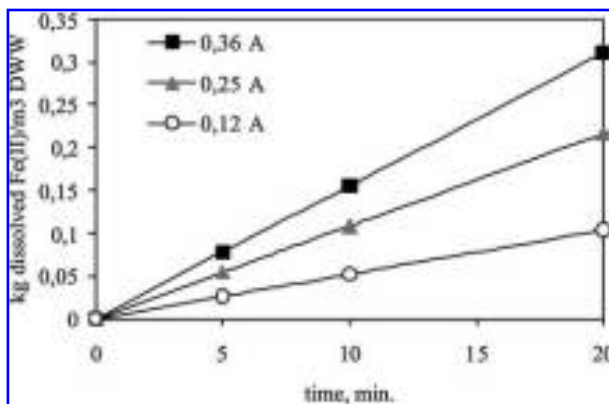

Figure 11. Amount of iron dissolved from the electrode for 1 m³ DWW treated.

Table 2. Sludge amounts produced with EC and CC processes.

Process	mL/100 mL
EC	12.5
EC with H_2O_2	6.5
Fe^{2+}CC	17.5
Fe^{3+}CC	16.5
Fe^{2+}CC with H_2O_2	10.5

difficulty on the settling capability in the wastewater and sludge matrix (Liu, 2003).

Developing iron concentrations by EC

Iron as an electrode can be preferred as a material, because it is a very cheap and nondangerous material. According to the results in Fig. 11, all of the pollution parameters for RLDWW are decreased to discharge limits of the local authority using 0.12 A for 15 min where 78 g Fe is dissolved to treat 1 m^3 DWW. If 0.25 A and 0.36 A were used, the values increased to 140 g Fe in 13 min and 155 g Fe in 10 min.

In WLDWW, the amounts of dissolved iron needed to treat 1 m^3 DWW for the applications of 0.36 A–4.5 min, 0.25 A–5 min, and 0.12 A–8 min were determined as 70 g, 54 g, and 41 g, respectively.

CONCLUSIONS

In this lab-scale study, relatively loaded and weakly loaded DWW was successfully treated by an EC process at very short times (5–15 min). Optimum electrical power and current intensity requirements supplying the local sewage discharge limit was determined as 0.6 Watt and 0.12 A, respectively. Therefore, the energy consumption of the process changes based on the pollutant strength of DWW and applied electrical power.

In RLDWW, using lower electrical power—0.6 W instead of 5.4 W (nine times smaller)—decreased energy consumption from 2.1 kWh to 0.4 kWh (about six times less) for each m^3 DWW treated. In WLDWW, a similar tendency (about four times; from 1.0 kWh to 0.25 kWh for 1 m^3 DWW treated) in energy consumption was observed for the same electrical power applications. These findings put forward a normal expectation for the wastewater strength—an energy consumption relationship.

For WLDWW, the removal efficiencies of SS and turbidity were obtained at 70% under optimal conditions (0.6 W and 8–15 min electrolysis time).

The levels of sludge production were between 1.5 and 2% in optimal reaction times of 8–15 min. Although no chemical was applied, the inorganic fraction of precipitated solids was 2.6%. When EC treatment was applied, the inorganic fraction of obtained sludge was enhanced as a function of current intensity, and changed from 14 to 23%, for powers between 0.6 and 5.4 W. By using H_2O_2, sludge volume of $Fe(OH)_3$ was less by about 50% in comparison with the sludge volume of $Fe(OH)_2$.

The amount of iron dissolved from the anode to treat 1 m^3 WLDWW was realized at about 40 g under optimal conditions. The amount dissolved for RLDWW was about 75 g. When all of the results were evaluated, the EC process for the treatment of DWWs can be put forward as an advanced treatment method because of its efficiency, low energy requirement, lower, and more stable sludge production compared with conventional (biological) treatment methods. It is considered searching a pilot scale work for the future perspective of this work.

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