

## Research Article

# Application of Electrocoagulation Process for Dairy Wastewater Treatment

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Dairy industry wastewater is characterized by high biochemical oxygen demand (BOD<sub>5</sub>), chemical oxygen demand (COD), and other pollution load. The purpose of this study was to investigate the effects of the operating parameters such as applied voltage, number of electrodes, and reaction time on a real dairy wastewater in the electrocoagulation process. For this purpose, aluminum electrodes were used in the presence of potassium chloride as electrolytes. It has been shown that the removal efficiency of COD, BOD<sub>5</sub>, and TSS increased with increasing the applied voltage and the reaction time. The results indicate that electrocoagulation is efficient and able to achieve 98.84% COD removal, 97.95% BOD<sub>5</sub> removal, 97.75% TSS removal, and >99.9% bacterial indicators at 60 V during 60 min. The experiments demonstrated the effectiveness of electrocoagulation techniques for the treatment of dairy wastewaters. Finally, the results demonstrated the technical feasibility of electrocoagulation process using aluminum electrodes as a reliable technique for removal of pollutants from dairy wastewaters.

## 1. Introduction

Food processing industries cause severe environmental problems because of generation of strong wastewater characterized by high biological oxygen demand (BOD<sub>5</sub>) and chemical oxygen demand (COD) [1]. Among the food industries, the dairy industry is the most polluting in terms of volume of effluent generated as well as in terms of its characteristics too, generating about 0.2–10 L of effluent per liter of processed milk [2].

The dairy industry is generally considered to be the largest source of food processing wastewater in many countries. Water is used throughout all steps of the dairy industry, including cleaning, sanitization, heating, cooling, and floor washing; naturally, the industry's need for water is huge [3]. In general, wastes from the dairy processing industry contain a high concentration of organic material such as proteins, carbohydrates and lipids, high BOD<sub>5</sub> and COD, and high concentrations of suspended solids and suspended oil grease. The BOD<sub>5</sub> concentrations for dairy factory wastewaters can

vary widely depending on the season or product cycle. For instance, it has been reported that cheese factories have BOD<sub>5</sub> ranging from 588 to 5000 mg/L, whereas factories producing cream have BOD<sub>5</sub> in the range of 1200–4000 mg/L [4].

All of these require specialized treatments to prevent or minimize environmental problems. Dairy wastewaters (DWs) are also characterized by wide fluctuations in flow rates, related to discontinuity in the production cycles of different products [5, 6]. Therefore, dairy wastewater treatment is very important from environment and water requirement point of view for dairy industry.

Dairy wastewaters are generally treated usually using biological methods such as activated sludge process, aerated lagoons, aerobic bioreactor, trickling filters, sequencing batch reactor (SBR), upflow anaerobic sludge blanket (UASB) reactor, upflow anaerobic filters, and biocoagulation, [4, 7–10]. Aerobic biological processes are high energy intensive, whereas anaerobic treatment of dairy wastewater reflects very poor nutrient removal, and effluents treated by anaerobic biological processes need additional treatment [6, 11]. On

TABLE 1: Characteristics of the raw dairy wastewater used for this study.

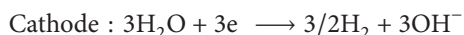
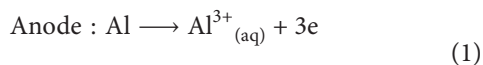
Parameters	Raw wastewater Mean $\pm$ S.D.	12 h settled wastewater Mean $\pm$ S.D.	Permissive levels (Iran standard)
Number of samples	24	24	—
Total COD (mg/L)	7855.25 $\pm$ 703.05	6114.25 $\pm$ 74.52	60
Total BOD <sub>5</sub> (mg/L)	3486.3 $\pm$ 235.17	2919.3 $\pm$ 45.27	30
Total suspended solids (mg/L)	1724.17 $\pm$ 149.14	734.43 $\pm$ 22.13	60
Total coliforms (TCs)	4.39 $\times 10^6 \pm 2.5 \times 10^5$	3.53 $\times 10^6 \pm 1.1 \times 10^5$	1000
Fecal coliforms (FCs)	3.27 $\times 10^6 \pm 1.3 \times 10^5$	2.75 $\times 10^6 \pm 7.2 \times 10^4$	400
Conductivity ( $\mu$ S/cm)	8010 $\pm$ 1300	8073.67 $\pm$ 59.53	—
pH	7.65 $\pm$ 0.02	7.24 $\pm$ 0.07	6.5–8.5

the other hand, the physical/chemical methods that have been proven to be successful are coagulation/flocculation [12, 13].

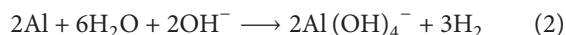
In the recent years, investigations have been focused on the treatment of wastewaters using electrocoagulation (EC) because of the increase in environmental restrictions on effluent wastewater. Electrocoagulation (EC) is an electrochemical method for treating polluted water which has been successfully applied not only for the treatment of soluble or colloidal pollutants, such as slaughterhouse wastewater [14], vegetable oil refinery [15], dairy industry wastewater [16, 17], slaughterhouse wastewater [18], nitrate-bearing wastewater [19], wastewaters containing heavy metals, and pesticides and phenolic compounds [20–25], but also drinking water for fluoride and humic acid removal [26–28].

Electrocoagulation involves the generation of coagulants in situ by dissolving electrically either aluminium or iron ions from aluminum or iron electrodes, respectively. The metal ions generation takes place at the anode, and hydrogen gas is released from the cathode. The hydrogen gas would also help to float the flocculated particles out of the water. The electrodes can be arranged in a monopolar or bipolar mode. The materials can be aluminium or iron in plate form or packed form of scraps such as steel turnings and millings [29, 30].

An examination of the chemical reactions occurring in the electrocoagulation process shows that the main reactions occurring at the electrodes (aluminum electrodes) are



The cathode may also be chemically attacked by  $\text{OH}^-$  ions generated during  $\text{H}_2$  evolution at high pH [31]:



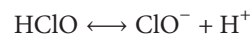
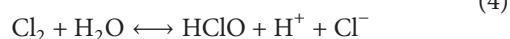
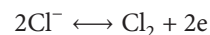
$\text{Al}^{3+}_{(\text{aq})}$  and  $\text{OH}^-$  ions generated by electrode reactions (1) react to form various monomeric species such as  $\text{Al}(\text{OH})^{2+}$ ,  $\text{Al}(\text{OH})_2^+$ ,  $\text{Al}_2(\text{OH})_2^{4+}$ , and  $\text{Al}(\text{OH})_4^-$  and polymeric species such as  $\text{Al}_6(\text{OH})_{15}^{3+}$ ,  $\text{Al}_7(\text{OH})_{17}^{4+}$ ,  $\text{Al}_8(\text{OH})_{20}^{4+}$ , and  $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$ ,  $\text{Al}_{13}(\text{OH})_{34}^{5+}$ , which transform finally into  $\text{Al}(\text{OH})_3$  according to complex precipitation kinetics [32].

Freshly formed amorphous  $\text{Al}(\text{OH})_3$  “sweep flocs” have large surface areas which are beneficial for a rapid adsorption of soluble organic compounds and trapping of colloidal particles. These flocs polymerize as



and they are easily removed from aqueous medium by sedimentation and by  $\text{H}_2$  flotation. On the other hand, the aluminum hydroxide flocs normally act as adsorbents and/or traps for pollutants. Therefore, they would eliminate them from the solution [21, 30].

Secondary anodic reactions occur also during electrocoagulation process; for example, in neutral and acidic chloride solutions, native and free chlorine and hypochlorite are formed which are strong oxidants [33, 34].



In this study, real dairy industry wastewater has been treated using aluminum electrodes in a batch reactor. This study elucidates the effects of parameters such as applied voltage, reaction time, and number of electrodes on the BOD<sub>5</sub> and COD removal efficiencies. Information regarding the electrical energy consumption and electrode consumption are also included to provide an estimation of the cost of pollutants removal by an electrocoagulation system.

## 2. Experimental

**2.1. Dairy Wastewater.** The wastewater used in this work was taken from the local dairy factory in Iran (Sistan and Baluchestan province), with 25000 (mean value) kg milk per day processing capacity. Although the quality of the effluent was variable, it was colored milk, and typical characteristics of the raw dairy wastewater are presented in Table 1. Samples were collected in polypropylene bottles, shipped cold and kept at 4°C before use. The length of the storage before

starting experiments varied from one day to six weeks. The effluent has been sampled at different times during this study, and the initial characteristics varied with time (Table 1). This effluent initially contained high concentrations of soluble and suspended materials.

**2.2. Setup and Procedure.** In each run, wastewater (supernatant) after 12 h settling time was poured into the electrocoagulation cell. All experiments were performed in a bipolar batch reactor (Figure 1), with six aluminum electrode connected in parallel. Only the outer electrodes were connected to the power source, and anodic and cathodic reactions occurred on each surface of the inner electrode when the current passed through the electrodes. The internal size of the cell was  $15\text{ cm} \times 15\text{ cm} \times 25\text{ cm}$  with an effective volume of  $2000\text{ cm}^3$ . The volume ( $V$ ) of the solution of each batch was 2 L. The active area of each electrode (plate) was  $14\text{ cm} \times 20\text{ cm}$  (submerged area) with a total area of  $280\text{ cm}^2$ . The distance between electrodes was 2 cm. A power supply having an input of 220 V and variable output of 0–60 V (10, 20, 30, 40, 50, and 60 V) with maximum current of 5 ampere was used as direct current source. The temperature of each system was maintained at  $25 \pm 2^\circ\text{C}$ . Different samples of 100–150 mL were taken at 15 min intervals for up to 1 h and filtered before being analysed to determine  $\text{BOD}_5$ , COD, TSS, total coliforms, fecal coliforms, and other parameters. During the runs, the reactor unit was stirred at 150 rpm by a magnetic stirrer to allow the chemical precipitate to grow large enough for removal. During electrocoagulation, an oxide film formed at the anode. In order to overcome electrode passivation at the anode, the electrodes were rinsed in diluted HCl solution (5% v/v) after each experiment and rinsed again with tap water and finally weighted. Also, the electrodes were reweighted to calculate sacrificial electrode consumptions. These weights are used in the calculations of the total operating cost. In addition, the electrical energy consumed per unit volume of treated wastewater has been calculated for different experimental conditions. All analyses were conducted in duplicate for reproducibility of the experimental results, and all of the data in the figures and tables were the average ones.

**2.3. Analytica.** COD,  $\text{BOD}_5$ , total suspended solids (TSSs), conductivity, total and fecal coliforms, and pH determinations were determined according to the standard methods (APHA, 2005). COD was measured using COD reactor and direct reading spectrophotometer (DR/5000, HACH, USA). Five-day biological oxygen demand ( $\text{BOD}_5$ ) was determined by the manometric method with a respirometer OxiTop system (WTW). The pH and conductivity were adjusted to a desirable value using NaOH or  $\text{H}_2\text{SO}_4$  and NaCl and measured using a pH meter model UB-10 (Ultra Basic, US) and a conductivity meter model Cond 3110, respectively.

### 3. Results and Discussion

The effects of parameters such as applied voltage and reaction time have been evaluated under specific conditions for a constant temperature.

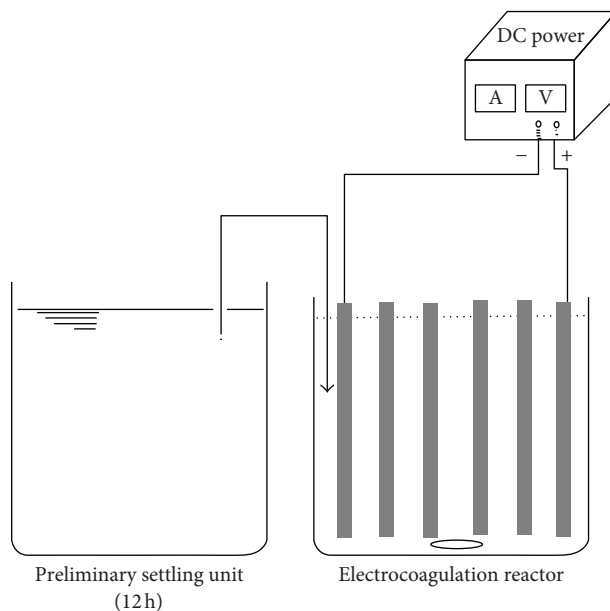


FIGURE 1: Experimental setup of electrocoagulation unit.

**3.1. Wastewater Characterization.** Table 1 presents the real dairy wastewater characteristics prior to any treatment, after 12 h settling time and the guidelines from Iran for effluent discharge in the sewage urban works. The values of the pollution parameters were lowered after 12 h of preliminary settling time. Also, the comparison of these values showed that the COD,  $\text{BOD}_5$ , and the concentration of TSS, were much greater than those recommended by Iran. Consequently, the dairy effluent needed to be treated before discharge.

**3.2. Effect of Preliminary Settling Time.** Preliminary settling process is a natural treatment method that requires no chemical addition. Although some workers realized the importance of the natural settling process, there is little information available in the literature on the effect of the preliminary settling time on pollutants removal capacity.

In this study, the raw dairy wastewater was allowed to settle in a preliminary settling tank before the electrocoagulation process. The process had an effect on  $\text{BOD}_5$ , COD, TSS, and total and fecal coliforms removals on the first 12 h. COD was reduced from  $7855.25 \pm 703.05$  to  $6114.25 \pm 74.52\text{ mg/L}$  (resulting in more than 22% COD removal efficiency), whereas  $\text{BOD}_5$  was reduced in the wastewater from  $3486.3 \pm 235.17$  to  $2919.3 \pm 45.27\text{ mg/L}$  (resulting in 16%-17%  $\text{BOD}_5$  removal efficiency). Furthermore, TSS concentration was reduced from  $1724.17 \pm 149.14$  to  $734.43 \pm 22.13\text{ mg/L}$  (resulting in more than 57% TSS removal efficiency). Similar results were reported by Amuda and Alade [35].

Also, data revealed that the effluent of the settling unit is characterized by high load of organic matter. The ratio  $\text{BOD}_5/\text{COD}$  of approximately 0.48 indicates that 50% of the COD of this wastewater is easily able to be degraded by biological treatment. Nevertheless, the remainder COD is high,

TABLE 2: Influence of electrocoagulation process using aluminum electrodes on dairy wastewater quality parameters (mean values).

Applied voltage (V)	Quality parameters of treated effluent after electrocoagulation unit				
	COD (mg/L)	BOD <sub>5</sub> (mg/L)	TSS (mg/L)	TC (MPN/100 mL)	FC (MPN/100 mL)
10	2405.96	892.2	372.43	1115480	476850
20	1390.99	484.47	298.18	209329	89925
30	1143.98	432.55	167.89	59340	38500
40	681.74	379.35	131.76	57892	38225
50	649.94	310.27	26.73	49420	18975
60	70.92	43.45	16.52	361	28

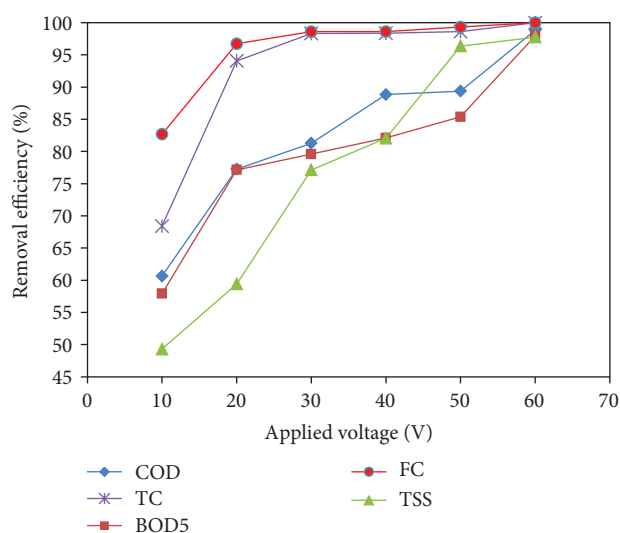


FIGURE 2: Effect of applied voltage on pollutants removal from dairy wastewater (reaction time = 60 min).

which indicates the necessity of an efficient physicochemical treatment for this wastewater.

**3.3. Effect of Electrocoagulation Process.** Electrocoagulation processes a direct current source between metal electrodes immersed in wastewater. The electrical current causes the dissolution of metal electrodes commonly iron and aluminum into wastewater. The dissolved metal ions, at an appropriate pH, can form wide ranges of coagulated species and metal hydroxides that destabilize and aggregate the suspended particles or precipitate and adsorb dissolved contaminants [29, 36].

The effect of applied voltage and reaction time on electrocoagulation process of real dairy wastewater treatment were determined. The results of effect of operating parameters on pilot scale electrocoagulation process are shown in Table 2 and Figure 2. The applied voltage is expected to exhibit a strong effect on electrocoagulation process, especially on the BOD<sub>5</sub> abatement and COD removal: the higher the voltage, the shorter the treatment. The supply of voltage to the electrocoagulation system determines the amount of Al<sup>3+</sup>

ion released from the respective electrodes and the quantity of resulting coagulant. Thus, more Al<sup>3+</sup> ions get dissolved into the solution, and the formation rate of Al(OH)<sub>3</sub> is increased. Also, it is well known that electrical potential not only determines the coagulant dosage rate but also the bubble production rate and size and the flocs growth which can influence the treatment efficiency of the electrocoagulation process [37, 38].

Figure 2 and Table 2 illustrate the effect of applied voltage on pollutants removal efficiency at pH of 7.24. Removal efficiencies of more than 97% were achieved after 60 min electrocoagulation for all voltages (10–60 V). Also, it is clear that by increasing electrical potential from 10 to 60 V, the pollutants concentration in the effluent decreased significantly. As an example, COD concentration has decreased from 6114.25 to 2405.96 mg/L (approximately 60.6% COD removal efficiency) after electrocoagulation process with electrical potential of 10 V. Again, by increasing electrical potential to 60 V, the COD concentration in the effluent decreased to 70.9 mg/L in 60 min (approximately 98.8% COD removal efficiency). Also, BOD<sub>5</sub> concentration has reduced from 2919.3 to 892.2 mg/L (nearly 57.9% BOD<sub>5</sub> removal efficiency) after electrocoagulation process with electrical potential of 10 V, and by increasing electrical potential to 60 V, the BOD<sub>5</sub> concentration in the effluent declined to 43.45 mg/L in 60 min (around 98% BOD<sub>5</sub> removal efficiency). The same trend was observed for TSS concentration in wastewater after electrocoagulation process, so the removal efficiency increased from 49.29% at electrical potential of 10 V to 97.75% at electrical potential to 60 V. Furthermore, as can be seen from Figure 2 and Table 2, the removal efficiency of bacterial indicators (TC and FC) is very high, and efficiency was raised with increase in applied voltage from 10 to 60 V. Maximum removal efficiency (>99.9%) was obtained in applied voltage 60 V, and thus the effluent quality was reached to permissive levels (lower than 1000 and 400 for TC and FC, resp.), and hence from the viewpoint of microbial aspects, discharge of this effluent to environment is safe. Also, minimum removal efficiency occurred in the lowest electrical potential (10 V). This is ascribed to the fact that at high voltage, the amount of oxidized aluminum increased, resulting in a greater amount of precipitate for the removal of pollutants. In addition, it was demonstrated that bubbles density increases and their size decreases with increasing



current density [39], resulting in a greater upwards flux and a faster removal of pollutants and sludge flotation. In fact, the most important mechanisms for removal of pollutants in this process are rapid adsorption of soluble organic compounds and trapping of colloidal particles in “sweep flocs” ( $\text{Al}(\text{OH})_3$ ). The results of this study are in agreement with the results of researches done by Nouri et al. [24] on zinc and copper removal from aqueous solutions by aluminum electrodes, Bazrafshan et al. [22] on the efficiency of EC with iron and aluminum electrodes in removal of chromium, Mansoorian et al. [40] on nickel removal from aqueous solutions by EC process with iron-rod electrodes, Njiki et al. [41] on mercury removal from water by EC process with aluminum and iron electrodes, Bazrafshan et al. [28] on humic acid removal from underground waters by EC process using iron electrodes, Bazrafshan et al. [25] on the efficiency of EC with aluminum and iron electrodes in removal of phenol, and also Bazrafshan et al. [27] on the efficiency of EC with aluminum and iron electrodes in removal of fluoride from aqueous solutions. Nevertheless, Bayar et al. [42] reported that increase in the current density does not cause an expected removal efficiency increase; on the contrary, it can cause a relatively negative effect on it. Also, a similar trend was seen in the study of Holt et al. [38].

Figures 3, 4, 5, 6, and 7 represent the effect of reaction time on pollutants removal from dairy wastewater at various applied voltages. Theoretically, based on Faraday's law, duration of electrolysis affects the amount of released electrode in a system with aluminum electrodes and determines the amount of produced  $\text{Al}^{3+}$  from this electrodes [43]. EC process includes two steps, destabilization and accumulation. The first step is usually short, and the second one is relatively long. With increase in reaction time, both energy and electrode consumption increase and this shows that reaction time is a very important parameter due to affecting the cost effectiveness of EC process in polluted waters [44].

In the present study, as it can be seen from Figures 3–7, more than 50% of wastewater quality parameters were removed in the first minutes (only 15 min), especially for applied voltages 30, 40, 50, and 60 V, while in later minutes, the percentage of removal was low. Also, more than 50% removal efficiency of COD and  $\text{BOD}_5$  was obtained at all applied voltages at 15 min (Figures 3 and 4). Furthermore, as the time of electrolysis was increased, comparable increases in the pollutants removal rate were observed for all applied voltages. On the other hand, for a given time, the removal efficiency increases significantly with an increase in applied voltages. As an example, after 60 min of electrolysis (Figure 3), it can be seen that 60.65%, 77.25%, 81.29%, 88.85%, 89.37%, and 98.84% of COD were removed for applied voltages of 10, 20, 30, 40, 50, and 60 V, respectively. Also, as the applied voltage was increased, the required time for the electrocoagulation process decreased. This was ascribed to the fact that at high current, the amount of oxidized metal increased, resulting in a greater amount of precipitate for the removal of pollutants. Similar trends were observed for other quality parameters of dairy wastewater.

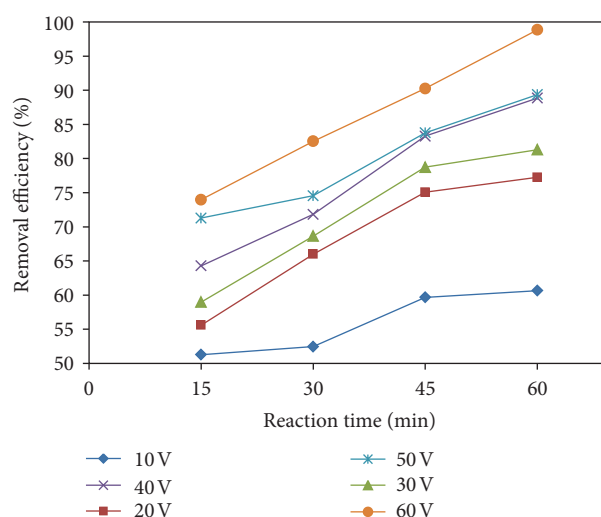


FIGURE 3: Effect of reaction time on COD removal from dairy wastewater at various applied voltages.

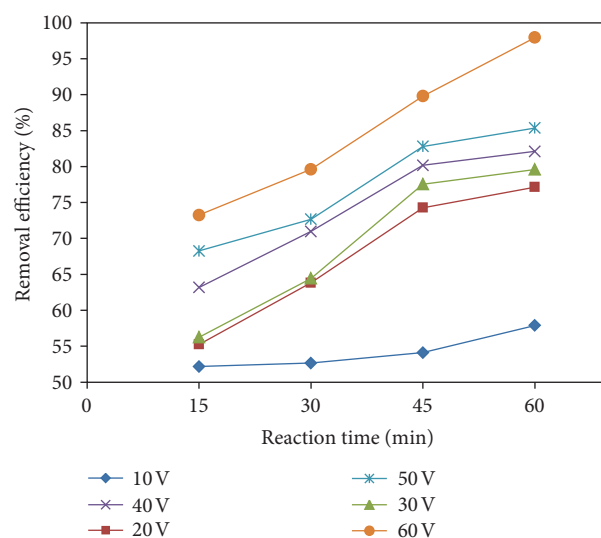


FIGURE 4: Effect of reaction time on  $\text{BOD}_5$  removal from dairy wastewater at various applied voltages.

The results of this study is in accordance with the results of researches done by Wan et al. [45] on removal of arsenic from water by using EC, Ugurlu et al. [46] on the removal of lignin and phenol from paper mill effluents by EC, and also Merzouk et al. [47] on removal of turbidity and separation of heavy metals using electrocoagulation-electroflotation technique.

**3.3.1. Electrical Energy and Electrode Consumption.** Electrical energy consumption is a very important economical parameter in the electrocoagulation process. Therefore, for the same operating conditions, after 60 min of electrocoagulation, consumption of energy and aluminum electrode were determined, that is, represented in Table 3. It can be understood from Table 3 that electrical energy and electrode

TABLE 3: Electrical energy and electrode consumption during electrocoagulation process (reaction time = 60 min).

	Applied voltage V					
	10	20	30	40	50	60
Energy consumption (Kwh/L)	0.012	0.027	0.074	0.080	0.089	0.095
Electrode consumption (g)	0.249	0.585	1.841	1.457	2.312	2.672

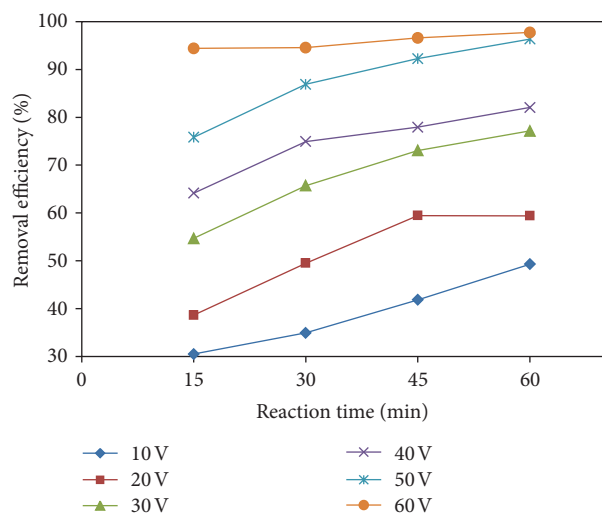


FIGURE 5: Effect of reaction time on TSS removal from dairy wastewater at various applied voltages.

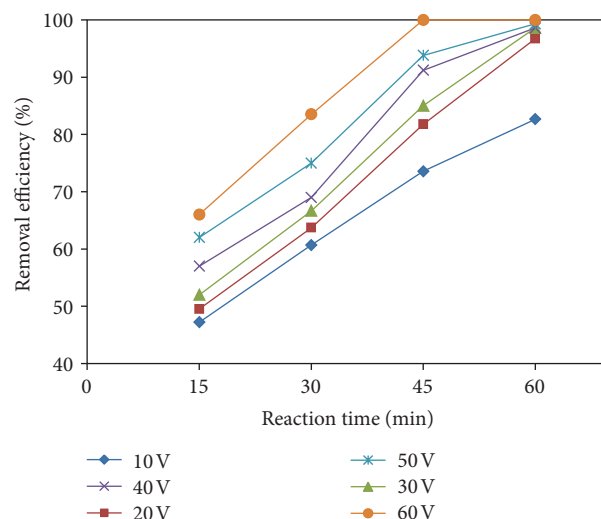


FIGURE 7: Effect of reaction time on FC removal from dairy wastewater at various applied voltages.

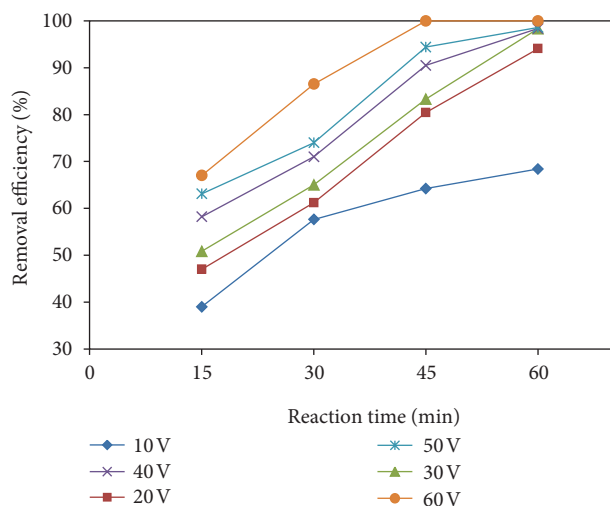


FIGURE 6: Effect of reaction time on TC removal from dairy wastewater at various applied voltages.

consumption were found to increase with increasing the applied voltage as would be expected in any other electrolytic process. An increase in applied voltage from 10 to 60 V causes an increase in energy consumption from 0.012 to 0.095 kWh/L. Similar trends were seen in the study of Bayar et al. on Poultry slaughterhouse wastewater treatment [42] and Bazrafshan et al. on carwash wastewater treatment by electrocoagulation process [48].

Also, as it is presented in this table, electrode consumption varied between 0.249 and 2.672 g/L. It can be concluded that the higher the voltage of the system applied, the higher the weight of the electrode consumed in the process. On the other hand, an increase in the applied voltage causes a proportional increase of the consumption of electrode by the system. As the table represents, the electrode consumed at 60 V in the process is much more than that of the process conducted with 10 V. This result is in agreement with the findings of Bazrafshan et al. on slaughterhouse wastewater treatment by electrocoagulation process [18].

#### 4. Conclusion

Batch electrocoagulation studies were performed to evaluate the influence of various experimental parameters such as applied voltage and electrolysis time on the removal of pollutants from dairy wastewater. The results of this study have shown the applicability of electrocoagulation in the treatment of real dairy industry wastewater. The treatment rate was shown to increase upon increasing the applied voltage and reaction time. However, increasing the applied voltage caused the energy consumption to increase. Indeed, the highest voltage produced the quickest treatment with an effective reduction of COD, BOD<sub>5</sub>, and bacterial indicators (TC and FC) concentration. Consequently, it can be inferred that electrocoagulation is a comparatively suitable process for removal of COD, BOD<sub>5</sub>, and other pollutants

using aluminum electrodes to effectively treat dairy industry wastewater.

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