original article

Treatment of hospital wastewater by electrocoagulation using aluminum and iron electrodes

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ABSTRACT

Aims: The main goal of this study was to determine of the removal efficiency of chemical oxygen demand (COD) from educational hospital waste-water using electrocoagulation process by using iron and aluminum electrodes.
Materials and Methods: A laboratory-scale batch reactor was conducted to determine the removal efficiency by the electrocoagulation method. Fifty-five samples of Shahid Mohammadi Hospital waste-water in Bandar Abbas were collected for the periods of 6 months according to standard methods. The removal of COD from the waste-water was determined at pH 3, 7, and 11 in the voltage range of 10, 20, and 30 V at the operation time of 30, 45, and 60 min. Data were analyzed in SPSS (version 16) using Pearson's correlation coefficient to analyze the relationship between these parameters.
Results: The removal efficiency is increased by 6.2% with decreasing pH from 11 to 3 at the optimal condition of 30 V and 60 min operation time. By increasing the reaction time from 30 min to 60 min at voltages (10, 20, and 30 V), the removal efficiency was increased from 32.3% to 87.1%. The maximum COD removal efficiency was observed at pH 3 and voltage of 30 V and 60 min reaction time using four iron electrodes. Pearson correlation analysis showed a significant relationship between voltage and the reaction time with the removal efficiencies ($P < 0.01$).
Conclusion: Due to the high efficiency of the electrocoagulation process and also the simplicity and relatively low-cost, it can be used for removing COD from hospital waste-water.

Key words: Bandar Abbas, chemical oxygen demand, electrocoagulation, hospital waste-water

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INTRODUCTION

Hospital waste-water is one of the most dangerous types of pollution. These waste-waters are contaminated with pathogens, such as bacteria, viruses, and parasites, as well as hazardous chemical compounds, pharmaceutical compounds, and radioactive isotopes. Protection of water resources in a country like Iran, which has many climatic constraints, is very important. It is therefore necessary to

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treat the sources of pollution such as sanitary waste-water and industrial waste-water before being discharged on to receiving waters.[1]

In general, the most important goals of waste-water treatment are controlling the pollution, preventing the infectious, and chronic diseases, protecting the environment, and reusing the waste-water.[2] Hospital waste-water contains a large number of pathogens and this reveals the importance of the sources of pollutants. In case hospital waste-water is properly treated, it can be reused for agricultural purposes.[3] Electrical coagulation is a process of waste-water treatment by electrochemical method through, which direct current electricity is used in order to remove the contaminants from the solution. In this process, a coagulant is produced in place through electrolytic oxidation of an anode which is made of appropriate materials. Then, charged ionic species are removed by allowing the reaction to the opposite charge or the metal hydroxides produced within the waste.[4]

Recently, the electrocoagulation technology is highly acceptable for waste-water treatment, due to the need for simple and easy operation, good settling ability of the sludge, lower sludge production, bigger produced flocks compared to the chemical treatment, and reducing the secondary pollution by not using the chemical compounds and also being economic, safe and environmentally friendly.[5-8] The other capability of this process is the removal of contaminants, such as heavy metals. For instance, it is utilized for the removal of chromium, colloidal and suspended solids, fat, oil, grease, organic compounds, bacteria, viruses, cysts, dye mono-azo acid red, and dye oranges from aqueous environment.[5-13] Electrocoagulation process is suitable for a wide variety of waste-water treatment plants, such as dairy products,[14] removal of cyanide, biochemical oxygen demand, and chemical oxygen demand (COD) from olive oil waste-water[11] and removal of detergent from industrial waste-water of automobile industry.[15] The results showed an increase in the removal of dexamethasone (up to 38.1%) with a rise of the current applied and a decrease of the electrode distance, in aqueous solutions.[16] Bazrafshan *et al*. indicated that electrocoagulation process is able to remove COD from real dairy waste-water up to 98.84% at 60 V during the 60 min operation.[17] The maximum COD removal efficiency was measured 82% at 100 mg/l dye concentration by electrocoagulation.[18] The removal of COD from cardboard paper mill effluents was investigated using aluminum and iron electrodes. The maximum removal efficiencies of COD under optimal operating conditions (pH = 5.29 for Al electrode and pH = 7.21 for Fe electrode) with a current density of 4.41 mA/cm^2 and operating time of 10 min were 99.93% and 99.92% for Al and Fe electrode, respectively.[19] The removal of heavy metals and COD from real industrial waste-water were investigated by electrocoagulation. COD was removed up to 83.94% and 53.83% by Al and Fe electrodes, respectively.[20]

In cities with a sewage collection system, it is possible to dispose the hospital waste-water to the networks, however, if a city has no sewage collection systems, complete hospital waste-water treatment must be provided. Conventional waste-water treatment plants are not able to meet the effluent quality standards for the hospital waste-water effluent. Although the electrocoagulation process has been used for the treatment of many synthetic waste-waters, so far this method has not been used for the treatment of hospital waste-water. Furthermore, the effect of interfering compounds present in the real waste-water has been intensively studied using the electrocoagulation method. Therefore, the objective of this study was to

i. Evaluate the feasibility of using the electrocoagulation process for the treatment of Shahid Mohammadi hospital waste-water and
ii. Determine the optimal condition for maximum COD removal efficiency.

MATERIALS AND METHODS

All the tests were performed at a bench-scale batch reactor mode at room temperature and normal pressure. The study was conducted in a laboratory scale using two-fold and four-fold iron and aluminum electrodes [Figure 1]. An electrochemical Pyrex cell made of 10 mm thickness glass and 2.25 lvolume with dimensions of 12 cm × 12 cm × 16 cm and iron and aluminum plate with dimensions of 12 cm × 10 cm × 2 mm was used as the electrode. The electrodes were vertically separated by 2 cm from each other. The end of each electrode was connected to a direct current (DC) power supply. Table 1 summarized the pilot characteristics were used for the removal of COD from Shahid Mohammadi hospital waste-water using the electrocoagulation method. The mixing was performed by a magnetic stirrer at a constant speed of 100 rpm. Control was also used for this study to decrease the effect of volatile organic content in the waste-water.

All chemicals were purchased from Merck (Germany). Hydrochloric acid of 15% wt was used to clean the electrodes before starting the experiment. The pH was adjusted by sodium hydroxide (NaOH) and sulphuric acid (H_2SO_4).

Figure 1: The Schematic design of electrocoagulation

The educational hospital used in this study is one of the largest hospitals in Hormozgan province, which includes 450 beds and 23 wards. The volume of the sewage produced by the hospital is over 1000 m^3/day. The extended aeration activated sludge is used to treat the waste-water which is not efficient in removing COD. Fifty-five samples (3 l) of Shahid Mohammadi hospital waste-water in Bandar Abbas were collected for the periods of 6 months according to standard methods.[21] The waste-water characteristics such as COD, phosphate, nitrate, and turbidity were measured.[21] Data regarding the chemical and physical properties of the waste-water sample were summarized in Table 2. The effects of different parameters (pH, voltage, reaction time, and electrode type) on the reduction rate of COD (216 samples) were determined at three replications. All the data were presented based on the mean. In this research, influent waste-water samples were studied in order to determine the optimal conditions. The studied parameters were electrode materials (aluminum and iron) with the arrangement of iron-iron, aluminum-aluminum and aluminum-iron, the number of electrodes (2 and 4 electrodes), operation time (30, 45, and 60 min), voltage (10, 20, and 30 V) and the current intensity between 1 and 5 amperes at different pH (3, 7 and 11). The effect of distance between two electrodes (2 or 3 cm) was also investigated. All the experiments were done in three replicates. In each set of experiments, the samples were taken from the reactor and COD was determined using the titration method according to the standard method (5220).[16] Moreover, turbidity and phosphate were measured by a nephelometer (HACH Company, USA) and spectrophotometer (DR 5000) according to the standard method (2130) and (P-4500), respectively. COD removal efficiency was calculated according to the formula (1).

$$E = C_0 - C/C \qquad (1)$$

Where:

E = COD removal efficiency,
C_0=Influent COD before electrocoagulation process,
C = Effluent COD before electrocoagulation process.

The optimal conditions of different parameters were determined according to COD removal efficiency. The data were analyzed using the SPSS statistical software (Version 16) by **Pearson's** correlation coefficient to analyze the relationship between these parameters.

RESULTS

According to previous studies conducted on different waste-water, pH, a voltage and operation time were selected in the range of pH 3, 7 and 11, the voltage of 10, 20 and 30 V and the operation time of 30, 45, and 60 min, respectively. Since, the current study was conducted on the real hospital wastewater, the fluctuation of COD was observed during the period of 6 months sampling of waste-water. First of all, the optimal voltage to achieve the maximum COD removal efficiency from Shahid Mohammadi waste-water was determined. According to Figure 2, COD removal efficiency during the electrocoagulation process using two pairs of iron-iron electrodes at the pH of 3 and 30 min operation time and the voltages of 10, 20, and 30 V were 32.3%, 42.1%, and 54% respectively. COD removal efficiency was increased by increasing the voltage. The same trend was seen by increasing operation time as well. The maximum removal efficiency at the applied voltage of 30 V at 60 min operation time was 87.1%.

Next, the effect of pH on removing COD from the waste-water was examined. According to Figure 3, COD removal efficiency

Table 1: Pilot characteristics used for the removal of COD from Shahid Mohammadi hospital waste-water using the electrocoagulation method

Reactor characteristics	
Length (mm)	120
Width (mm)	120
Height (mm)	160
Reactor dimensions	
Thickness (mm)	10
Volume (L)	2.25
Free board (cm)	4
Electrode dimensions	
Length (mm)	120
Width (mm)	100
Thickness (mm)	2

COD: Chemical oxygen demand

Table 2: The chemical and physical properties of Shahid Mohammadi Hospital waste-water using the electrocoagulation method

Test	Unit	Result
pH	–	6.2-8.3
Flow rate	m^3/day	1000
Turbidity	NTU	186
COD	mg/l	398
Phosphate	mg/l	35
Nitrate	mg/l	1

COD: Chemical oxygen demand, NTU: Nephelometry turbidity unit

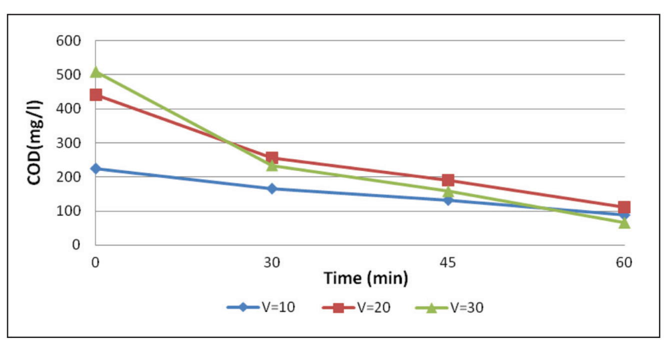

Figure 2: The reduction of chemical oxygen demand versus time during electrocoagulation process using four pairs of iron-iron electrodes at different voltages and optimal pH = 3

during the process of electrocoagulation using two pairs of iron-iron electrodes at voltage of 30 V, the operation time of 30 min and pH of 3, 11, and 7 were 54%, 47.1%, and 44%, respectively. By decreasing pH to 3, the removal efficiency was increased. The removal efficiency at the operation time of 45 min and pH of 3, 11, and 7, were 69%, 62.2%, and 58.3%, respectively. By increasing the operation time to 60 min, the removal of COD at different pH of 3, 11, and 7 were 87.1%, 80.1% and 76.62%, respectively. Therefore, the optimal pH to achieve the maximum removal efficiency was pH = 3.

After determining the optimal voltage and pH, the next attempt was to examine the effects of the electrode material and also the number of electrode on the COD removal efficiency. Using one pair of iron-aluminum electrodes at the optimal operation condition (pH = 3, the operation time of 60 min and applied voltages of 10, 20 and 30 V), the removal efficiencies were 20%, 27% and 36%, respectively. The removal efficiencies for one pair of aluminum electrodes for the optimal operation condition were 23%, 29.5%, and 39.1%, respectively. Interestingly, using one pair of iron-iron electrodes their removal were 30.8%, 37.8% and 42%, respectively. Obviously, the maximum removal efficiency of 42% was obtained with one pair of iron-iron electrodes. Using two pairs of iron-aluminum electrodes at the optimal operation condition, the removal efficiencies were 42%, 53% and 66.7%, respectively. Using two pairs of aluminum-aluminum electrodes, the removal efficiencies were 42%, 59.2% and 75.1%, respectively. The results indicated that using two pairs of iron-iron electrodes, the removal efficiencies were 64%, 75%, and 87.1%, respectively. The experiment demonstrated that using two pairs of iron-iron electrodes are the most efficient and we succeeded to achieve the COD removal efficiency of more than 87% from the hospital waste-water.

Another parameter that affects the COD removal efficiency was the time of electrolysis. Using one pair of iron-aluminum electrodes at the optimal condition (pH = 3 and voltage of 30 V, the operation time of 30, 45 and 60 min) resulted the removal efficiency of 16.8%, 25.5% and 36%, respectively. Using one pair of aluminum-aluminum electrodes, the removal efficiencies were 19%, 27.1% and 39.1%, respectively. While using one pair of iron-iron electrodes, the removal efficiencies of 25%, 33% and 42% were achieved [Figure 4]. Using two pairs of iron-aluminum electrodes, the removal efficiencies were 26.3%, 44.5%, and 66.7%, respectively. Whereas using 2 pairs of aluminum-aluminum electrodes, the removal efficiencies were 35.1%, 51.1%, and 75.1%, respectively. Using two pairs of iron electrodes, we achieved the removal efficiencies of 54%, 69%, and 87.1%, respectively [Figure 5]. The results indicated that by increasing the operation time the removal efficiency was increased. The maximum COD removal efficiency of 87.1% was achieved using 2 pairs of electrodes and the operation time of 60 min, whereas the minimum removal was obtained using one pair of iron-aluminum electrodes and the operation time of 30 min.

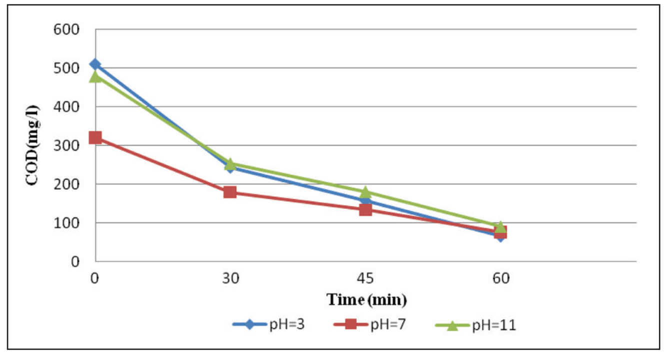

Figure 3: The reduction of chemical oxygen demand versus time during electrocoagulation process using four pairs of iron-iron electrodes at different pH and optimal voltage of 30 V

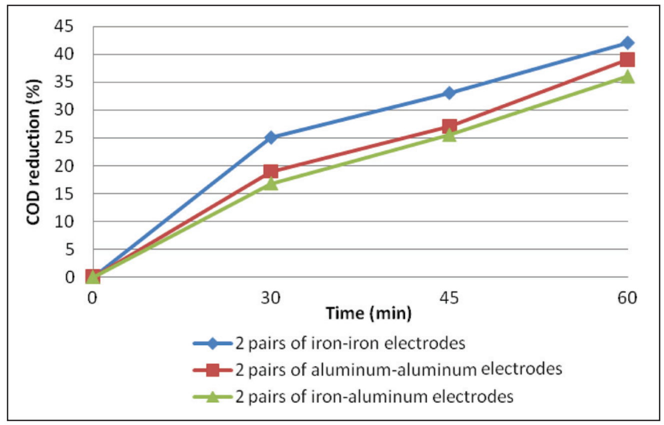

Figure 4: The percent reduction of chemical oxygen demand versus time during electrocoagulation process using different types of two pairs of electrodes at optimal voltage of 30 V and pH = 3

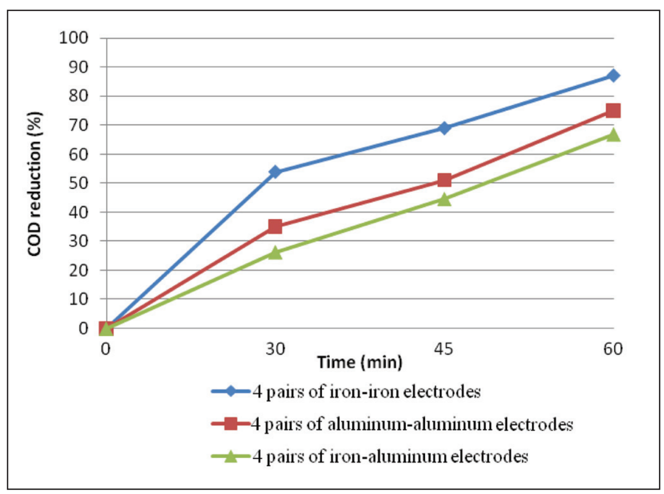

Figure 5: The percent reduction of chemical oxygen demand versus time during electrocoagulation process using different types of four pairs of electrodes at optimal voltage of 30 V and pH = 3

By increasing the distance between the two electrodes (2 cm, 3 cm), COD removal efficiency was reduced from 87.1% to 68%. In addition, using 2 pairs of iron-iron electrodes at the optimal condition (pH = 3, voltage of 30 V and the operation time of 60 min) the removal efficiencies of turbidity, phosphate and nitrate were 100%, 93.57%, and 75%, respectively.

DISCUSSION

According to Pearson correlation coefficient, there is an inverse relationship between the applied voltage and the removal efficiency of COD from waste-water ($P < 0.01$). The results obtained in this study showed that by increasing the voltage at a constant electrolysis time, the removal efficiency is increased. In other studies, researchers have reached the same result. Rahmani's studies also declared that the COD removal efficiency is increased by increasing the voltage.[22] By increasing the applied voltage, the current density is increased between the electrodes. Since the rate of electron flow is increased, the productions of ionic metals are accelerated and therefore the rate of the electrocoagulation process was enhanced.

In addition, the number of pairs of electrodes used in the electrocoagulation process has a large effect on the rate of COD removal efficiency. In fact, two pairs of electrodes result in a much greater removal efficiency compared to one pair of electrodes. On the other hand, the electrode material is also an important parameter affect the removal efficiency. The maximum removal is achieved using iron pair electrodes. The comparison of the results obtained in this work demonstrated that the orders of the removal efficiencies are as follows: iron-iron electrodes > aluminum-aluminum electrodes > iron-aluminum electrodes.

According to the results of this study, there is a significant relationship between pH and the COD removal efficiency, i.e., the COD removal efficiency decreased with increasing pH from 3 to 11. The same results were obtained by other studies which revealed that the removal efficiency of pollutant had an inverse relationship with increasing pH.[23] Generally, initial pH and final pH of electrochemical cell have an effect on the dissolution of electrodes and the form of aluminum or iron species are mainly depend on pH of the solution.

Increasing the operation time had a major role in the performance of the electrocoagulation process. There are many electrochemical reactions occurring simultaneously at the anodes and cathodes. The main reaction is the destabilization of pollutants. Electrodes which produce coagulants into water are made from either iron or aluminum. Iron and aluminum ions dissolve from the anodes. Released ions neutralized the charged particles and hence, the electrocoagulation process was performed. The removal efficiency was directly related to the concentration of ions generated on the electrodes. The ions concentration increased with increasing the time of electrolysis which in turn caused hydroxide flocks to increase. The results showed that the highest COD removal efficiency occurred at the operation time of 60 min. The effect of electrolysis time has been also considered as the main parameter in other studies. Many different studies demonstrated that increasing the electrolysis time resulted higher removal efficiency of COD, color, heavy metals and phosphate.[22-25] Pearson correlation revealed that there was a significant relationship between COD removal and the electrolysis time ($P < 0.01$). In addition, there is a significant relationship between effluent COD and electrical conductivity ($P < 0.01$).

The results showed that the electrode material had affected COD removal efficiency. Using optimal condition (pH = 3, voltage of 300 V and operation time of 60 min), the removal efficiency of COD decreased from 87.1% (two pairs of iron-iron electrodes) to 75.1% (two pairs of iron-aluminum electrodes). In addition, the COD removal is decreased to 66.7% when four iron-aluminum electrodes pair was used. The results exhibited the same trend for two electrode pairs, i.e., the highest removal was obtained using iron-iron electrodes, whereas the lowest removal related to iron-aluminum electrodes. This study showed that the maximum COD removal rate was associated with iron electrodes. In typical aqueous environments, iron can dissolve in divalent Fe (II) and trivalent Fe (III) forms, whereas aluminum dissolves only in trivalent form Al (III). Fe (II) can further oxidize to Fe (III) if oxidation reduction potential and pH conditions are suitable. Sengil *et al.* studies also demonstrated that using iron electrodes resulted in getting the highest COD removal efficiency from sewage.[14] Un *et al.* studies also declared that iron electrodes were more effective in removing COD and turbidity,[11] while Irdemez *et al.*, researches demonstrated that aluminum electrodes were more efficient than iron electrodes.[26] Many studies revealed that aluminum electrode is more suitable electrode material for electrocoagulation applications because it produces Al (III) species.[24-26] Metal ions and hydroxides produced by aluminum electrodes are more effective in the destabilization of pollutants. However, iron electrode can be effective in removing COD and turbidity.[11,14]

Since, economic evaluation is an important parameter in selecting an appropriate process for waste-water treatment; optimization was performed regarding electrical energy consumption. Based on the results obtained, electrical energy consumption during the electrocoagulation process and under optimal conditions for iron-iron, aluminum-aluminum and iron-aluminum pair electrodes were 30.6, 47.4, and 52.5 V/h/l, respectively. It can be concluded that the energy consumption rate with iron-iron electrodes was more economical. Operating cost calculations have been made in a few articles. Calculations typically include the cost of chemicals, electrodes and energy. Many studies showed that the cost of electrocoagulation was much cheaper than chemical precipitation. A comparative study showed that electrocoagulation was faster and more economic, consumed less material and produced less sludge, and pH of the medium was more stabilized than chemical coagulation for COD and turbidity removals. According to the results of this study, electrocoagulation can be an economically viable solution for the removal of COD from hospital waste-water.[27-30]

In conclusion, operating system parameters at the optimal condition can provide the COD removal efficiency of more than 87%. The results indicated the effectiveness of electrocoagulation for the treatment of hospital waste-waters. Moreover, data obtained in the present study demonstrated the technical feasibility of the electrocoagulation process using iron electrodes as a reliable method for the removal of COD from hospital waste-water. Due to the high efficiency of the electrocoagulation process and also the simplicity and relatively low cost, it might be considered as a reliable, flexible, fast, effective, and economical method for hospital waste-water treatment.

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