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RESEARCH ARTICLE

Evaluation of electrochemical treatment of real hospital wastewater with different electrode materials

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ABSTRACT

In this paper, the treatment of real hospital wastewater (HWW) by electrocoagulation process (EC), which is one of the electrochemical treatment methods, has been evaluated. In the EC process, aluminum (Al) and iron (Fe) plates as anode and cathode are used. Experimental studies were conducted at 5, 10, 20, 30 voltage (V) and 5, 10, 20, 30, 45 minutes (min) exposure times. pH, temperature, and conductivity were monitored. COD and phenol removal were evaluated. As a result of experimental studies, Al and Fe electrodes were effective in the treatment of HWW with EC. The highest COD removal efficiency was 93% at 30V 10 min and 95% at 30V 5 min for Al and Fe electrode, respectively. The highest phenol removal efficiency is 97% at 10V 10 min and 97% at 10V 5 min for Al and Fe electrode. When all parameters are evaluated, optimum electro kinetic conditions for treatment of HWW was obtained for 10V 5 min by the Fe electrode.

Keywords: Hospital wastewater, electrocoagulation, aluminum electrode, iron electrode

1. INTRODUCTION

Hospital wastewater (HWW) is a wastewater similar to the quality of domestic wastewater, whose properties are not well known [1], [2], [3]. HWW contains many hazardous ingredients, including hazardous chemical components, heavy metals, disinfectants, and special detergents used in laboratories and research [4], [5], [6]. HWW is a wastewater obtained from all hospital activities, both medical and non-medical, including activities such as radiology cafeterias and examination rooms. In addition, higher concentrations of pharmaceutical ingredients can be found than are found in domestic wastewater [7], [8]. The degree of pollution of HWW and its polluting properties can vary from day to day from hour to hour [1]. Therefore, treatment disposal and proper management of HWW is an increasing international concern. HWW is generally discharged into city sewer systems. In domestic wastewater treatment facilities, it has to be treated together with domestic wastewater. In different countries, specific treatment methods have been used for HWW output [9], [10], [11], [12].

In recent years, electrochemical treatment methods, which provide more economical and high treatment efficiency, are widely preferred in the treatment of wastewater. Electrocoagulation (EC) is one of the simple and effective electrochemical methods for water and wastewater treatment [13]. In this technique, which is characterized by simple equipment and easy operation, soluble metal hydroxide is produced by electrolytic oxidation of a suitable anode material to a coagulant at a suitable pH to remove large contaminants [14]. These types of metal hydroxides neutralize electrostatic charges on suspended solids to facilitate their separation from the aqueous phase, flocculation and coagulation [15], [16].

This method is used efficiently in various types of wastewater. In this study, the treatment of wastewater obtained from a real hospital with two different electrode types by EC method was evaluated. Also, pH, conductivity changes, COD and total phenol removals were evaluated at different voltage (V) and exposure times and a comparison was made for two electrodes.

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2. MATERIALS AND METHODS

2.1. Experimental setup

EC sets of HWW were conducted to plexiglass reactor (9 cm diameterx13 cm height) (Fig 1). Used wastewater volume was 600-mL for a set. Electrodes were preferred aluminum (Al-Al) and iron and (Fe-Fe) plate as anode and cathode. Electrode dimensions were 6 cm width, 11.5 cm height, 0.1 cm thickness, and their effective areas were 46.2 cm². Distance between electrodes was 2 cm. Before each set, electrodes were cleaned after distilled water and before acetone on electrodes. With DC power supply, an electric field were supplied by between 5 and 30 V in exposure times in EC (Table 1). At the end of each set, coagulated materials allowed to settle for a few hours in Imhoff and then supernatants were taken from Imhoff and analyzed.



Fig 1. Experimental setup

Experiment Sets	Exposure Time (min)	Voltage (V)
Control	0	0
1	5	5
2	10	5
3	20	5
4	30	5
5	45	5
6	5	10
7	10	10
8	20	10
9	30	10
10	45	10
11	5	20
12	10	20
13	20	20
14	30	20
15	45	20
16	5	30
17	10	30
18	20	30
19	30	30
20	45	30

Table 1. E	Experiment sets f	for Al and	Fe electrodes
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2.2. Analytic procedure

In real HWW by EC, pH, temperature and conductivity parameters were measured by a pH 510 Eutech Inst. meter in end of EC sets. COD and phenol were analyzed in supernatants. COD and phenol were analyzed according to Standard Methods 5220-D and 5530-D, respectively (APHA, 2005). In the direct photometric method for phenol, samples were measured at 500 nm by UV/VIS Spectrophotometer. The removal rates of COD and phenol were calculated using the following equation: (%) = $(A0 - A1)/A0 \times 100$ where A0 and A1 represent influent and effluent concentrations of the parameter, respectively. All analyses were conducted in triplicate. All chemicals used analyzed were analytical reagent grade.

2.3. HWW characterization

HWW samples were collected from the discharge point of the HWW to the sewer system in Sisli Hamidiye Etfal Training and Research Hospital, Istanbul, Turkey. Top [1] determined that the pollutant load density of the HWW sent to the sewerage systems is at noon. Therefore, the wastewater sample was taken at 13:00. All experiments have been done on the same wastewater. HWW characterization was given Table 2.

Parameters	Value
COD (mg L ⁻¹)	772
TP (mg L ⁻¹)	0.345
Suspended Solids (mg L ⁻¹)	185
Phenol (mg L ⁻¹)	3.26
Sulfate (mg L ⁻¹)	174
Chloride (mg L ⁻¹)	158
рН	6.87
Temperature (°C)	20.6
Conductivity (µS cm ⁻¹)	929

Table 2. Characteristics of used real HWW

3. RESULTS AND DISCUSSION

3.1. рН

The pH is one of parameters affected EC process. It is responsible for the speciation and solubility of metal oxides and hydroxide in wastewater. Also, pH change in EC contributes to the removal of pollutants in wastewater. Changes in pH during an EC are related to the electrolysis of water and the chemistry of the coagulation process. The rise in the final pH with applying electric field can be easily explained by Equations (Table 3).

The pH values obtained as a result of EC treatment of HWW with Al and Fe electrodes and the first wastewater pH range is wider at the Fe electrode, indicating that a different chemical process will occur with two reagents (Al and Fe) in the coagulation of pollutants. Fig 2 shows the pH change in HWW with Al-Al and Fe-Fe electrodes. According to the results, it was seen that pH gradually increased with the increase in voltage and duration due to the dominant activities of the cathode, depending on the activities of the anode and cathode.

Anode	Cathode
$40H^- + 4e^- = 2H_2O + O_{2(g)}$	$2H_3O^+ + 2e^- = H_{2(g)} + 2H_2O$
	(in aside solution)
$2H_2O + 4e^- = O_{2(g)} + 4H^+$	$2H_2O + 2e^- = H_{2(g)} + 2OH^-$
	(in alkali solution)
$2Cl^- + 2e^- = Cl_{2(g)}$	$O_{2(g)} + 2H_2O + 4e^- = 4OH^-$
Al-anode	
$Al_{(k)} + 3e^{-} = Al_{(aq)}^{3+}$	$Al_{(k)} + 40H^{-} = [Al(0H)_{4}]^{-} + 3e^{-}$
	(in very-high pH)
$Al_{(aq)}^{3+} + 3H_20 = Al(OH)_3 + 3H^+$	
Fe-anode	
$Fe_{(k)} + 2e^{-} = Fe_{(aq)}^{2+}$	
$Fe_{(aq)}^{2+} + 2H_20 = Fe(0H)_2 + 2H_20$	$Fe(OH)_3 + OH^- = [Fe(OH)_4]^-$
$Fe^{2+} + e^{-} = Fe^{3+}$	$[Fe(OH)_4]^- + 2OH^- = [Fe(OH)_6]_3^-$
$Fe^{3+} + 3H_2O = Fe(OH)_3 + 3H^+$	(in very-high pH)

Table 3. Reactions occurred by EC in Wastewater [17]

During the exposure period, the lowest pH increases occurred at 5V. The longest exposure time at 45 min was 7.18 for Al and 8.2 for Fe. When the voltage was 30, the pH increased from 6.87 to 8.57 with the Alelectrode, and from 6.87 to 9.32 with the Fe electrode. The pH increase in the Fe-electrode was higher than in the Al electrode. The higher the pH level, the higher the dose of coagulants in the solution.



Fig 2. pH changes in Al and Fe electrodes

3.2. Temperature and conductivity

10 15 20

Temperature changes in HWW during EC are given in Fig 3. Temperature increase with time is an expected

25

Time (min)

30 35

45 50

situation arising from electrolytic reactions in EC processes [18]. Wang et al. [19] and Ilhan [18] found that there was an increase in temperature depending on the time in EC in their study. Depending on the contact time, the type of electrode, and the applied electrical field, temperatures increase as a result of electrolytic reactions. In these studies, when Al and Fe electrodes were used, the highest temperature was measured at 30V and 41 °C at the Fe electrode. The temperature of the two electrode types varied between 20-41 °C at different voltages and exposure times.





Fig 3. Temperature changes in Al and Fe electrodes

Also, the electrical conductivity changes depending on the contact time and the applied electrical field. The conductivity change is shown in Fig 4. The electric field applied in EC reduces conductivity [20]. In addition, the conductivity is higher in acidic pH than neutral pH. The reason for this is related to hydroxide compounds produced by the presence of Al and Fe ions emitted from the electrode surface, which can adsorb cations and anions into the solution. As a matter of fact, this is the case here. Conductivity starts to decrease with the increase of pH in the Al electrode, this situation is seen more clearly in the Fe electrode. Conductivity decreased at all voltage values after 20 min. of exposure. However, an increase was observed in the conductivity values of Al and Fe electrodes between 0-20 min. In some EC studies, it has been reported that disinfectants or dissolved ions in wastewater may cause an increase in conductivity [21], [22]. Consequently, it is estimated that disinfectants or dissolved ions in HWW may have caused an increase in conductivity.



Fig 4. Conductivity changes in Al and Fe electrodes

3.3. COD removal

Under different voltage, the effect of COD concentration on the treatment of HWW with different electrodes was investigated. In Fig 5, the percentages of COD removal with both Al and Fe electrodes are given. The high removal efficiency was achieved with both electrode materials. The performances of the electrodes varied under the same conditions. Fe electrodes were obtained more COD removal efficiency than Al. For example, the lowest exposure time is 5 min. and the Al and Fe COD removal efficiencies at 5V are 88% and 94%, respectively. However, COD removal efficiencies during the highest voltage and highest exposure (30V 45 min) were 85% and 87% for Al and Fe, which were the lowest removal efficiencies obtained in the study. The reason for this is that high rates of metal coagulants are produced with the increase of pH in EC processes.

Maximum COD removal efficiency was achieved faster at low voltages than at higher voltages [23]. However, in this study, the highest COD removal was obtained at the highest voltage. The reason for this is that the conductivity of HWW is lower than that of other wastewater, so $Al(OH)_3$ compounds and $Fe(OH)_2$ formation have been achieved more in the wastewater at the highest voltage. The increase in pH with the exposure time also increased the bubbles formed in EC and these bubbles caused an increase in COD removal efficiency by increasing the buoyancy of the cell [24]. After 10 min of exposure time at the Al electrode, COD removal efficiency decreased at all voltage values. The decrease in the COD yield at the Al electrode at high pH may be due to the [Al(OH)n] complex formations and the dissolution of Al(OH)₃ in solution (Table 3). After 10 min of exposure for Al, more bubbles are produced from the anode, which means a higher oxygen generation. Therefore, competition occurs between Al dissolution and oxygen formation. Thus, Al(OH)₃ formation was reduced, leading to a decrease in COD removal efficiency [25], [26]. In their study, Verma et al [27] determined that the maximum removal efficiency occurred at neutral pH and the decrease in removal efficiency was due to amphoteric Al(OH)₃ at acidic pH.



Fig 5. COD % removal of HWW with Al and Fe electrodes with EC processes

A different situation occurred in the Fe electrode compared to Al (Fig 5). After exposure time 5 min., the COD removal efficiency started to decrease. As is known, there is a relationship between solution pH and COD removal. The higher the solution pH, the more coagulants (Fe(OH)₂ and Fe(OH)₃) were formed [28]. Subsequently, they adsorbed the contaminant, thus achieving COD removal efficiency at higher solution pH. However, it has been stated that when the solution pH at the Fe electrode is higher than 8.00, the COD removal efficiency will decrease [29], [30]. COD removal efficiencies at all voltages decreased as the reaction time increased. It has been stated that the decrease in efficiency at higher reaction time and voltage may be associated with high production of drug degradation products, competition with major contaminants for coagulant, and reactive species. Similar results have been reported in the study conducted by Ahmadzadeh et al. [31] for the treatment of HWW by EC process. The highest COD removal efficiency was 93% at 30V 10 min and 95% at 30V 5 min for Al and Fe electrode, respectively.

3.4. Phenol removal

Phenol is very soluble in water. If the phenol in wastewater is not removed, it may pose a risk in receiving environments. Due to their toxic effects, phenolic contaminants can damage sensitive cells. Phenolic micropollutants in HWW can cause health and environmental problems. For this reason, it is important to eliminate phenolic compounds in HWW. In Fig 6, phenol removal efficiencies of HWW with Al and Fe electrodes with EC processes are given. Phenol removal increased at all voltages during the 10 min exposure with Al electrodes. Treatment efficiency continued to increase up to the 20 min exposure time, except 20 and 30 V. The highest phenol removal efficiency was achieved at 10V 10 min at 97%. In the Fe electrode, it is 97% in 10V 5 minutes. It has been reported that phenols and organic acids can chemically interact with trivalent cations to form insoluble species through compound complexing, precipitation or coagulation processes [32]. The resulting Fe(OH)3 and Al(OH)₃ and phenol molecules were removed from the water through the formation of surface complexes. Phenol molecules undergo physical adsorption by Van der Waals forces to gelatinous amorphous Fe(OH)3 flocks, which are found on the surfaces of hydroxide flocs (co-precipitation) [33].

Phenol removal efficiency trend from wastewater was found to be similar to COD. Efficiency decreased as the voltage and contact time increased. The low phenol removal efficiency was obtained at high pH. Similar results were found in Abdelwahab et al. [34] was also obtained in the study.

Removal efficiency is very low at low pH or high pH. The higher phenol removal efficiency was obtained with a neutral pH. This behavior has been attributed to the amphoteric character of Al(OH)₃, which does not precipitate at very low pH [14]. It has also been determined that high pH leads to the formation of Al(OH)₄⁻, which is soluble and useless for the adsorption of phenol [35]. Therefore, it has been reported that further increasing the initial pH value decreases the phenol removal efficiency.

According to Faraday's Law, the electrolysis time in the EC process affects the rate of metal ions released into the system, therefore, as the EC time increased in this study, the COD and phenol removal efficiencies decreased compared to the low voltage and EC time [36]. Therefore, the treatment of HWW with a Fe electrode has been found more appropriate economically and environmentally.

In addition, the total suspended solids determination in the study was made at the end of EC, but the effluent TSS concentration was <0.1 mg L^{-1} in most of the experiment sets. The positively charged metal ions generated during EC can reduce the TSS concentration by destabilizing the negatively surface charged colloidal pigment particles by scavenging [37], [38].



Fig 6. Phenol % removal of HWW with Al and Fe electrodes with EC processes

4. CONCLUSIONS

In the study, the treatment of real HWW by EC was evaluated using Al-Al and Fe-Fe electrodes. During EC, both electrode materials increased with increasing pH exposure time and voltage. Similarly, the temperature change was between 20-41 °C in all sets with increased temperature. The highest COD removal efficiency from wastewater was 93% at 30V 10min and 95% at 30V 5min for Al and Fe electrode, respectively. The highest phenol removal efficiency is 97% at 10V 10 min and 97% at 10V 5 min for Al and Fe electrode. High voltage and EC time increase the operating cost in EC studies. when COD and phenol removal Accordingly, evaluated, efficiencies were the optimum electrokinetic conditions in the treatment of HWW were 10V 10 min and 10V 5 min for Al and Fe, respectively. Therefore, economically and environmentally, optimum electro kinetic conditions in treatment of HWW was obtained for 10V 5min by Fe electrode.

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