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The investigation of COD treatment and energy consumption from urban wastewater by batch electrocoagulation system for small settlements

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ABSTRACT

In this study, electrochemical treatment of urban wastewater with electrical conductivity of 1000 µS/cm, chemical oxygen demand of 250 mg/L was investigated using variables of circulation rate, initial pH value, constant pH value, current intensity and supporting electrolyte type and concentration. Electrocoagulation was used in which aluminum and stainless steel were selected as the electrochemical treatment process. The data obtained in experimental studies show that the COD removal efficiency increases in experiments where the circulation rate is 100 mL/min and the initial pH value is 7. Although the increase in current intensity from 5 A–20 A increases the recovery efficiency from 46%–80%, the COD removal efficiency at a current intensity of 25 A is significantly decreased. While the use of support electrolyte did not contribute to the efficiency of COD removal, it significantly reduced the value of energy consumption. The best removal efficiency according to optimum results was obtained as about 90%, which is 10 A current intensity, 100 mL/min circulation rate, constant pH 7 and no support electrolyte experiments.

Keywords: Urban wastewater; Electrocoagulation; Current intensity; Circulation rate

1. Introduction

Restrictions on water resources and the environmental impacts of dirty wastewater on planetary health are an undeniable worldwide concern today. Water pollution and water recycling is one of the greatest environmental problems of XXI century [1]. In this context, water treatment technologies are emerging as the most direct solution to reduce pollution in water bodies. Central water and wastewater treatment facilities are trying to overcome this environmental problem. Among all water technologies, physico-chemical processes are the most used technologies because they have been known and practiced for hundreds of years to make water available for human needs [2]. However, due to technological development and industrial activity today, pollutants in the water are different in terms of quantity and pollution load over time. Thus, water treatment technologies have become a very important research topic in order to prevent pollution.

Urban wastewater treatment plants generally make it possible to reduce the content of organic matter, nitrogen and phosphorus in urban wastewater. However, there is a serious reduction in water availability due to the increase in water demand and the intensive use of water resources. For this reason, the treatment and reuse of the wastewater is a matter of great interest for the elimination of water stress [3–6].

Electrocoagulation (EC) is an electrochemical technology with a wide range of applications that can effectively reduce the presence of various contaminants, including heavy metal-resistant organic pollutants [7–14].

Electrocoagulation is by now a well-known process and could be a good choice for water treatment because of the following reasons: (1) the amount of required chemicals is much lower, (2) a smaller amount of sludge is produced, (3) no mixing of chemicals is required, (4) coagulant dosing as well required over potentials can be easily calculated and controlled, and (5) operating costs are much lower when compared with most of the conventional technologies [15].

Electrocoagulation consists of an in situ generation of coagulants by an electrical dissolution of iron or aluminum

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electrodes. The metal ions generation takes place at the anode; hydrogen gas is released from the cathode. The hydrogen gas would also help to float the flocculated particles out of the water, and therefore, the process sometimes is named as electroflocculation [16]. Typically, aluminum, iron, carbon, mild steel, graphite and titanium plates are used as electrodes in the electrocoagulation process. Iron and aluminum have been reported to be very effective and successful in pollutant removal at favorable operating conditions. In the case of aluminum, main reactions are as follows:

Anode:

$$Al \rightarrow Al_3{}^{+}{}_{(aq)} + 3e^{-} \quad (1)$$

Cathode:

$$2H_2O_{(l)} + 2e^{-} \rightarrow H_{2(g)} + 2OH^{-}{}_{(aq)} \quad (2)$$

In the solution:

$$Al_3{}^{+}{}_{(aq)} + 3H_2O \rightarrow Al(OH)_3 + 3H^{+}{}_{(aq)} \quad (3)$$

$$nAl(OH)_3 \rightarrow Al_n(OH)_{(3n)} \quad (4)$$

Amorphous $Al(OH)_3$(s) flocks having large surface areas formed in aluminum anode are active in rapid adsorption of soluble organic compounds and trapping of colloidal particles and are easily separated from aqueous medium by sedimentation or H_2 flotation [17,18].

The objective of this study is optimize urban wastewater removal by electrocoagulation. Aluminum were used as anode materials and stainless steel was used as cathode in the monopolar configuration. The effects of circulation rate, initial pH, constant pH, current intensity, time and type and concentration of supporting electrolyte on the COD removal efficiency were investigated.

2. Materials and methods

In the study, domestic wastewaters with weak characteristics were used [19]. Its COD was 250 mg/L while its BOD_5 was 175 mg/L and SS was 80–100 mg/L. Its conductivity was 1000–1050 µS/cm and temperature was 15 ± 3°C. In the intermittently working electrocoagulation system, a tubular reactor with total volume of 500 mL was used. It consisted of a telescopic stainless steel cathode with inner diameter of 60 mm and aluminum anode with outer diameter of 50 mm. A Chroma brand digitally controlled direct-current power supply (62024P–40–120 model 0–40 V, 1–120A) was used to supply the required power. A WTW brand multi-meter was used to adjust pH, conductivity and temperature of the wastewater in the beginning of the reaction and to read these values instantly during the reaction. Total surface area of the electrodes was 1400 cm^2. The distance between the electrodes was 5 mm. In these experiments, effect of wastewater supply rate, supporting electrolyte types like NaCl, Na_2SO_4 and $NaNO_3$, initial pH value of the wastewater and current intensity were examined as wastewater parameters. The experimental assembly is given in Fig. 1.

Experimental conditions and work intervals are shown in the table below (Table 1).

Chemical oxygen demand (COD) analyses were used in examining the parameters having an effect on the process in domestic wastewater treatment through electro-coagulation process. COD analyses were conducted according to the closed system (reflux) method as specified in the standard methods [20]. The following equation was used in calculation of the experimental data:

1. Calculation of treatment efficiency

$$\eta(\%) = \left(\frac{C_0 - C_e}{C_0}\right) \times 100 \quad (5)$$

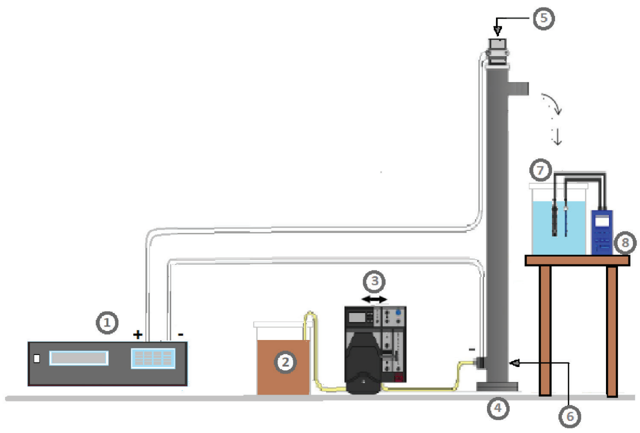

Fig. 1. Experimental setup – 1: Power supply, 2: wastewater inlet, 3: peristaltic pump, 4: reactor, 5: anode electrode (aluminum), 6: katode electrode (stainless steel), 7: wastewater outlet, 8: pH pH control cell.

Table 1
The parameters having an effect on the process in domestic wastewater treatment through electro-coagulation process

Parameters	Parameter intervals	Constant variables
Supporting electrolyte type	NaCl, Na_2SO_4 and $NaNO_3$	pHi ≈ 7.8, T = 15 ± 3°C
Supporting electrolyte concentration (M)	5 – 10 – 25 – 50 mM	pHi ≈ 7.8, T = 15 ± 3°C
Initial pH (pHi)	5 – 6 – 7 – 7.8	*T* = 15 ± 3°C
Current intensity (A)	5 – 10 – 15 – 20	pHi ≈ 7.8, T = 15 ± 3°C

C_0, initial pollutant concentration (mg/L), C_e, the concentration of the pollutant remaining in the wastewater at time t (mg/L).

2. Calculation of energy consumption

$$W\left(\frac{kW-h}{m^3}\right)=\frac{V*I*t}{\vartheta} \quad (6)$$

W: The value of energy consumption (kW-h/m^3), I: Applied current intensity (A), V: potential difference in the system (Volt), t: reaction time (minute), υ: total waste water volume (m^3).

3. Results and discussion

A cylindrical reactor, whose feature are given in Fig. 1, was used in examining the parameters having an effect on treatment of domestic wastewater through the electro-coagulation process with an intermittent system. Because it was impossible to stir content of the column type reactor from inside or outside due to its nature, content of the reactor was constantly circulated with the help of a peristaltic pump to dissolve and distribute Al^{3+}, which was produced during the electrochemical reaction, homogenously inside the reactor during the entire reaction. The purpose was to ensure homogeneity inside the reactor and to minimize warming of the reactor content due to the potential difference to be caused by the applied constant current intensity.

3.1. Effect of circulation rate on COD removal

Accordingly, first, experiments were conducted at constant current intensity of 5 A and natural pH values of the wastewater and circulation rates of 100, 250, 500 and 1000 mL/min. Experiment time was chosen as 60 min and the variables like ambient pH value, temperature and potential difference, which would occur during the electrochemical process, were not intervened. Electric field was ceased at 5th, 10th, 15th, 20th, 30th, 45th and 60th min of the experiments conducted at different circulation rates to collect samples and effluent pH value, temperature, conductivity and potential difference were determined and recorded. The wastewater's initial pH value was around 7.8. This value reached the value of 8.4 after 60 min at the circulation rate of 100 mL/min and 8.5 at the circulation rate of 250 mL/min, 8.7 at the circulation rate of 500 mL/min and 9.3 at the circulation rate of 1000 mL/min respectively. On the other hand, temperature values were measured as 30, 24, 23, 21°C at the circulation rates of 100, 250, 500 and 1000 mL/min respectively. COD removal efficiency and the consumed energy amount are shown in Fig. 2.

Although constant current intensity of 5 A was supplied for each circulation rate, different energy consumption values occurred according to Eq. (6) depending on different potential difference values occurred in the system. It was concluded that the reason of different energy consumption values was essentially caused by high shear forces occurred as a result of rapid fluid transfer affecting electron transfer negatively in the anode and cathode regions depending on the increase in circulation rate. The result caused by this situation was also supported by the literature [21]. Considering circulation rate's COD removal efficiency, the increase in circulation rate reduced COD removal efficiency significantly. Therefore, circulation rate was taken as 100 mL/min in examining the parameters having an effect on COD removal in domestic wastewater through the electrocoagulation system.

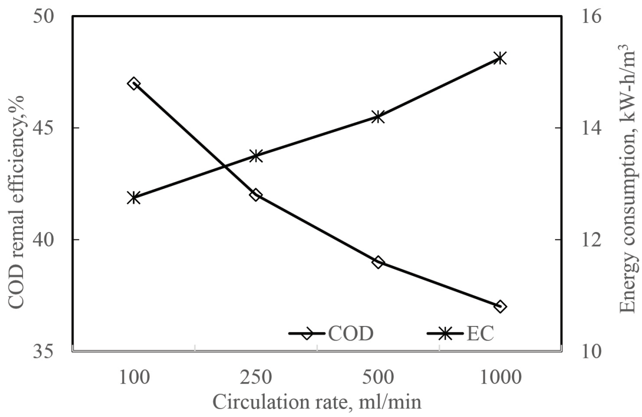

Fig. 2. The effect of circulation rate on COD removal and energy consumption, 5 A current density, initial pH 7.8, temperature 15ºC.

3.2. Effect of the initial pH value of wastewater on COD removal

To examine effect of the initial pH value of wastewater on COD removal efficiency, four different pH values were chosen: 5, 6, 7 and 7.8. pH value of wastewater should be examined as one of the important parameters having an effect on domestic wastewater treatment through the electro-coagulation process because it has an effect on the electrochemically dissolving anode electrode type existing in the reactor. For examining effect of the initial pH value of wastewater on treatment efficiency, studies were conducted with current intensity of 10 A, circulation rate of 100 mL/min in electrolyte-free media. pH value of wastewater was not intervened during the reaction in all experiments. For the experiments conducted in the electrochemical reactor, electric field was ceased to collect samples at 5th, 10th, 15th, 20th and 30th min and effluent pH value, temperature, conductivity and potential difference values, which occurred, were recorded. It was observed in the experiments for each initial pH value that pH values increased during the reaction. At current intensity of 10 A, pH 5 increased to 6.15 at the end of the reaction. Similarly, pH 6 increased to 7.04, pH 7 increased to 8.13 and pH 7.8 increased to 8.95. Furthermore, effluent temperature variations, which occurred during the reaction time as a result of different initial pH values, were also examined. Initial temperature of domestic wastewater was approximately 15°C. This value varied for each different pH value during the reaction time. The reason for variation was associated with the reduction in electrical resistance of the system due to the increase in conductivity of the system depending on the initial pH value. Effluent temperature value of the system was 25°C at the end of the reaction for pH 5 while it was 29 for pH 6. 33 for pH 7 and 39°C for pH 7.8. Energy consumption variations resulted from the initial pH values occurred during the reaction were also examined. The most important factor causing variation of energy consumption under the constant current intensity during the reaction was the change in the applied potential difference values. Because natural pH value of the domestic wastewater was 7.8, HNO_3 was used to obtain reducing initial value. This increased electrical conductivity and consequently, decreased the potential difference and energy consumption values. COD values showing electric energy consumption and treatment efficiency obtained from the experimental data were shown in Fig. 3.

According to Fig. 3, the best removal efficiency was obtained when the initial pH was 7. It is believed that the reason is the fact that, for the same time interval, pH values were obtained below the pH interval suitable for $Al(OH)_3$ at low pH values and above the pH interval suitable for $Al(OH)_3$ at high pH values. Al dissolution diagram showing this exists in previous publications of the authors [22]. This result influenced efficiency of the treatment.

3.3. *Effect of constant initial pH value of wastewater on COD removal*

Experiments were conducted by keeping initial pH values constant during the reaction. In these experiments, current intensity was kept constant at 10 A, circulation rate at 100 mL/min and reaction time as 30 min. Experiments were conducted for pH 5, 6, 7 and natural pH value 7.8 by keeping these values constant. Fig. 4 shows energy consumption and treatment efficiency values resulted from these experiments graphically.

Because concentrated HNO_3 was added continuously to the reactor during the reaction to keep pH constant at constant current intensity, electrical conductivity of the wastewater increased. As seen in Fig. 4, the lowest energy consumption value was obtained at pH 5 at which highest conductivity occurred while the highest energy consumption value was obtained at natural pH value at which the lowest electrical conductivity occurred. The reason for the variation in the treatment efficiency by approximately 26% (86–60) in the experiments in which

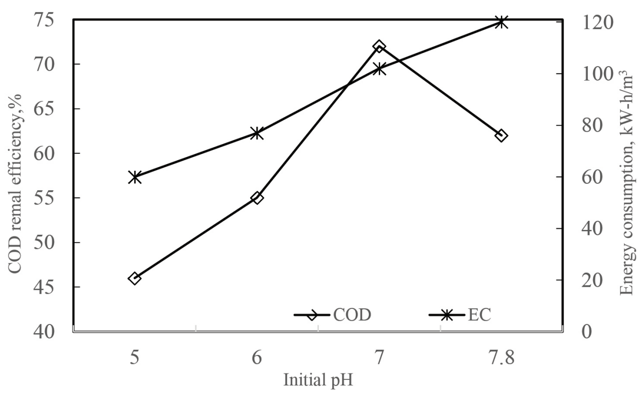

Fig. 3. Effect of the initial pH value of wastewater on COD removal and energy consumption, current density of 10 A, circulation rate of 100 mL/minute.

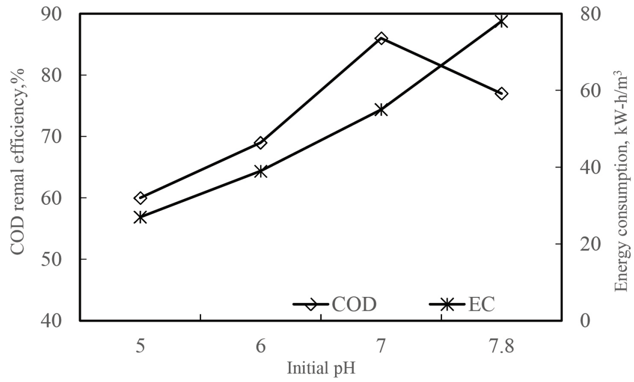

Fig. 4. Effect of the constant initial pH value of wastewater on COD removal and energy consumption, current density of 10 A, circulation rate of 100 mL/minute.

pH value was kept constant during the reaction was the differentiation in Al types depending on ambient pH. The most suitable pH interval for $Al(OH)_3$ formation for the highest flocking and the lowest dissolution is approximately 5.5–8.2 [23]. In experiments where the pH value was kept constant, the rate of formation of $Al(OH)_3$ decreased at the lowest and highest pH values. The decrease in the efficiency of treatment for these pH values may be explained as the decrease in $Al(OH)_3$ formation rate. The fact that the best treatment efficiency was obtained at pH 7 may be explained by higher $Al(OH)_3$ formation rate obtained at this pH value compared to those obtained at other pH values. According to the results from the experiments in which the initial pH value was not kept constant, it was seen that pH value of the wastewater was 7.6 when the efficiency of treatment reached approximately 63% for the 5th min of the experiment that was conducted with pH 7. For the same experiment, when the efficiency of treatment reached approximately 72% at the end of the reaction time of 30 min, effluent's pH value was approximately 8.45. In the experiments in which pH value was continuously kept constant, the treatment efficiency reached approximately 74% at 5th minute at pH 7 at which the best treatment efficiency was obtained and the efficiency reached approximately 86% at the end of the reaction time of 30 min. The fact that the efficiency of treatment values obtained in the experiment in which pH value was kept constant and was not kept constant are different is caused by the different $Al(OH)_3$ formation amounts. Similarly, treatment efficiency values varied depending on ambient pH values while energy consumption values decreased depending on the increase in the wastewater's electrical conductivity due to the addition of concentrated acid to suppress pH values in the experiments in which pH value was kept constant. According to the results obtained from the experiments in which the initial pH values were not kept constant, energy consumption was 102 kW-h/m^3 for pH 7 at the end of the reaction time of 30-minute while energy consumption reduced up to 54 kW-h/m^3 for pH 7 in the constant pH experiments. Keeping the pH value constant during the reaction time resulted in an increase in the treatment efficiency and a decrease in energy consumption.

3.4. Effect of current intensity on COD removal

Experiments with current densities of 5, 10, 15, 20 and 25 A were conducted to examine effect of current intensity on COD removal from domestic wastewater. Wastewater pH value was taken as 5, 6, 7 and natural pH in these experiments. It was seen that the initial pH value of the waster increased during the 5 min reaction time at all current intensities under study. Rapid treatment efficiency was obtained in the first 5 min of the reaction at all current intensities under study and the data from the first 5 min were used because the treatment efficiency rate decreased for the later reactions times. Because the increase in current intensity caused more Al^{3+} ions were dissolved in the anode of the electrochemical reactor and faster H_2 gas release in the cathode region, the effluent's pH value increased [24]. Directly proportional increase in pH value with the increase in current intensity during the reaction prevented reaching the expected values with respect to the treatment efficiency because pH value was drifted away from the pH value at which $Al(OH)_3$ dissolution was lower and its flocking tendency was higher although more coagulants were dissolved. The increase in current intensity increased COD removal efficiency for every initial wastewater pH value under study because it caused more electrochemically dissolving coagulant entered the medium. It should be kept in mind that $Al(OH)_3$ flock formation, which is aimed when aluminum electrode is used in electro-coagulation process, absolutely depends on ambient pH value in the electro-coagulation. According to the data, in the experiments in which the domestic wastewater pH value was 7, effluent pH value reached 7.5 after the 5 min reaction time with current intensity of 5 A while this value reached 7.7 with 10 A, 7.74 with 15 A, 7.85 with 20 A and 8 with 25 A. According to the temperature variation results, the effluent temperature increased for each current intensity value under study. This may be explained as the fact that some of electrical energy supplied to the system converted into heat energy due to the increase in electrical resistance formed in the system depending on the increased current intensity. In the experiments conducted with pH 7, effluent temperature raised to 24.7°C with 5 A 34°C with 10 A, 51°C with 15 A, 62°C with 20 A and 69°C with 25 A. Variation in the treatment efficiency obtained from the experiments in which effect of current intensity on COD removal was examined was shown in Fig. 5 graphically.

According to Fig. 5, when the initial pH value rose to 7 and current intensity rose from 5 A to 20 A, COD removal efficiency increased significantly. However, when current intensity was raised from 20 A to 25 A, the expected improvement in COD removal efficiency did not occur. Furthermore, this caused significant decrease in the efficiency. It is believed that the reason may be very rapid increase in the wastewater temperature depending on the high electrical resistance formed in the system due to the increased current intensity because the domestic wastewater's conductivity was not very high. The increase in the wastewater temperature in high amount prevented $Al(OH)_3$ formation although Al^{3+} ion dissolved in sufficient amount because it affected the flock, which must have formed electrochemically. However, when Fig. 5 is studied carefully, it is seen that COD removal efficiency failed to reach the required increase rate because the increased current intensity raised the increase rate of the wastewater's pH value in the unit reaction time. The difference with respect to the removal efficiency between the lowest pH value (pH 5) and the highest pH value (pH 7) depending on the initial pH value under the current intensity of 5 A is 27% (48–21) while this difference is 17% (77–60) under the current intensity of 20 A. There are two basic reasons for why the expected treatment efficiency cannot be achieved with high current intensities: Ambient pH value increases very rapidly in the unit time with high current intensities and high effluent temperatures due to the increased electrical resistance affect flock formation negatively. The variation in energy consumption values is shown in Fig. 6 graphically.

As seen in Fig. 6, the increased current intensity along with the increased potential difference values applied to the system depending on current intensity caused that energy consumption values increased exponentially in the wastewater having the same electrical conductivity.

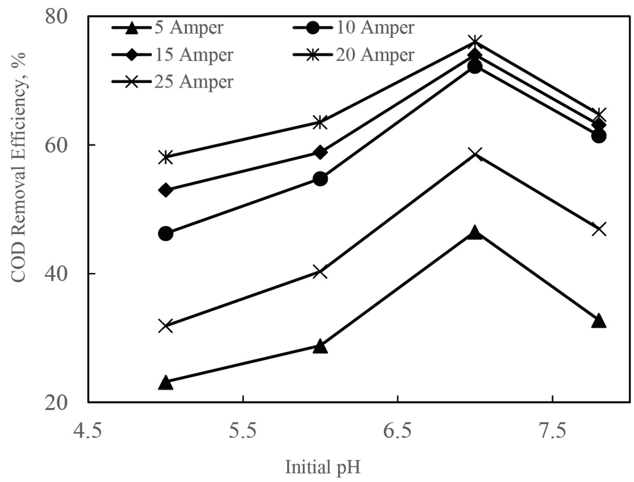

Fig. 5. The effect of current density on COD removal.

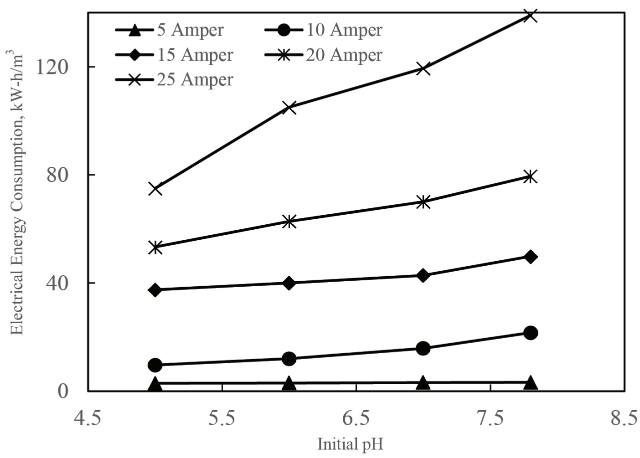

Fig. 6. The effect of current density on energy consumption.

3.5. Effect of supporting electrolyte type and concentration on COD removal

To examine effect of supporting electrolyte type in the studies conducted for raising efficiency of domestic wastewater treatment, effect of supporting electrolyte types like NaCl, Na_2SO_4 and $NaNO_3$ were investigated at concentrations of 5, 10, 25, and 50 mM, at pH 7, at room temperature with circulation rate of 100 mL/min and current intensity of 10 A. The results of COD removal efficiency and electrical energy consumption are given in Table 2. Furthermore, the results obtained from the experiments conducted under the same conditions in which no supporting electrolyte was used are given for comparison with each supporting electrolyte type under study.

COD removal efficiency was found as 73% at the end of the 30-min reaction time in the experiments conducted without using any electrolyte. According to the figures given in Table 2, the use of supporting electrolyte significantly reduced the efficiency of COD removal from domestic wastewater through electro-coagulation. The reduced COD removal efficiency depending on type and concentration of the supporting electrolyte added to wastewater was seen in the studies in the literature also [25]. Energy consumption occurred as 23.8 kW-h/m^3 at the end of the 30 min reaction time when 10 A of current intensity applied to the system without supporting electrolyte in which wastewater pH was 7. Although the use of supporting electrolyte affects efficiency of the treatment, the increased supporting electrolyte concentrations resulted in decreased electrical energy consumption value for each supporting electrolyte type because this reduced the potential difference value applied to the system.

Although the three different supporting electrolyte types under study reduced energy consumption values at different ratios, it was concluded that the use of supporting electrolyte in domestic wastewater treatment through electro-coagulation process is not suitable because they affected COD removal efficiency.

4. Conclusions

In this study, electrochemical treatment of urban wastewater with electrical conductivity of 1000 μS/cm, chemical oxygen demand of 250 mg/L was investigated using variables of circulation rate, initial pH value, constant pH value, current intensity and supporting electrolyte type and concentration. As a result, current intensity of 5 A and the initial pH value of 7.8, which was natural pH value of the wastewater, were kept constant to investigate effect of wastewater circulation rate on COD removal and energy consumption. It was seen at the end of the experiments that the best COD removal efficiency was obtained at the circulation rate of 100 mL/min. It is believed the reason why the increased circulation rate decreased COD removal efficiency was the fact that some of the flocks, which were produced in the electro-coagulation reactor, were broken down in the peristaltic pump at high circulation rates and the increased shear force effect prevented coagulant and contaminant from being combined.

It was understood that the wastewater initial pH value should be 7 as a result of the experiments conducted by using aluminum anode electrode through intermittent electro-coagulation process to investigate effect of wastewater initial pH value on COD removal effect.

COD removal efficiency of 85.5% was obtained as a result of the studies in which effect of constant pH value on COD removal efficiency was investigated in which the highest removal efficiency was achieved when the wastewater initial pH value was 7.

Effect of current intensity on COD removal efficiency was investigated in the electro-coagulation studies conducted for organic matter removal from domestic wastewater and the optimum result was obtained with 20 A.

Effect of supporting electrolyte type on COD removal efficiency was investigated at pH 7 in the situations in which wastewater temperature was not intervened during the reaction by using the supporting electrolyte types of NaCl, $NaNO_3$ and Na_2SO_4. The experiments showed that each electrolyte decreased COD removal efficiency at different ratios depending on the increased concentration. It was found that $NaNO_3$ is the supporting electrolyte least affecting removal efficiency.

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Table 2
Effect of supporting electrolyte type and concentration on COD removal and energy consumption

	COD removal efficiency, %					Electrical energy consumption, kW-h/m^3				
	0	5 mM	10 mM	25 mM	50 mM	0	5 mM	10 mM	25 mM	50 mM
NaCl	73	55.56	49.73	41.74	24.24	23.8	13.18	11.32	8.40	7.00
Na_2SO_4		53.66	46.67	39.02	34.15		19.13	11.78	7.93	6.77
$NaNO_3$		33.33	30.91	25.71	18.24		21.00	13.53	10.03	8.40

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