

# Electrocoagulation with iron and aluminum electrodes in a continuous reactor for COD removal in the effluent of an industrial wastewater treatment plant: A water reclamation approach

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## Abstract

*In this study, the performance of an electrocoagulation process in terms of COD removal from the effluent of an industrial wastewater treatment plant was investigated. By using metal electrodes made of iron and aluminum, hydrogen gas was produced at the cathode and  $Fe^{2+}$  and  $Al^{3+}$  ions were generated at the anode. Since a set of operational factors such as electrolysis time, initial pH of the feed, distance between electrode plates, and electric field strength are influential and interdependent in the efficiency of the electrocoagulation process, optimal values for these parameters were determined while suitable initial values have been considered. According to the experimental results, an increase in electrolysis time and application of a higher electric field strength led to an increase in COD removal rate. Additionally, the maximum COD reductions in iron and aluminum electrodes at electrical field equal to 5 were achieved equal to 53 and 54.3, respectively. The optimum values for pH were found to be equal to 7, and a distance between the electrodes equal to 5 cm and 4 cm for iron and aluminum electrodes, respectively. The energy consumption and corrosion of iron and aluminum electrodes under different conditions were also calculated per volume of treated wastewater.*

**Keywords:** Electrocoagulation, Industrial wastewater treatment, Water reclamation, COD removal, Continuous reactor.

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## I. INTRODUCTION

The ongoing global water crisis, driven by factors such as population growth, urbanization, and climate change, has intensified the need for sustainable water management strategies [1]. The discharge of industrial wastewater containing organic and inorganic pollutants poses a significant threat to aquatic ecosystems and the availability of clean water resources [2]. COD removal in industrial wastewater is crucial not only to meet environmental regulations but also to enable the safe reuse of the treated effluent, thereby contributing to the circular economy and alleviating freshwater scarcity [3].

Emerging electrochemical techniques, such as electrocoagulation, have gained attention as promising alternatives for the treatment of complex industrial wastewater [4, 5]. Electrocoagulation using iron and aluminum electrodes was first registered in the United States in 1909. Drinking water treatment by this method was first carried out on a large scale in 1946 [2]. This process utilizes the in-situ generation of metal hydroxides, such as those from iron and aluminum electrodes, to destabilize and precipitate the pollutants through a combination of electrochemical and physicochemical processes [4, 6].

Several recent studies have explored the application of electrocoagulation for the removal of COD from industrial wastewater. Shukla et al. investigated the use of aluminum electrodes for the treatment of textile industry wastewater, reporting COD removal efficiencies of up to 92% [7]. Similarly, Izadi et al. examined the performance of iron and aluminum electrodes in removing COD from pulp and paper industry wastewater, achieving removal rates of 70-90% [8]. Jing et al. conducted a comprehensive and updated review on the use of electrocoagulation for the treatment of various industrial effluents, highlighting the performance and efficiency of this method [9].

Nonetheless, there are a limited number of studies that employ electrocoagulation as a tertiary treatment to enhance the quality of treated industrial wastewater, for better application of the effluent, such as RO feed.

The present study aims to determine the efficient operational parameters of an electrocoagulation process in terms of COD removal enhancement in the effluent of an industrial wastewater treatment plant. For

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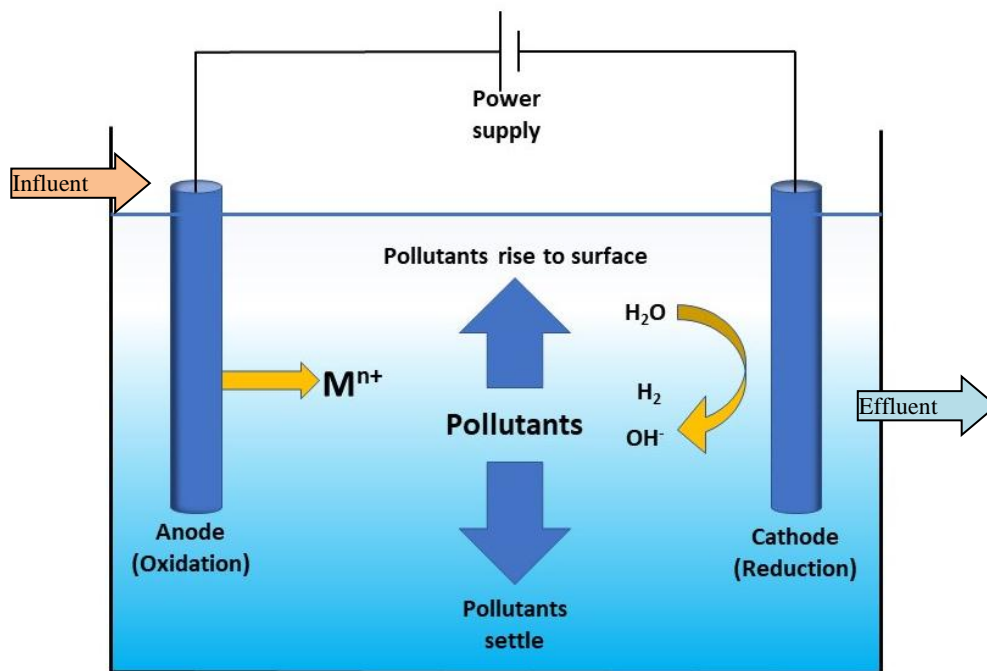
this purpose, the method of electrocoagulation using iron and aluminum electrodes in a continuous reactor setup has been evaluated. This water reuse approach seeks to enhance the quality of the treated effluent, thereby expanding opportunities for its safe and sustainable reintegration into industrial or agricultural water supply networks.

The continuous reactor configuration offers several advantages over batch processes, including improved process control, increased throughput, and reduced operational costs [2]. By using iron and aluminum plates as the electrode in the system, the setup performance has been evaluated and compared. Furthermore, optimum range for influential operational parameters such as the electrolysis duration, distance between electrodes, pH of the reaction medium and the electric field intensity have been determined to maximize the COD removal from the influent. Energy consumption and also the rate of electrodes' corrosion were estimated and the optimal values have been suggested.

The findings of this research contribute to the growing body of knowledge on innovative wastewater treatment strategies that address the challenges of water scarcity and environmental protection.

### 1.1 Fundamentals of electrocoagulation process

The electrocoagulation process involves the in-situ generation of coagulating agents through the electrolytic oxidation of a sacrificial anode, typically made of aluminum or iron. This results in the destabilization of pollutants, allowing for their subsequent removal through precipitation, flotation, or adsorption [10]. This process is schematically shown in Figure 1.



**Figure1: Overall scheme of electrocoagulation process**

In this system, the electrode ions are transferred through the electrolyte. Undesirable pollutants are separated through chemical reactions, precipitation, or by coagulating colloidal materials, and then they are separated by electrolytic flotation through settling and filtration.

The electrocoagulation process is driven by a series of electrochemical reactions that occur at the anode and cathode. At the anode, the metal (usually aluminum or iron) undergoes oxidation, releasing metal ions into the solution [11]. These metal ions then undergo hydrolysis, forming various metal hydroxide complexes that act as coagulating agents.

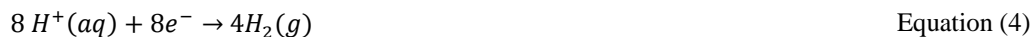
In general, electrocoagulation introduces metal cations electrochemically by consuming sacrificial anodes at the site. Hence the reactions at the anode can be represented as follows:





Where M represents the metal (Al or Fe), n is the valence of the metal ion, and e<sup>-</sup> is the electron.

At the cathode, the reduction of water molecules takes place, generating hydrogen gas and hydroxide ions:



The formation of these hydroxide ions contributes to the overall pH increase in the solution, further promoting the coagulation process [12].

In case of using iron or aluminum in an electrocoagulation system, regardless of the electrode material, the reactions at the cathode involve the reduction of water, generating hydrogen gas and hydroxide ions, as described in Equation (5) to (13).

If an iron electrode is used, the following reactions occur in the anode:



The generated iron ions then undergo hydrolysis reactions, forming various iron hydroxide complexes:



When an aluminum electrode is employed, the anodic reaction involves the oxidation of the aluminum metal, which releases Al<sup>3+</sup> ions into the solution.



The generated aluminum ions then undergo hydrolysis reactions, forming various aluminum hydroxide complexes:



In any case, the amount of metal dissolution and consequently the breakdown of the metal anode depends on the amount of electric current passing through the water and follows Faraday's law [13].

## 1.2 Materials and Methods

The present study is a laboratory experiment conducted as a pilot project in the wastewater treatment plant of Shokoohieh Industrial Town in Qom, Iran. In this research, the influent wastewater to the reactor was collected from the effluent of the sand filter unit of the Shokoohieh Industrial Wastewater Treatment Plant. The quantitative and qualitative specifications of the feed stream to the pilot are presented in Table 1 below.

**Table 1: Quantitative and qualitative characteristics of the influent and effluent wastewater of the Shokourieh wastewater treatment plant.**

Item	Parameter	Unit	Value	
			Average in WWTP influent	Average at the effluent of sand filter
1	BOD5	mg/L	1000	100
2	COD	mg/L	2000	200
3	TSS	mg/L	300	300

In this study, the effect of some important variables such as electrode material, applied voltage, electrolysis time, electrode spacing, pH, as well as electric field on COD removal was investigated, and their optimal values have been determined. The reactor chamber was cuboid-shaped and made of Plexiglas. Considering a hydraulic retention time of 8 hours and a flow rate of 40 liters per hour, its active volume was equal to 32 liters. The influent wastewater tank was located at the top of the chamber and the flow inside the reactor was controlled by an electric valve and level detector. Two pairs of perforated metal plates made of iron and aluminum with an 8 mm hole diameter were used in the reactor chamber to facilitate the flow of electric current towards the membrane during electrolysis.

According to the theory of electrolysis, when the electrodes are connected to the power supply, one of the metal plates acts as an anode and the other acts as a cathode. The power supply used in this study was made in Japan (Toshiba model), with a range of electric current alteration from zero to 3 amperes and a voltage range varying from zero to 30 volts. Additionally, a sludge discharge valve was installed at the bottom of the reactor. Hydrochloric acid and sodium hydroxide were used to adjust the pH. In each series of experiments, samples were taken from the reactor outlet valve, and after filtration, the COD of the samples was measured using a spectrophotometer manufactured by Jen Way Company (England). The removal efficiency after treatment was calculated from the following formula (Equation 14):

$$R(\%) = \frac{C_0 - C}{C_0} \times 100 \tag{Equation 14}$$

in which  $C_0$  is the initial concentration (influent to the reactor) and  $C$  is the final concentration after the process. The amount of iron or aluminum consumed can be calculated from Faraday's law (Equation 15):

$$\Delta m_{theo} = \frac{ItM}{nF} \tag{Equation 15}$$

in which,  $I$  is the current intensity passing through the electric field (A),  $t$  is the electrolysis time (s),  $M$  is the molar mass used (g/mol),  $n$  is the number of exchanged electrodes in anode and cathodes, and  $F$  is Faraday's constant ( $F=96487$  C/mol). Additionally, the amount of electrical energy consumption can be calculated from Equation (16):

$$E = V.I.t \tag{Equation 16}$$

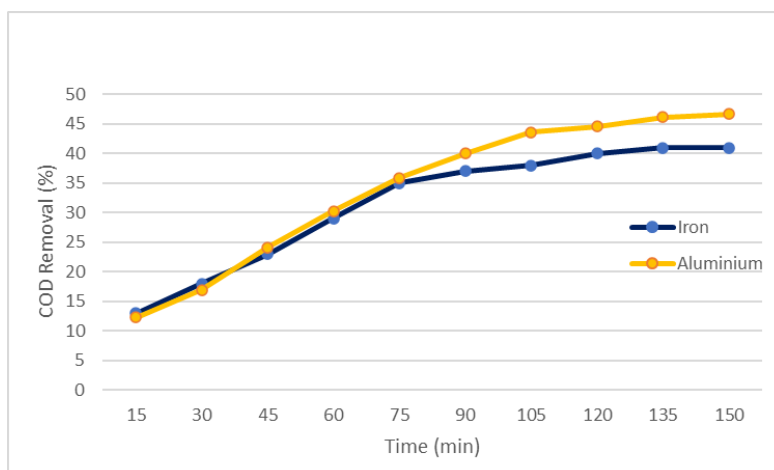
where  $E$  is the amount of electrical energy consumption (kWh),  $V$  is the applied voltage (v),  $I$  is the current intensity (A), and  $t$  is the electrolysis time (s).

## II. RESULT AND DISCUSSION

As discussed earlier, a set of influential operational parameters in the electrolysis process, such as electrode type, electrolysis time, applied current density, initial pH of the wastewater, and electrode spacing, are dependent on each other. In order to have a better approach to the optimized values, initial values for these parameters have been selected on the light of the literature and previous studies to determine the optimal value for each of these parameters, specifically for the conditions applied in this study.

### 2.1 Investigation of the effect of electrolysis time on COD removal efficiency.

Electrolysis time refers to the application of the electrical potential difference between the anode and cathode to establish a constant current density in the electrical circuit. Initially, with an initial pH value of 7, an electric field of 3 V/cm, electrode spacing of 4 centimeters, and an electrolysis time of 15 to 150 minutes, the experiment was conducted on the feed stream with both iron and aluminum electrodes. The results of these conditions on COD removal are shown in Figure 2.



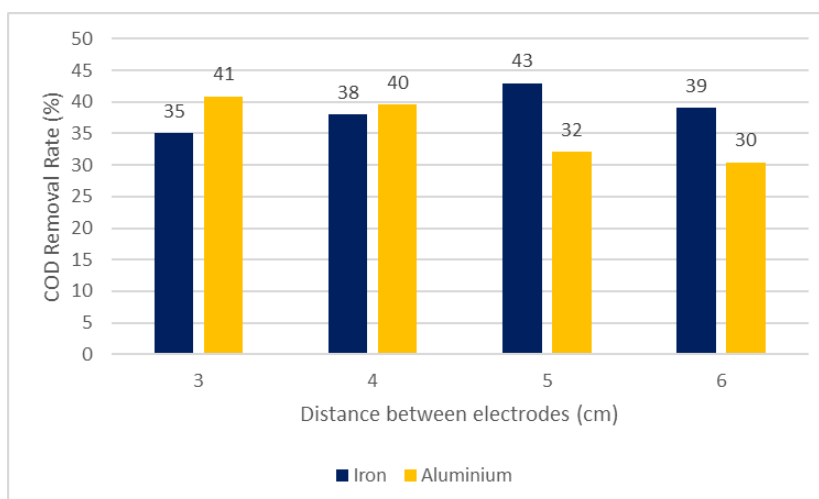
**Figure 2: The changes in COD removal versus time of electrolysis in the electrocoagulation process**

The initial investigation results show a significant decrease in COD levels after electrolysis for both electrodes. According to Figure 2, regardless of the electrode material, around 35% of the total COD has been removed in 75 minutes. Afterwards, the aluminum electrode demonstrated rather better performance in comparison with iron electrode. However, the treatment rate decreased further to the point where there was not much change in the separation range between 105 to 150 minutes (reduction in slope of the curve).

At this stage of the experiment, the maximum removal efficiency at an initial pH of 7, an electric field of 3 V/cm, electrode spacing of 4 centimeters, and after an electrolysis time of 150 minutes was found to be equal to 41% and 46% for iron and aluminum, respectively. Due to the negligible difference in treatment efficiency between the time intervals of 105 to 150 minutes (around 3%), subsequent experiments were conducted within a 105-minute timeframe for energy savings and optimal use.

## 2.2 Investigation of the effect of electrode spacing on COD removal efficiency.

In the next step, variations in electrode spacing were examined to determine the optimal distance under operational conditions of pH=7, initial electric field of cm/3 V, and the electrolysis time from zero to 105 minutes. The experiments were conducted with different distances between the anode and cathode at d=3cm, d=4cm, d=5cm, and d=6cm, for two types of electrodes. Figure 3.a and 3.b demonstrates COD removal results obtained through these sets of conditions.



**Figure 3: The effects of changes in the distances between the iron and aluminum electrode plates on the reduction of COD under operational conditions of pH=2, electric field=1 V/cm, and d=1, 2, 4, 5 cm.**

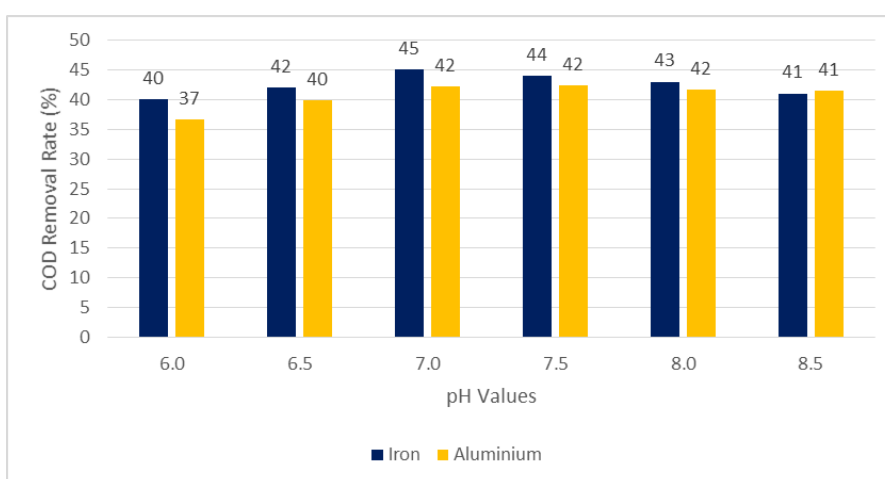
In the tests performed by iron electrodes, results indicated that changes in electrode spacing led to different COD removal rates. From Figure 3, it can be concluded that increasing the distance from 3 to 5 centimeters would lead to increase in the COD removal. However, further increasing this distance to 6 centimeters led to a gradual decrease in the maximum efficiency of COD removal. The same trend occurred for

aluminum electrodes, nonetheless when the distance increased to 4 centimeters, the COD removal rate remained rather constant, but when the distance increased to 5 and 6 centimeters, it drastically decreased.

At this stage and for iron electrodes, the maximum removal efficiency under operational conditions of pH=7, electric field of 3 V/cm, electrolysis time of 105 minutes, and electrode spacing of 5 cm was found to be 43%. Similarly, when pH=7, electric field equals 3 V/cm, electrolysis time equals 105 minutes, and electrode spacing equals 4 cm, the maximum COD removal for aluminum electrodes was 40%, which is slightly less than the results obtained from iron electrodes. Therefore, the optimal distance between electrodes was chosen as 5 cm and 4 cm for iron and aluminum, respectively, for subsequent stages.

### 2.3 Investigation of the effect of pH on COD removal efficiency.

pH plays a crucial role in purification operations through electrolysis and pollutant removal. In treatment systems, especially in biological processes, pH normally ranges between 6 and 8. Considering the influence of pH levels on iron and aluminum complexes ion production, a set of experiments were conducted with an electric field of 3 V/cm, electrolysis time of 105 minutes, electrode spacing of 5 cm and 4 cm (for iron and aluminum electrodes, respectively), and different pH values. The results are presented in Figure 4.



**Figure 4: Variations of pH in the amount of COD removal under operational conditions of d=5 cm for iron and d=4 cm for aluminum electrodes and electric field = 1 v/cm**

The results showed that COD removal efficiency at pH=6 for iron electrodes was slightly lower compared to the data for pH=7 and pH=8. There is not much difference between pH=7 and pH=8, thus the optimal value for pH to maximize the COD removal rate when iron electrodes were employed was equal to 7. The same results have been obtained when aluminum electrodes were used in the system. In detail, the COD removal rate increased while the pH leveled up to 7 and afterwards, it remained rather constant, indicating that the optimal pH value for maximum COD removal in presence of aluminum electrodes was equal to 7. Various studies have been conducted on the effects of pH on pollutant removal efficiencies, which rather confirms the results achieved in this study. Therefore, the optimal pH value for consequent experiments was set equal to 7, for both iron and aluminum electrodes.

### 2.4 Investigation of the effect of electric field intensity on COD removal efficiency.

To examine the effects of electric field intensity on COD reduction efficiency, experiments were conducted with variations in electric field intensity at values of 1, 2, 3, 4, 5 and 6 v/cm under constant operational conditions with pH=7, electrolysis time t=105 minutes, and electrode spacing equal to 5 cm and 4 cm for iron and aluminum electrodes, respectively. The results are presented in Figure 5.

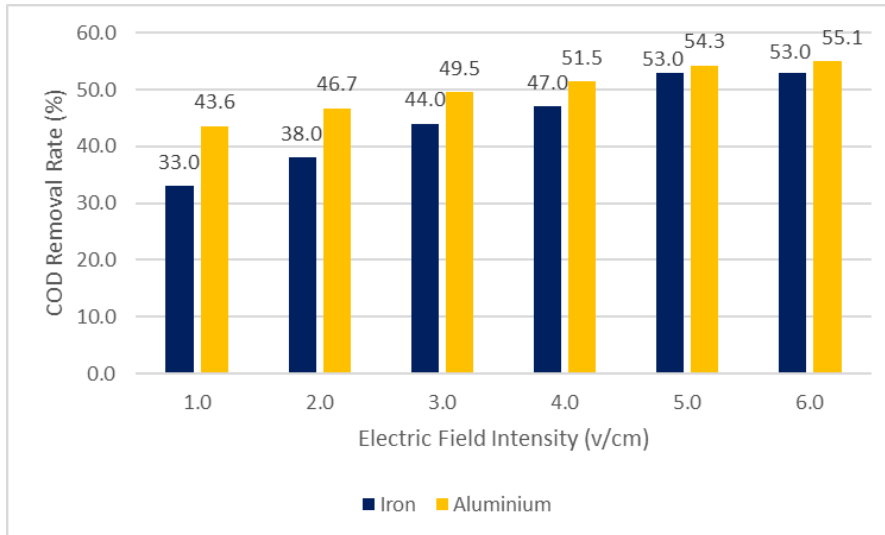


Figure 5: Variations of the electric field in the amount of COD removal under operational conditions of  $d=5$  cm for iron and  $d=4$  cm for aluminum electrodes and  $pH=7$

According to the results obtained, for both electrodes, COD removal gradually rises by the increase in electric field intensity. When the EF reaches to the value of 5 v/cm, it remains rather constant or it changes slightly. Since the COD removal maintains stability after  $EF=5$  and there is a slight and negligible alteration afterwards, the optimum value for EF parameter is considered equal to 5 for both iron and aluminum electrodes. What is more, it can be obviously figured out from the results that in general, aluminum electrodes demonstrate a better performance in COD removal in comparison with the iron electrodes.

### 2.5 Energy consumption and electrode usage.

As previously mentioned, two key parameters that affect the application of electrical coagulation processes in industrial water and wastewater treatment plants are energy consumption and electrode usage during electrolysis. Figure 6 illustrates the usage of iron and aluminum electrodes (as anode) for different electrical fields, under optimal conditions obtained during this study.

Furthermore, consumed electrical energy versus the volume of treated WWTP effluent as a function of applied voltage (v) to the cell has been calculated and presented in Figure 7.

The results indicate that as the electric field grows up, the consumption of anode electrode increases rather linearly, while the energy consumption rises exponentially. Considering the amount of energy consumption and iron and aluminum consumption as anode in the process, the value of EF equals to 2 v/cm has been suggested as the optimum value.

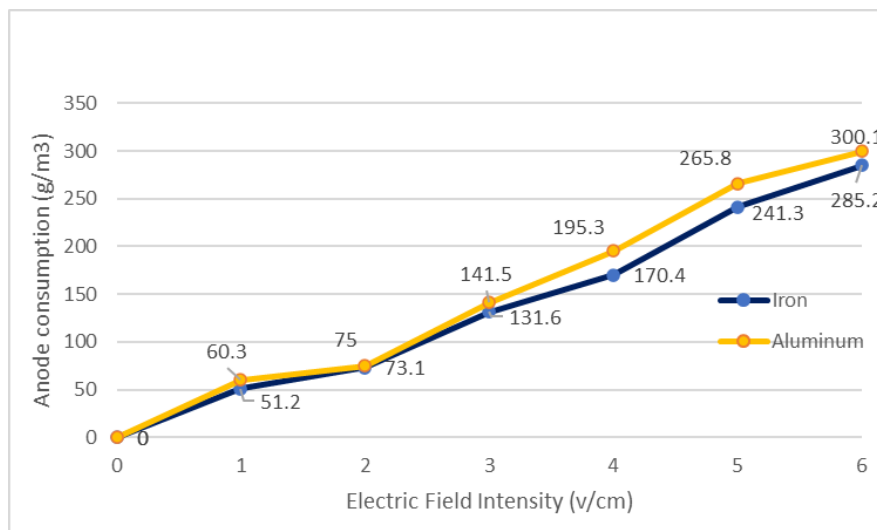


Figure 6: The trend of changes in iron and aluminum consumption in the anode versus increasing the electric field.

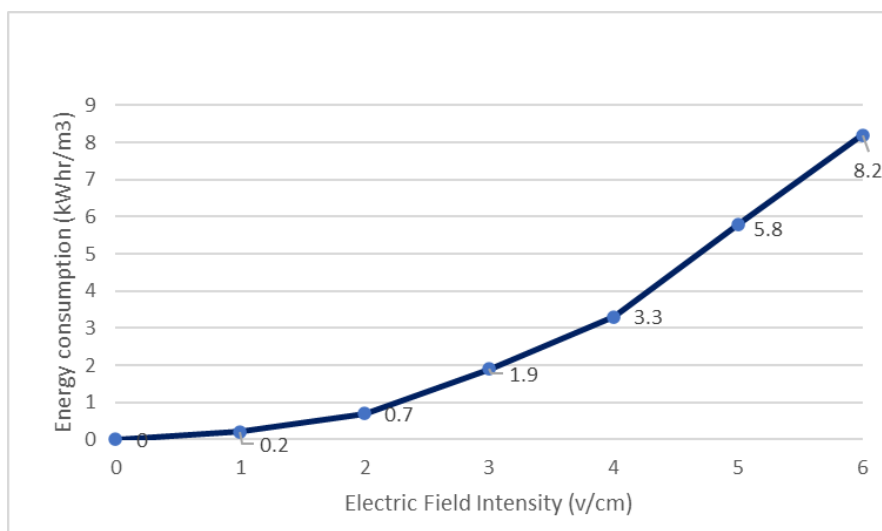


Figure 7: The trend of electricity consumption versus the increase in electric field.

## 2.5 Discussion.

To achieve the optimal state of utilizing the electro-coagulation treatment method on the effluent sample from the industrial wastewater treatment plant in Shokouhieh, the results of experiments with iron and aluminum electrodes were analyzed and evaluated based on the interdependence of operational factors in the efficiency of the electro-coagulation process, such as initial electrolysis time, electrode spacing, pH values and applied electric field. Considering the suitable initial values for these parameters, the optimal values for these parameters were determined. Based on the results obtained, it is shown that the efficiency of this process increases with an increase in electrolysis time and electric field intensity. Due to the decrease in the rate of efficiency increase at higher times, and regardless of the electrodes' material, optimal values for these parameters are suggested to be 105 minutes and 2 v/cm, respectively. Additionally, the results indicate that the pH value for optimal COD removal is equal to 7 for both iron and aluminum electrodes. Nonetheless, the optimal distance between electrodes was found to be dependent on the electrodes' materials, in such a way that the optimal distance between iron electrodes was found to be equal to 5 cm, while for aluminum electrodes, the optimal distance was figured out to be equal to 4 cm.

Regarding the electrolysis time, according to Faraday's law (equation (15)), with a constant current intensity, an increase in electrolysis time results in the production of more iron and aluminum in the reactor environment, leading to an increase in removal efficiency. This explains the initial increase in COD removal efficiency at the beginning of the experiment. The decrease in efficiency at the end of the experiment can be attributed to the removal of a significant portion of pollutants in the effluent within the first 75 minutes of the experiment. Also, due to a decrease in the capacity of iron and aluminum ion loading, remaining pollutants cannot be easily removed.

Furthermore, concerning the electrode spacing, since the space between the anode and cathode is the main location for reactions between pollutants in the effluent and free iron and aluminum hydroxide ions during electrochemical reactions, increasing the distance between the electrodes creates a higher probability of interaction between more pollutant particles in the wastewater and produced hydroxides. Therefore, an increase in electrode spacing from 3 to 5 centimeters leads to an increase in COD separation efficiency. However, increasing the spacing beyond 5 cm for iron electrodes and 4 cm for aluminum electrodes, results in an increase in wastewater volume between the electrodes. Despite providing more contact between wastewater and hydroxide ions by increasing electrode spacing beyond 5 and 4 cm, it ultimately leads to a decrease in removal efficiency due to the increased resistance of wastewater conductivity and capacity of pollutants from hydroxide ions.

Additionally, the results obtained from applying different electric field intensities show that the COD removal rate increases with an increase in electric field intensity. With an increase in electric field intensity and subsequently an increase in current intensity and density, the degradation of iron and aluminum electrodes increases, leading to an increase in coagulation agents' concentration in the wastewater. Therefore, pollutant removal from the wastewater accelerates. Moreover, with an increase in electric field intensity, the production of bubbles increases and at the same time, their diameter decreases. The simultaneous effect of both factors on increasing pollution separation by H<sub>2</sub> bubbles is beneficial. The efficiency removal curves of COD obtained from applying different current fields with a difference of 3 to 5% in COD removal efficiency almost ascend in parallel manner. Considering the decreasing trend in removal efficiency increase with an increase in electric



field intensity and negative impact of increased energy consumption costs on assessing the economic feasibility of employing electrolytic coagulation method, further increasing electric field intensity is not justified.

### III. CONCLUSION

In this study, the removal of COD pollutant from the effluent of an industrial wastewater treatment plant was investigated. It is concluded that the electrolytic coagulation process can be used as a rapid, effective, and flexible method for supplementary treatment of effluents from industrial wastewater treatment plants, depending on consumption needs.

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