



Operating parameters and costs assessments of a real dyehouse wastewater effluent treated by a continuous electrocoagulation process



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ABSTRACT

Treatment of a real dyehouse wastewater was studied with continuous flow electrocoagulation (CEC) process. Removal efficiencies of COD, TOC and turbidity were obtained as 85%, 76% and 95% for Fe electrode and 77%, 72% and 95% for Al electrode at current density of 65 A/m², inlet flow rate of 0.010 L/min, operating time of 80 min (hydraulic retention time of 350 min), and current density of 65 A/m² (the optimum operating conditions). The decrease in the inlet flow rate (0.20–0.010 L/min) led to an increase in removal efficiencies of COD (58–85% for Fe, 55–77% for Al), TOC (51–76% for Fe and 46–72% for Al) and turbidity (70–95% for Fe and 72–95% for Al). Operating costs for Fe and Al electrodes at the optimum operating conditions were calculated as 1.562 \$/m³ or 7.282 \$/kg COD for Fe electrode and 1.851 \$/m³ or 14.257 \$/kg COD for Al electrode. The results presented in this study revealed that the CEC process can be effectively used for the removals of color, COD, TOC and turbidity from textile wastewater.

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1. Introduction

The textile industry utilizes about 10,000 dyes and pigments, and a high amount of these dyes can exist in effluents of dyeing processes [1]. Reactive dyes are very important class of textile dyes, whose losses through processing are particularly significant and difficult to treat. Under typical reactive dyeing conditions (pH: >10, temperature: >65 °C and salt: 60–100 g/L) as much as 20–50% of the initial mass of the reactive dye remains in the spent reactive dye bath in the hydrolyzed form which has no affinity for the fiber [2]. The dyeing and finishing are the two important processes generally applied in most of the textile manufacturing industries. In textile industries, substantial amounts of unfixed dyes are released into wastewater because of the low efficiency of dye fixing on textile fibers (60–90%) [3,4]. The direct discharge of textile wastewater into the water resources pollutes the water and affects the aquatic life (aquatic plants, microorganisms, fish and mammals). The direct discharge of this industrial effluent into sewage networks produces also disturbance in biological treatment processes. Textile wastewater is characterized by strong colors, high chemical oxygen demand (COD) and total organic

carbon (TOC), low biodegradability and high-salt content [5,6]. There are many processes such as biodegradation, adsorption, precipitation, membrane filtration, chemical degradation, photodegradation and chemical coagulation to remove dyes from colored effluents [7–9]. However, these processes are quite expensive and involve several operational problems. For these reasons, there has been an increasing interest in the use of electrochemical methods [10]. Electrochemical methods have advantages due to no requirement of chemicals before and after treatment, producing of less sludge, requirement of small area and low investment cost. [11]. Moreover, the high electrolyte (e.g. NaCl, Na₂CO₃, Na₂SO₄ inorganic salts) concentration used in the textile dyeing process offers an inherent advantage for treating dyeing dyebath effluent with electrocoagulation (EC) [12].

In recent years, EC has been applied successfully to treat textile dyes containing solutions or wastewater samples [10–23]. However, most of these studies have involved with treatment of aqueous synthetic dye solutions. EC treatments of these samples have been conducted on a laboratory scale in batch and continuous EC reactors showing almost removal efficiencies of 85–100% for color and 40–80% for COD at various operating conditions. These processes are found to be very efficient in color removal with low-energy consumption. Treatment of textile wastewater by continuous EC process (CEC) has been less investigated for especially

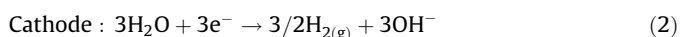
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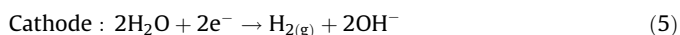
textile dyebath effluents [24–29]. Despite few scientific studies on treatment of actual textile wastewater by the CEC process using iron (Fe) and aluminium (Al) anodes, there have not been publications reported yet about technical and economical evaluation for treatment of actual exhausted textile dyebath effluents in the CEC process.

EC is an electrolytic process consisting of dissolution of sacrificial anodes (Fe or Al) upon application of a current between two electrodes for treatment of liquid wastewater containing inorganic or organic pollutants [29–39]. In the EC, the anodic reaction involves the dissolution of Al or Fe electrodes (Eqs. (1)–(3)), and the cathodic reaction (Eqs. (2) and (5)) involves the formation of hydrogen gas and hydroxide ions, then hydroxide ions formed at the cathode increase pH of the wastewater thereby inducing precipitation of metal ions as corresponding hydroxides and co-precipitation with iron hydroxides. The main anode and cathode reactions occurring at Al and Fe electrodes in EC process are as follows:

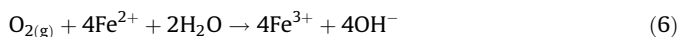
Anode and cathodes reactions for Al electrodes:



Anode and cathodes reactions for Fe electrodes:



In the case of Fe electrodes, dissolved iron (i.e., Fe^{2+} or Fe^{3+}) hydrolyses by increasing pH to form precipitates as $\text{Fe}(\text{OH})_{n(\text{s})}$. The rate of the oxidation of Fe^{2+} depends on the availability of dissolved oxygen [28].



In the case of used aluminium electrodes in EC process, $\text{Al}^{3+}/\text{Fe}^{3+}$ and OH^- ions generated by anode and cathode Reactions (1) and (2) to form various monomeric species depending on the pH of the aqueous medium. Consequently, the removal mechanism of pollutants from wastewater with both electrodes was related to forming of $\text{Fe}(\text{OH})_{3(\text{s})}$, $\text{Al}(\text{OH})_{3(\text{s})}$, monomeric and polymeric iron and aluminium species due to coagulation, precipitation, co-precipitation, and electrooxidation [11,28].

The objective of the present study was to investigate the influence of different operating parameters (current density of 20–85 A/m², operating time of 0–80 min, inlet flow rate of

0.01–0.20 L/min and COD or TOC concentration) on COD, TOC and turbidity removal efficiencies in the real dyehouse wastewater by the CEC process using sacrificial Fe and Al electrodes. Operating costs were also calculated in terms of energy and electrode consumptions.

2. Material and methods

2.1. Characterization of textile dyehouse wastewater

The dyehouse wastewater used in this study was obtained from a textile factory producing approximately 1000 m³ of wastewater per day in Istanbul, Turkey. The textile wastewater contained real dyehouse effluents from both dyeing and rinsing baths of cotton, rayon, silk, wool and nylon. Reactive, dispersive and acidic dyes are the major types of dye used in this textile factory. The wastewater was collected from balancing (equalization) tank immediately after the dyeing and rinsing processes, and then directly stored at below 4 °C in a dark condition to avoid any change in physico-chemical characteristics before use. The relatively large suspended particles in the colloidal ranges were removed from the wastewater before conducting the experiments. Characterizations of the textile dyehouse wastewater from the balancing tank are shown in Table 1.

2.2. Continuous EC reactor and experimental procedure

The EC experiments were carried out in a continuous flow reactor made from Plexiglas with dimensions of 250 mm × 190 mm × 80 mm. A schematic of the experimental setup for the CEC reactor is shown in Fig. 1. Two anodes and two cathodes (Fe or Al) of rectangle electrodes with dimensions of 220 mm × 50 mm × 4 mm were placed in the reactor with monopolar parallel connection mode. The space between each electrode was 20 mm and their position within the reactor was in perpendicular order according to the wastewater flow. The total effective electrode area was 660 cm². The electrodes were connected to a digital dc power supply (Agilent 6675A model) operated at galvanostatic mode. The current was adjusted on the dc power supply. The initial pH of wastewater was adjusted to 5.5, and then the wastewater was fed into the reactor from the bottom using a peristaltic pump (Cole-Parmer 7553-75 model). Volume of the dyehouse wastewater in the CEC reactor was 3.5 L. The CEC experiments for Al or Fe electrodes were run up to 80 min depending on variations of the operating time and other operating parameters. Flow rate of the textile wastewater was varied at 0.010, 0.050, 0.10 and 0.20 L/min. Effluent samples from the reactor were taken at different times during the experiment and the experiment was continued until steady-state concentrations were achieved. Effluent from the reactor was passed through a filter column with diameter of 30 mm filled with 10 mm glass beads to remove suspended

Table 1
The characterizations of the textile dyehouse wastewater.

Parameters	Before the CEC process	Average values before the CEC	After the CEC process	
			Fe	Al
pH	6.5–7.1	6.8	8.4	8.2
COD (mg/L)	1940–2060	2000	300	460
TOC (mg/L)	520–450	485	116	136
Turbidity (NTU)	1850–2600	2225	111	111
Conductivity (mS/cm)	2.25–2.35	2.30	1.85	1.96
Total SS (mg/L)	210–250	230	10	15
Temperature (°C)	18–22	20	26	27
Fe (mg/L)	–	–	0.014	–
Al (mg/L)	–	–	–	0.018

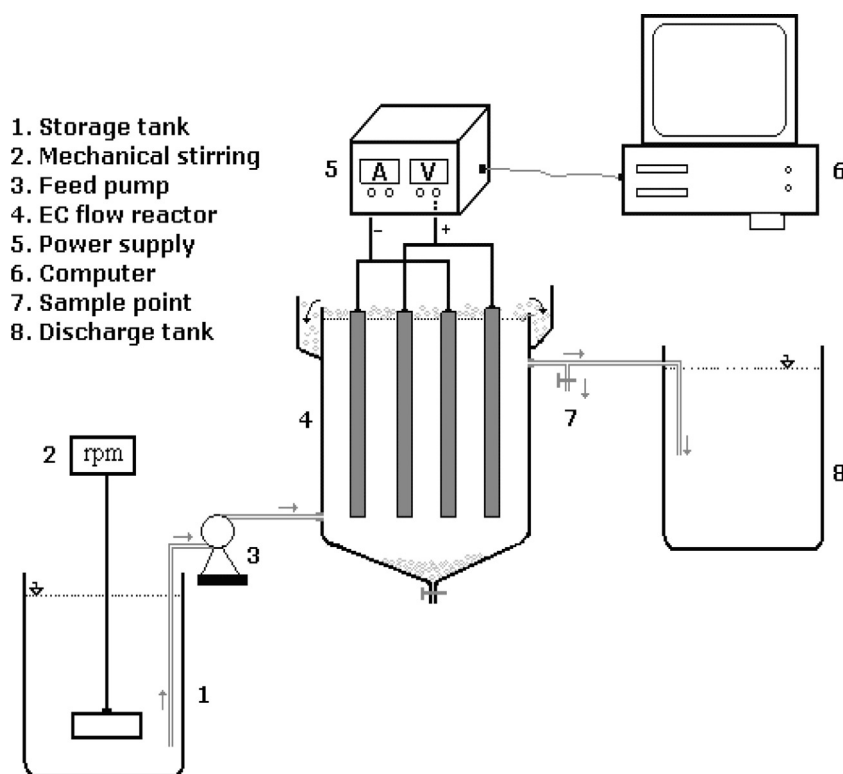


Fig. 1. Experimental setup of the CEC process.

solids/turbidity. Conductivity, pH, and turbidity of the effluent in the reactor were monitored during the course of each experiment. The effluent samples from the reactor were collected and then filtered by Millipore membrane filter with the pore diameter of $0.45 \mu\text{m}$ at the end of the run, and then all samples were analyzed for COD, TOC and turbidity measurements.

2.3. Analytical techniques

The operating parameters namely, pH, conductivity, COD, TOC and turbidity before and after the treatment by the CEC process were analysed with the standard methods [40]. A Hach COD reactor was used for digestion of the sample in COD vials. Values of COD were measured spectrophotometrically by a UV–vis spectrophotometer (PerkinElmer Lambda 35). The TOC levels were determined through combustion of the samples at 680°C using a non-dispersive IR source (Tekmar Dohrmann Apollo 9000). Turbidity (Nephelometric Turbidity Unit, NTU) was determined with a turbidimeter (Mettler Toledo 8300 model). Residual total metal (Fe and Al) concentrations of both raw textile wastewater and EC-treated samples after the filtration were measured with inductive coupled plasma spectrophotometer (PerkinElmer ICP-OES Optima 7000 DV). Removal efficiencies (R_e) of COD, TOC, and turbidity are expressed as a percentage and defined as:

$$R_e = \frac{(C_i - C_f)}{C_i} \times 100 \quad (7)$$

In Eq. (7), C_i and C_f are the COD or TOC or turbidity concentrations in the inlet and the exit streams of the CEC reactor, respectively. pH was determined using a pH-meter (Mettler Toledo 2050e model). A conductivity meter (Mettler Toledo 7100e model) was used to determine the ionic conductivity of the wastewater. The wastewater samples were filtered through a standard GF/F glass fibre filter to determine total suspended solids (TSS). The residual retained on

the filter was dried in an oven at 105°C to a constant weight. The increase in weight of the filter represents the TSS (APHA Method 2540 D). All the inorganic chemicals used were of analytical grade and all reagents were prepared in Millipore milli-Q deionized water.

2.4. Calculation of operating cost

Operating cost (OC) of the EC process includes material, mainly electrodes and electrical energy costs, as well as labour, maintenance, sludge dewatering and disposal, and fixed costs. The latter cost items are largely independent of the type of the electrode material [41,42]. In this study, energy and electrode material costs for calculation of the OC ($\$/\text{m}^3$ or $\$/\text{kg}$ removed COD) were taken into account as major cost items:

$$OC = aENC + bELC + cCC \quad (8)$$

where ENC is energy consumption (kWh/m^3) and ELC is electrode consumption (kg/m^3). Prices for a , b and c (chemical consumption cost (CC) (kg/m^3 or kg/kg COD)) in the Turkish market in September 2015 were $0.1064 \text{ \$/kWh}$ for electrical energy, $1.848 \text{ \$/kg}$ for Al and $0.952 \text{ \$/kg}$ for Fe electrodes, and $1.01 \text{ \$/kg}$ for NaOH, $0.40 \text{ \$/kg}$ for H_2SO_4 for adjustment of a desired pH. On the other hand, ENC and ELC were calculated from Eqs. (9)–(10) and Eqs. (11)–(12), respectively [19,25].

$$ENC(\text{kWh}/\text{m}^3) = \frac{U \times i \times t_{EC}}{v} \quad (9)$$

$$ENC(\text{kWh}/\text{kg COD}) = \frac{U \times i \times t_{EC}}{v \times (C_{i,\text{COD}} \times R_{e,\text{COD}})} \quad (10)$$

where U is cell voltage (V), i is current (A), t_{EC} is operating time (hour), v is volume (m^3) of the wastewater, $C_{i,\text{COD}}$ (kg/m^3) is COD

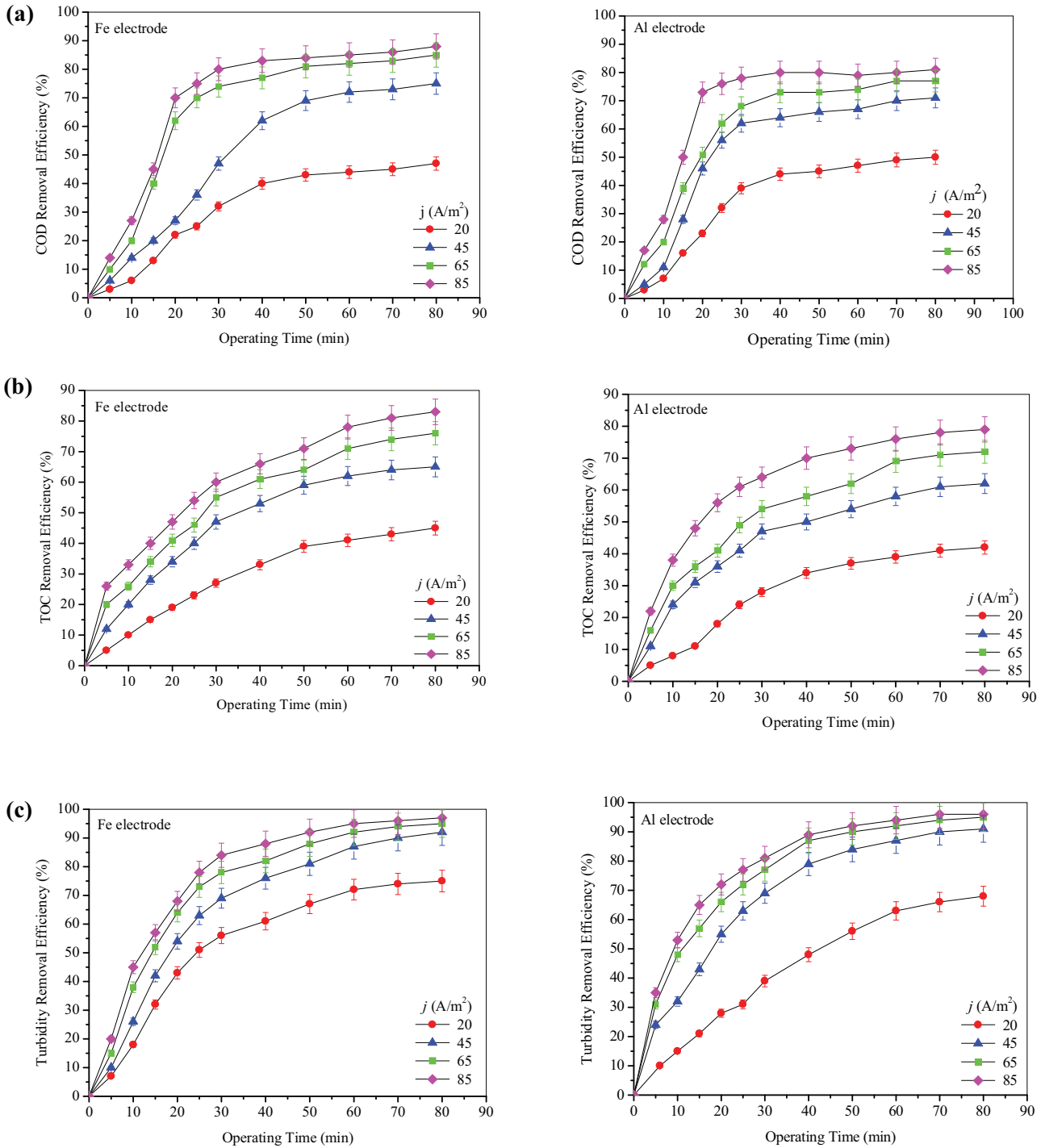


Fig. 2. (a) Effects of current density on COD removal efficiencies (pH_i 5.5, Q=0.01 L/min). (b) Effects of current density on TOC removal efficiencies (pH_i 5.5, Q=0.01 L/min). (c) Effects of current density on turbidity removal efficiencies (pH_i 5.5, Q=0.01 L/min).

concentration of wastewater in the inlet stream, and $R_{e,COD}$ is COD removal efficiency.

$$ELC(\text{kg}/\text{m}^3) = \frac{i \times t_{EC} \times M_w}{z \times F \times v} \quad (11)$$

$$ELC(\text{kg}/\text{kg COD}) = \frac{i \times t_{EC} \times M_w \times \varphi_{Al \text{ or } Fe}}{z \times F \times v \times (C_{i,COD} \times R_{e,COD})} \quad (12)$$

where M_w is molecular mass of electrode ($M_{w,Al}=0.02698$ kg/mol, $M_{w,Fe}=0.05585$ kg/mol), t_{EC} is operating time (s), z is number of electron transferred ($z_{Al}=3$, $z_{Fe}=2$), and F is Faraday's constant (96.487 C/mol). In addition, the faradic yield ($\phi_{Al \text{ or } Fe} = \Delta m_{exp}/\Delta m_{th}$) of electrode (Al or Fe) dissolution was calculated as the ratio of the weight loss of the electrodes during the experiments (Δm_{exp}) and the amount of electrode consumed theoretically (Δm_{th}) at the anode [25]. In this study, faradic yield or current efficiency was calculated as 102–120% for Al electrodes and 100–113% for Fe electrodes at 20–85 A/m².

3. Results and discussion

In the earlier study, optimization using response surface methodology for treatment of the textile dyehouse effluents in a batch EC reactor was studied with considering the effects of initial pH, current density and operating time. [43,44]. The optimum conditions using Al and Fe electrodes for the above study were pH_i 5.5, current density of 45 A/m² and operating time of 33 min. Treatment efficiencies of COD, TOC, turbidity and operating cost were determined to be 60%, 57%, 72% and 0.85 \$/m³ for Al electrode and 71%, 65%, 95% and 2.128 \$/m³ for Fe electrode, respectively. In the present study, the textile dyehouse wastewater was treated with the CEC process by considering three main operating parameters namely, current density, flow rate and operating time.

3.1. Effect of current density

In all electrochemical processes, current density is the most important parameter for controlling the reaction rate within the electrochemical reactor [11]. The current density was determined by dividing each current by the corresponding electrode area ($j = i/A_{\text{electrode}}$, A/m²). It is well known that current density determines the production rate of coagulant, adjusts also bubble production, and hence affects the growth of flocs [27]. A continuous electrocoagulation process using Fe and Al electrodes was carried out at pH_i 5.5 and an inlet flow rate of 0.01 L/min or τ_{react} = hydraulic residence time of 350 min in the CEC reactor to investigate the effect of current density ($j = 20\text{--}85$ A/m²) on the removal efficiencies of COD, TOC and turbidity. COD removal

efficiencies for Al and Fe plate electrodes versus operating time and current density are shown in Fig. 2(a–c). Maximum COD removal efficiencies of 50%, 71%, 77% and 81% for Al electrode and 47%, 75%, 85% and 88% for Fe electrode were obtained at current densities of 20, 45, 65, and 85 A/m². As shown in Fig. 2(a), increase in the current density from 20 to 85 A/m² led to decrease in the operating time required to achieve steady-state concentration, i.e. values on the plateau region and then became constant nearly at about 30–50 min for both electrodes, and increase in percentage of COD removal efficiencies for both electrodes. The reason may be related to increasing of current density led to both increasing of the coagulant production rate and the amount of iron or aluminium hydroxide available in solution to form complexes and precipitate ions. The increase in the current density yielded an increase in the removal efficiencies of COD from 47 to 88% for Fe electrode and from 50 to 81% for Al electrode. At the same time, the highest TOC removal efficiencies were obtained as 45%, 65%, 76% and 83% for Fe electrode and 42%, 62%, 72% and 79% for Al electrodes at 20, 45, 65 and 85 A/m², respectively (Fig 2(b)). Turbidity removal efficiencies at current densities of 20, 45, 65 and 85 A/m² and operating time of 80 min were determined to be 75%, 92%, 95% and 97% for Fe electrode and 68%, 91%, 95% and 96% for Al electrode (Fig. 2(c)). These results were ascribed to the fact that the extent of anodic dissolution of Al and Fe anodes increased at high current densities, resulting in a greater amount of Fe(OH)_{3(s)} or Al(OH)_{3(s)} particles for the removal of pollutants. As the current intensity increased, the anodic dissolution was favoured so that the metallic sludge residues increased and organic pollutants (dissolved dye molecules and other species in the textile wastewater) were

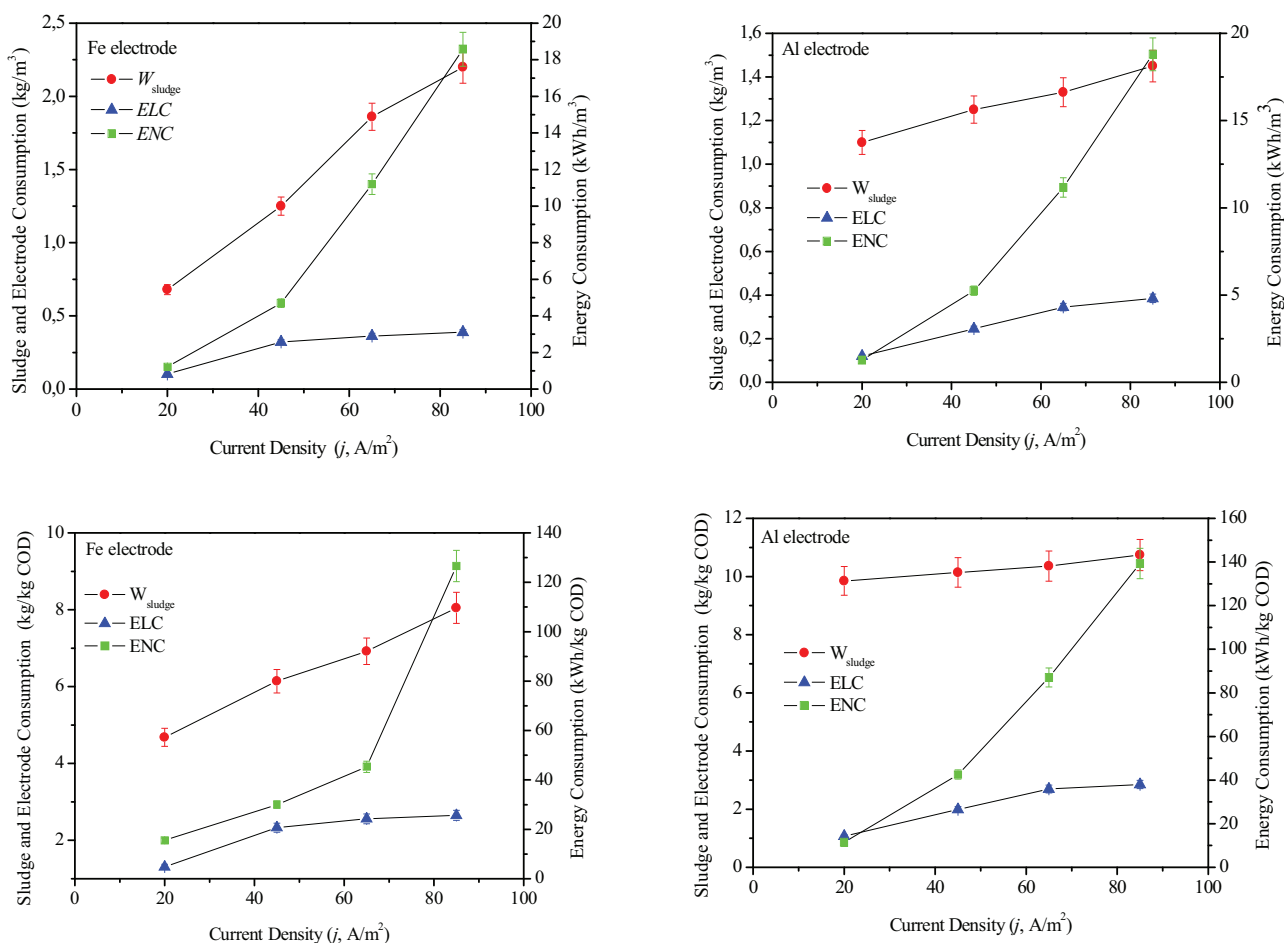


Fig. 3. The amount of sludge generated, electrode and energy consumptions for Fe and Al electrodes at different current densities in the CEC process.

effectively removed from the solution (by co-precipitation or adsorption on metallic sludge). The structure of aluminium sludge was in a white gelatine-type precipitate and was less stable while ferrous sludge formed a stable green colloidal precipitate in aqueous solution. Therefore, the ferrous sludge was more reactive and effective in removing organic pollutants by precipitation/adsorption [45]. Moreover, the bubble generation rate increased and the bubble size decreased with increasing the current densities. These effects were both beneficial for high pollutant removal by $H_{2(g)}$ flotation [27]. Removal efficiencies at 65 and 85 A/m² for Fe electrodes were determined to be 85% and 88% for COD, 76% and 83% for TOC, and 95% and 97% for turbidity at 80 min. On the other hand, removal efficiencies at 65 and 85 A/m² for Al electrodes were 77% and 81% for COD, 72% and 79% for TOC, and 95% and 96% for turbidity. The results indicated that Fe electrode required slightly lower current density than that of Al electrode for the same operating conditions. This probably resulted from the differences in the removal mechanisms of COD and TOC. The COD and TOC from the textile wastewater using Fe electrode were mainly removed by the EC process, while the COD and TOC removals by Al plate electrodes were due to the synergic effect of EC and electrooxidation [1,22]. Although, the higher current density gave slightly better results but, 65 A/m² was selected for both electrodes as the optimum current efficiency value for this study due to lowering the operating cost. Consequently, the CEC process involving Fe electrode gave rise to removal efficiencies of 85% for COD, 76% for TOC and 95% for turbidity at the optimum current density value.

The energy (ENC) and electrode (ELC) consumptions calculated on the basis of kWh/m³ and kWh/kg COD removed using Eqs. (9) and (12) for both electrodes are depicted in Fig. 3. Values of the energy consumptions at 20–85 A/m² were 1.22–18.57 kWh/m³ (15.57–126.61 kWh/kg COD) for Fe electrode, and 1.28–18.8 kWh/m³ (11.46–139.26 kWh/kg COD) for Al electrode, respectively. Values of the electrode consumptions at 20–85 A/m² for Fe and Al electrodes were 0.103–0.388 kg/m³ (1.31–2.65 kg/kg COD) and 0.120–0.385 kg/m³ (1.07–2.85 kg/kg COD) (Fig. 3). In addition, the amount of sludge occurred during the CEC process at 20–85 A/m² were varied from 0.68 to 2.20 kg/m³ (4.68–8.05 kg/kg COD) for Fe electrode, and from 1.10 to 1.45 kg/m³ (9.85–10.74 kg/kg COD) for Al electrode since the current density and operating time affected the amount of sludge generated. Therefore, the sludge production in the CEC process was proportional to characteristics of wastewater, settable solids

and matter destabilized by coagulation and flocculent concentration. According to the above results, the sludge production in the CEC process for both electrodes increased with increasing of current density since amounts of dissolved Fe and Al anodes at the constant operating time (80 min) were increased (Faraday's law).

Changes of the operating costs as a function of the current density for both electrodes are illustrated in Fig. 4. The variations in the operating costs for both electrodes were calculated on the basis of \$/m³ and \$/kg COD removed. The operating costs at 20–85 A/m² were 0.253–2.370 \$/m³ or 2.929–16.019 \$/kg COD removed for Fe electrode and 0.383–2.737 \$/m³ or 3.222–20.109 \$/kg COD removed for Al electrode (Fig. 4). On the other hand, the operating cost increased rapidly with increasing current density from 20 to 85 A/m². Fe electrode lowered the operating cost per m³ or removed kg COD of the wastewater as compared to Al electrode did. From the operating cost and removal efficiencies of COD, TOC and turbidity points of view, Fe electrode in the wastewater was clearly preferable since it lowered the operating cost significantly in the CEC process.

Residual metal (Al and Fe) concentrations are an important consideration for any wastewater treatment by EC process using sacrificial anodes such as Al and Fe. The residual metal concentrations may decrease carrying capacity of the sewer system because of precipitation of aluminium and iron hydroxides and cause toxic effects on organisms and increase turbidity in receiving water environments such as river and lake. There are also concerns that residual aluminium in drinking water may cause Alzheimer's disease. Residual aluminium or iron concentrations in the treated samples by the CEC process at current densities of 20, 45, 65, and 85 A/m² and operating time of 80 min, were obtained as 0.0392, 0.0241, 0.0192, and 0.0131 mg/L for Al electrodes and 0.0414, 0.0326, 0.0213, and 0.0203 mg/L for Fe electrodes. The residual Al and Fe concentrations in the treated effluents at different current densities were lower than the permissible limit for aluminium and iron in drinking waters (0.3 mg/L) set by Turkish Water Pollution Control Regulation. On the other hand, the permissible limits for sewer system discharging of textile wastewater after the treatment were 3 mg/L for Al and 5 mg/L for Fe.

3.2. Effect of inlet flow rate

The time evolution of treatment of wastewater by electrocoagulation is often presented in the literature by a plotted

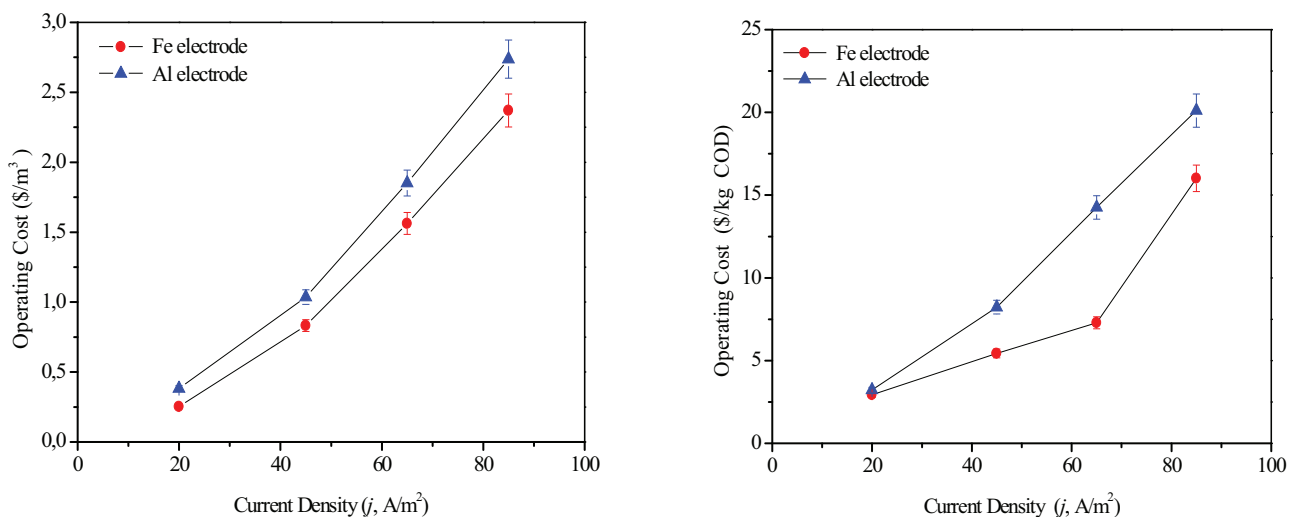


Fig. 4. Effects of current density for Al and Fe electrodes on the operating cost.

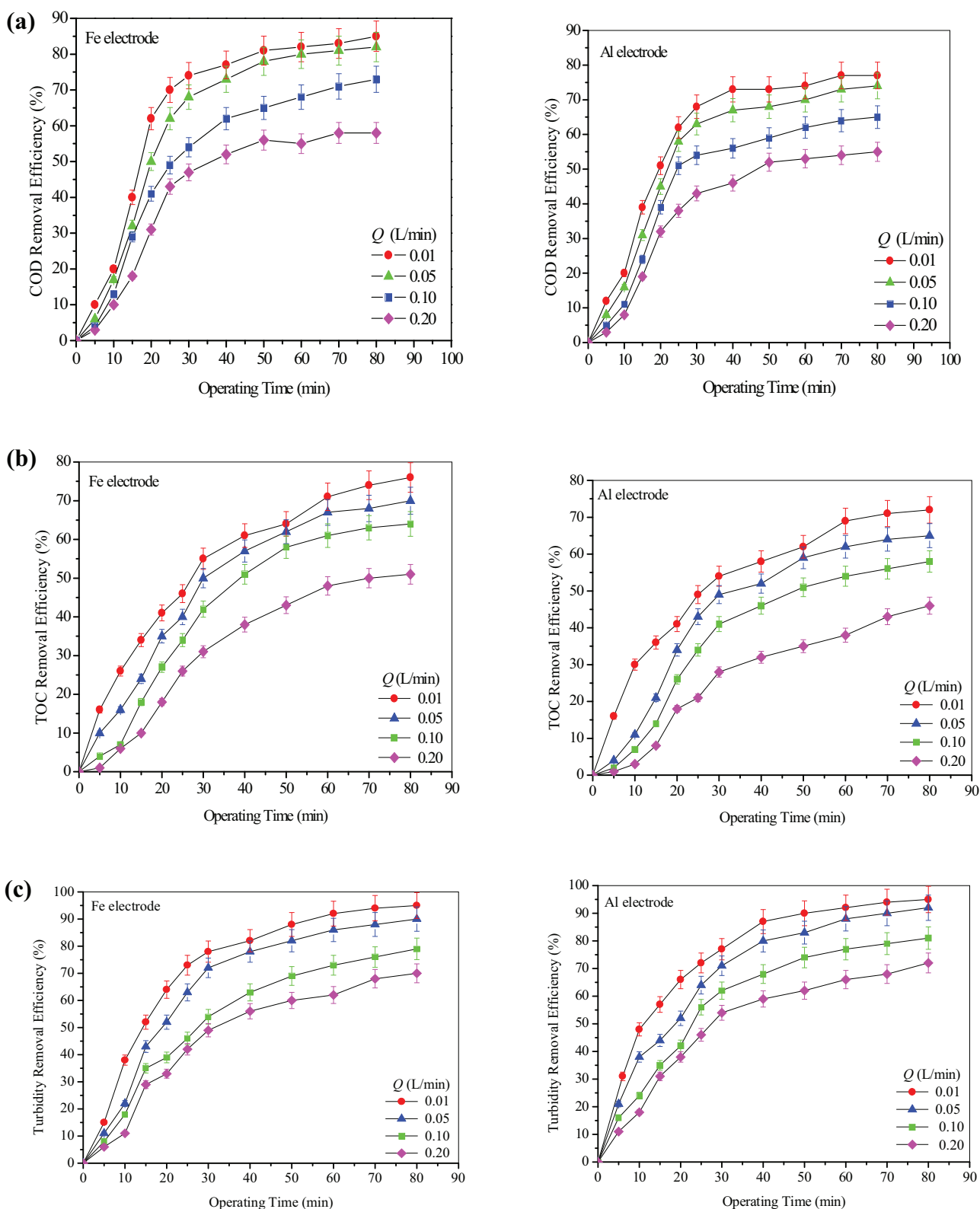


Fig. 5. (a) Effects of flow rate for both electrodes on COD removal efficiencies in the CEC process (pH_i 5.5, $j = 65 \text{ A/m}^2$). (b) Effects of flow rate for both electrodes on TOC removal efficiencies in the CEC process (pH_i 5.5, $j = 65 \text{ A/m}^2$). (c) Effects of flow rate for both electrodes on turbidity removal efficiencies in the CEC process (pH_i 5.5, $j = 65 \text{ A/m}^2$).

parameter to follow the treatment efficiency (COD, TOC, turbidity) vs. operating time. It is much more appropriate to normalize the time by the mean residence time to analyze properly the effect of the flow rate [24,29,46]. As the flow rate is decreased in the CEC

process, the hydraulic retention time in the CEC reactor is increased, so it can cause the restabilization of aluminium or iron hydroxides flocs. In other words, as the inlet flow rate increases for rapid mixing in the CEC reactor, the retention time decreases. The

rapid mixing is required in the initial condition of the EC process to grow larger flocs and to precipitate these enough in the solutions for the removal. Thus, it would be of practical interest to test how much an increase or decrease in the effluent inlet flow rate affects the COD, TOC, and turbidity removal efficiencies for the dyehouse effluent in the CEC process.

Effect of inlet flow rate on treatment of the dyehouse effluent in the CEC process at constant operating conditions (65 A/m^2 and initial pH_i 5.5) is shown in Fig 5(a–c). Because of constant total volume of the CEC reactor, an increase in the wastewater inlet flow rate led to a proportional decrease in the hydraulic retention time of the wastewater in the reactor. As the inlet flow rate was increased to 0.010, 0.050, 0.10 and 0.20 L/min, the hydraulic retention time directly decreased to 350, 70, 35 and 17.5 min (figure not depicted). In this way, such a reduction in the hydraulic retention time would lead to a decrease in the COD, TOC and turbidity removals from the wastewater. As seen in Fig. 5(a–c), the inlet flow rate increased from 0.01 to 0.20 L/min, COD, TOC and turbidity removal efficiencies were decreased from 85% (300 mg/L) to 58% (840 mg/L), from 76% (116 mg/L) to 51% (238 mg/L) and from 95% (111 NTU) to 70% (668 NTU) for Fe electrode and from 77% (460 mg/L) to 55% (900 mg/L), from 72% (136 mg/L) to 46% (262 mg/L) and from 95% (111 NTU) to 72% (623 NTU) for Al electrode, respectively. As seen in Fig. 5(a–c), increase in the dyehouse wastewater inlet flow rate from 0.010 L/min to 0.20 L/min reduced the COD, TOC and turbidity removal efficiencies only by 27%, 25% and 25% for Fe electrode and 22%, 26% and 23% for Al electrode. The CEC process using both electrodes

provided COD removal efficiency higher than 60% for all the operating conditions except for 0.20 L/min. The slower flow rate allowed the coagulant generated by electrochemically dissolved of the electrodes and the pollutants in the wastewater to mix properly and thereby improved the rate of coagulation.

The longer the hydraulic retention time of the wastewater, the more floc generated by the CEC process, so enhanced the oxidation, precipitation and adsorption process. In this case, the COD, TOC and turbidity removal efficiencies increased significantly. The electrode and energy consumptions at flow rates of 0.01–0.20 L/min were varied in the range of $0.363\text{--}0.135 \text{ kg/m}^3$ (or $2.56\text{--}3.45 \text{ kg/kg COD}$) and $11.20\text{--}3.34 \text{ kWh/m}^3$ (or $45.30\text{--}60.60 \text{ kWh/m}^3$) for Fe electrode and $0.345\text{--}0.147 \text{ kg/m}^3$ (or $2.69\text{--}2.91 \text{ kg/kg COD}$) and $11.17\text{--}3.10 \text{ kWh/m}^3$ (or $87.04\text{--}98.06 \text{ kWh/kg COD}$) for Al electrode (Fig. 6). These results indicated that sludge production, energy and electrode consumptions per m^3 treated wastewater for both electrodes decreased with increasing of the inlet flow rate. On the other hand, the amount of sludge in the CEC process varied in the range of $1.86\text{--}0.63 \text{ kg/m}^3$ (or $6.92\text{--}10.05 \text{ kg/kg COD}$) for Fe electrode and $1.33\text{--}1.05 \text{ kg/m}^3$ (or $10.36\text{--}15.96 \text{ kg/kg COD}$) for Al electrode at inlet flow rates of 0.010–0.20 L/min (Fig. 6). Operating costs were calculated as $1.562\text{--}0.599 \text{ \$/m}^3$ (or $7.069\text{--}9.757 \text{ \$/kg COD}$) for Fe electrode and $1.851\text{--}0.626 \text{ \$/m}^3$ (or $14.257\text{--}15.894 \text{ \$/kg COD}$) at inlet flow rates of 0.010–0.20 L/min (Fig. 7). The operating cost for both electrodes increased with increased flow rate per m^3 treated wastewater but the cost decreased with per removed kg COD. The residual aluminium and iron concentrations in treated

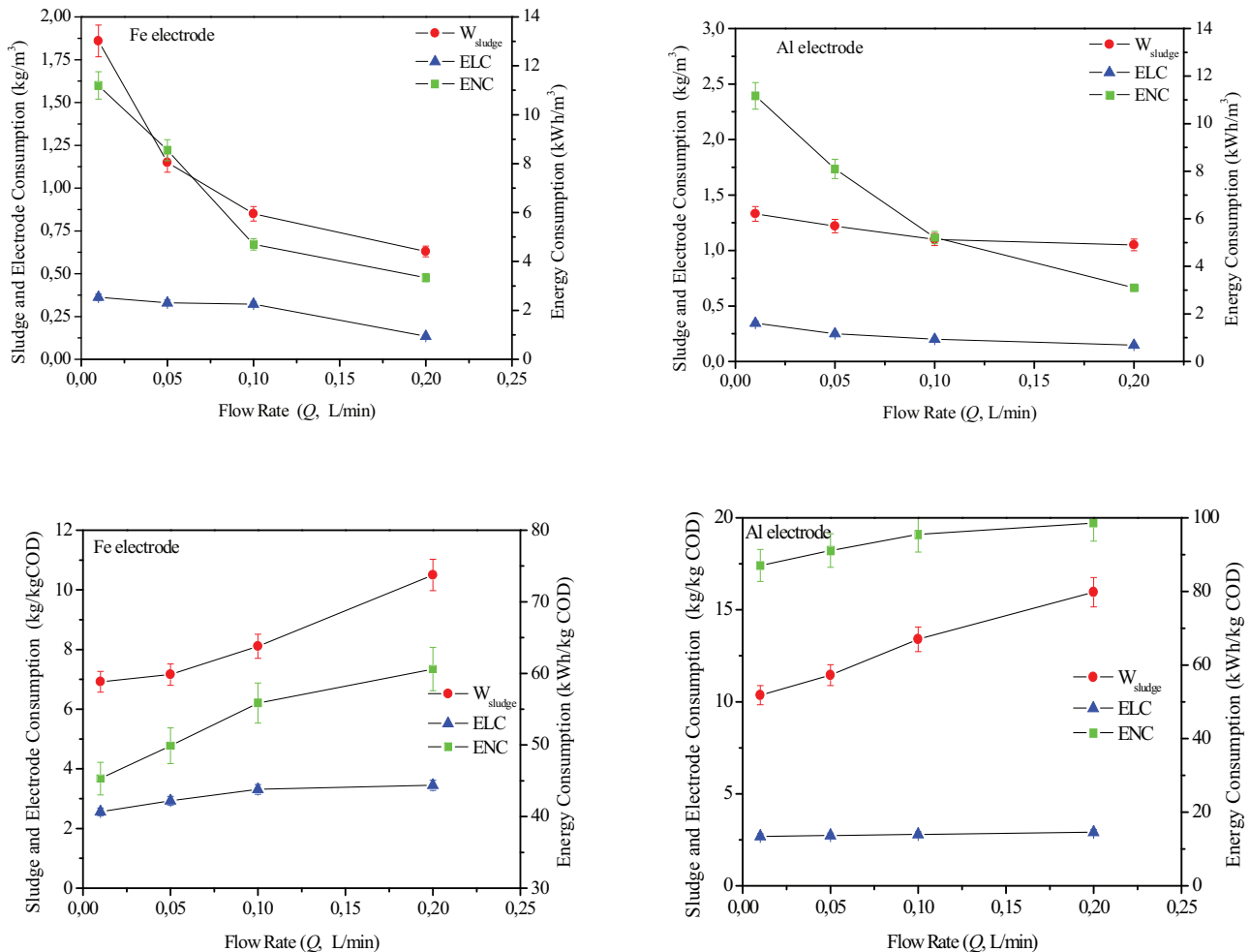


Fig. 6. Values of sludge, electrode and energy consumptions at different flow rates in the CEC process.

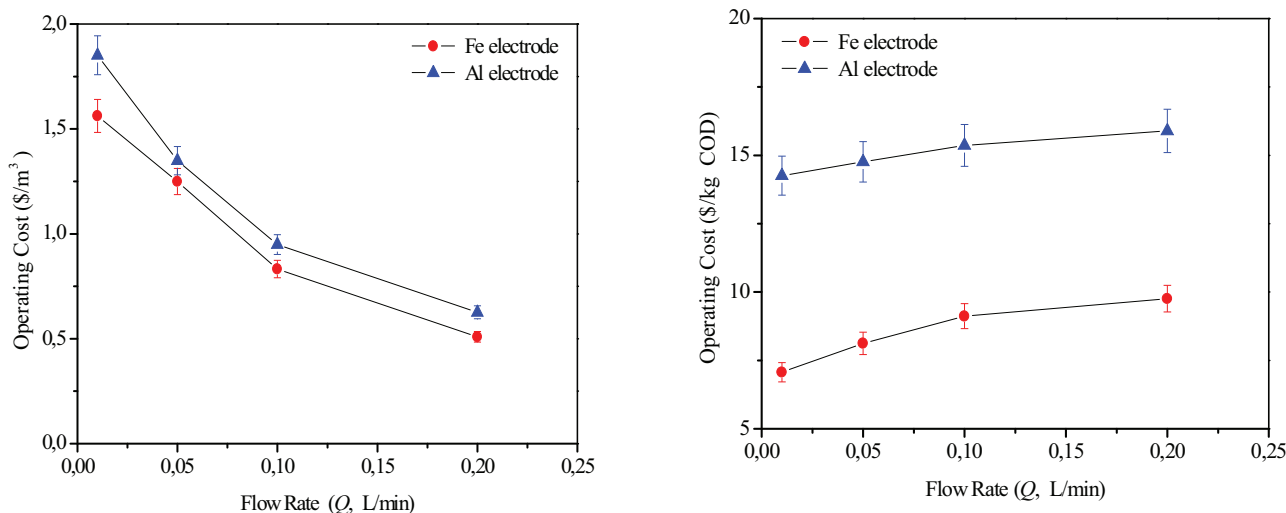


Fig. 7. Effects of flow rate on the operating cost for Al and Fe electrodes.

samples for inlet flow rates of 0.010, 0.050, 0.10 and 0.20 L/min at operating time of 80 min were obtained as 0.0192, 0.0245, 0.0312 and 0.0436 mg/L for Al electrodes and 0.0213, 0.0452, 0.0519 and 0.0603 mg/L for Fe electrodes.

3.3. Effect of COD concentration

Textile dyeing processes include pretreatment, dyeing, finishing and other technologies. Pre-treatment includes desizing, scouring, washing, and other processes [47,48]. Dyeing mainly aims at dissolving the dye in water, which will be transferred to the fabric to produce colored fabric under certain conditions. The primary source of wastewater in dyeing operations is spent dye bath and wash water. Pre-treatment wastewater accounts for about 45% of the total, and dyeing/printing process wastewater accounts for about 50–55%, while finishing process produces little. About 50% of the textile mills are using approximately 200–250 liters of water per kg of fabric produced while about 20% of the mills is using below 200 liters of water per kg of fabric depending on the process sequence and water utilization practices adopted in the mill [49]. Water pollution by the textile mills is mainly attributable to various waste liquors coming out of the unit operations in wet

processing such as desizing, scouring, bleaching, mercerizing, dyeing, printing and finishing. This process can be divided into continuous and batch type [5,7,47,48]. Taking the batch type as an example, the concentration of COD is up to 20,000–60,000 mg/L. The wastewater from the reduction process may account for only 5% of the volume of wastewater, while COD accounts for 60% or more in the conventional dyeing and finishing.

The COD concentration is the most important parameter required for the sewage discharge standards. The source of pollution in the textile wastewater arises mainly from dye rinse wastewater which contains high rate of pollution. Treatments of concentrated textile dyeing bath wastewater are more difficult than rinse bath wastewaters. These wastewaters are also mixed with the other wastewaters apart from the dyeing bath process which increases the amount of wastewater and hence, amount of COD in the wastewater changes. Characterizations of the wastewater obtained from the balancing tank of the textile dyehouse plant are shown in Table 1. However, the effluent wastewater from the dyeing bath of this dyehouse plant contained average values of 22,000 mg/L of COD, 6500 mg/L of TOC, 20,500 NTU of turbidity at pH, 6.8. The removal efficiency would be very low if only the dyeing bath wastewater is treated by the CEC process

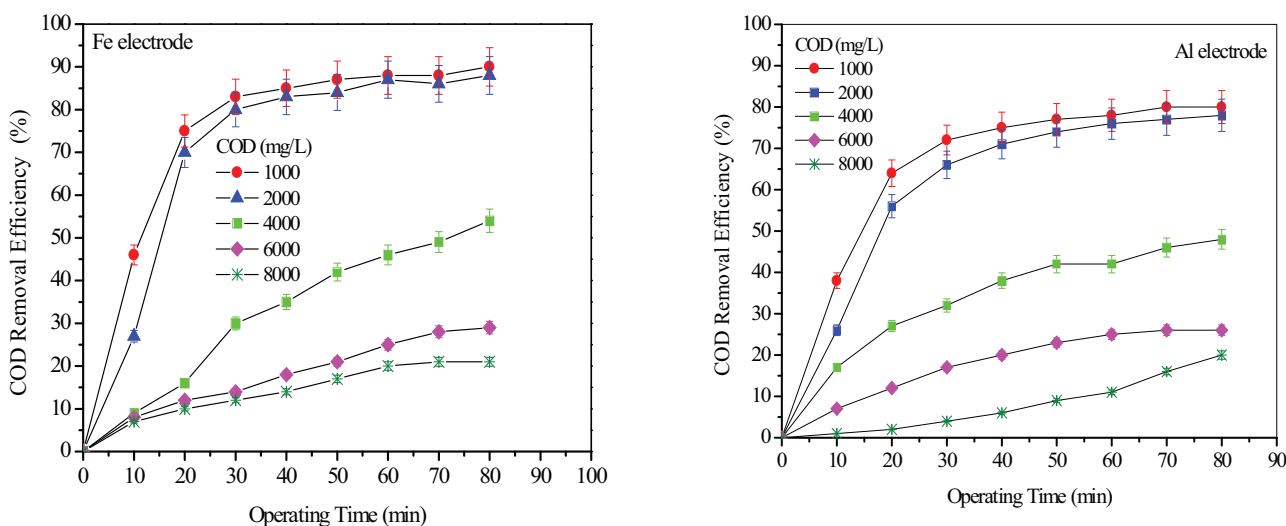


Fig. 8. Effects of COD concentration on the treatment of the wastewater in the CEC process (65 A/m², pH, 5.5).

due to high concentrations of COD and TOC in the wastewater. Therefore, the dyeing bath wastewater containing intensive pollution was added to the balancing tank wastewater to obtain different initial concentrations of COD. Then, the removal efficiencies of COD having different initial concentrations were studied and the results for Fe and Al electrodes are depicted in Figs. 8 and 9.

According to these figures, the removal efficiencies using Al electrodes in the CEC reactor were achieved to 80% and 78% for COD concentrations of 1000 and 2000 mg/L in the wastewater, 74% and 68% for TOC and 96% and 93% for turbidity. The removal efficiencies at high concentrations of COD present in the wastewater (4000, 6000 and 8000 mg/L) were also obtained as 48%, 26% and 20% for COD, 44%, 21% and 16% for TOC and 76%, 65% and 36% for turbidity, respectively. Effluent pH values at different concentrations of COD in the wastewater varied in the range of 8.9–8.1 after the CEC process (figures not depicted). Moreover, the removal efficiencies in the CEC reactor using Fe electrodes were 90% and 88% for COD concentrations of 1000 and 2000 mg/L in the wastewater, 83% and 78% for TOC and 93% and 91% for turbidity. The removal efficiencies at high concentrations of COD present in the wastewater using Fe electrode (4000, 6000 and 8000 mg/L) were 54%, 29% and 21% for COD, 48%, 22% and 18% for TOC and 72%, 52% and 30% for turbidity, respectively. Effluent pH values at different concentrations of COD in the wastewater varied in the range of 9.2–8.2 after the CEC process (figures not depicted). According to these results, the removal efficiency of COD was decreased significantly in the wastewater containing COD concentration of >2000 mg/L since there were not enough Al^{3+} and Fe^{3+} hydroxyl species from the electrodes, i.e., intensive pollutants concentrations in the wastewater demand larger electrogenerated coagulant mass. On the other hand, dye molecules and polymeric species like surfactants adsorbing the electrode surfaces (deposited) caused passivation of the electrodes as well. This situation can be attributed to the inhibition of the electrodes surfaces by the dye molecules and polymeric species in the wastewater deactivating the active sites of the electrode surfaces. Similar results were reported in the literature [50] and could be seen in Fig. 8. The removal efficiency was not increased much with increasing the operating time. This caused to decrease in the reaction rates. The dissolution of the anodes was not enough due to passivation, and efficiencies from dissolution through electrochemical (the faradic efficiency) were under 100% (45–80% for Al and 35–75% for Fe electrodes). Therefore, the temperature in the CEC reactor

increased from 20 to 28 °C at 80 min due to the increasing of mobility and ion collisions with the hydroxyl polymers. However, higher temperatures decreased the removal efficiency due to the formation of unsuitable flocks and/or solubility increase [51]. The removal efficiency was observed to be lower than as expected (≥ 2000 mg/L of COD) since adsorption of the wastewater on surface of the electrodes caused passivation of the electrode material.

Energy consumptions for initial COD concentrations of 1000, 2000, 4000, 6000 and 8000 mg/L were calculated as 6.35, 11.20, 13.8, 15.2 and 16.1 kWh/m³ for Fe electrode and 6.92, 11.17, 14.20, 15.71 and 16.84 kWh/m³ for Al electrode. As the initial COD concentration was increased, values of the energy consumption increased, the electrode surfaces were deactivated and potential between the electrodes was increased in the CEC reactor. Therefore, the energy consumptions were increased with respect to the obtained results. On the other hand, the electrode consumptions were 0.252, 0.363, 0.374, 0.320 and 0.301 kg Fe/m³ and 0.276, 0.345, 0.379, 0.328, and 0.321 kg Al/m³ at initial COD concentrations of 1000, 2000, 4000, 6000 and 8000 mg/L. Dissolved amount of Al and Fe electrodes per m³ was getting less when concentration of COD was greater than 2000 mg/L due to existence of layer of deposition film on surface of the electrodes (a layer of color precipitation related to the wastewater) and deactivation of the electrode surfaces.

The other important issue in the treated wastewater was to evaluate parameters with respect to discharge standards. The discharge standards for the textile industry in Istanbul are set by Istanbul Water and Sewerage Administration (ISKI) which controls and inspects the industrial wastewater discharges [52]. Industries are required to pretreat their wastewater to meet the standards set by ISKI. The sewage discharge standard of COD for the dyehouse effluents in Turkey is 800 mg/L. According to the obtained results, initial COD concentration was reduced from 2000 mg/L to 800 mg/L (i.e., COD removal efficiency of 60% for Al and Fe electrodes) at inlet flow rate of below 0.01 L/min, pH_i 5.5, current density of ≥ 65 A/m² and operating time of 80 min in the CEC process. It can be concluded with based on the obtained results that the optimum conditions ($j = 65$ A/m², $t_{\text{EC}} = 80$ min, $Q = 0.010$ L/min, and COD of 2000 mg/L) for COD, TOC and turbidity effluent concentrations from the wastewater in CEC process were COD of 300 mg/L, TOC of 116 mg/L and turbidity of 111 (NTU) for Fe electrode and COD of 460 mg/L, TOC of 136 mg/L and turbidity of 111 NTU for Al electrodes. According to ISKI standards, values of

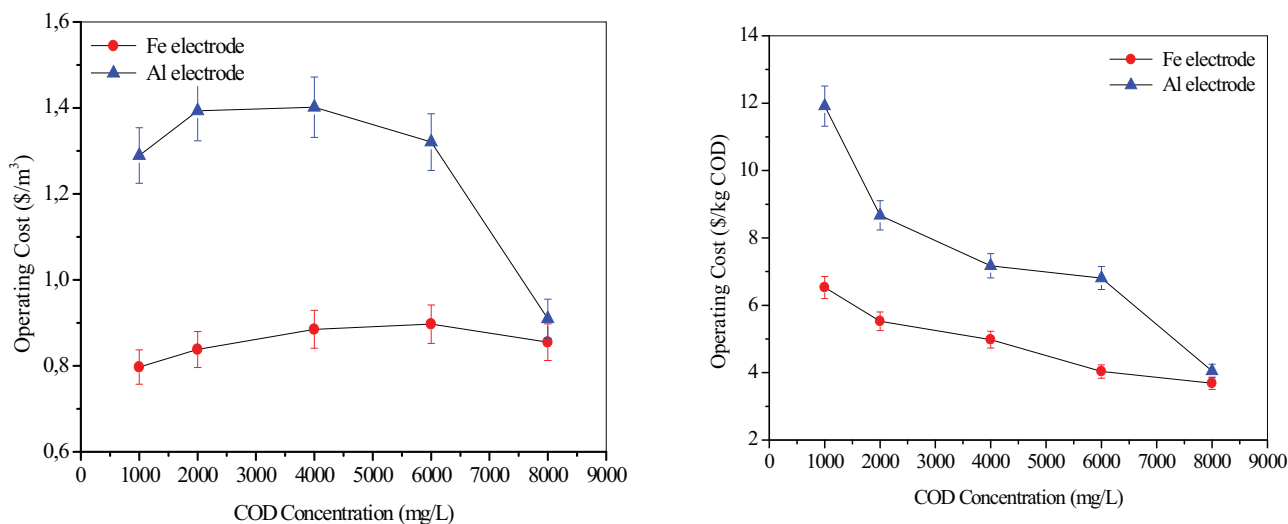


Fig. 9. Effects of COD concentration on the operating costs for Al and Fe electrodes.

Table 2

The summary of studies on treatment of synthetic and real textile wastewater by continuous flow electrocoagulation process.

Dye and textile wastewater	Experimental conditions*	Optimum conditions*	Ref.
Synthetic dye wastewater (a red dye: mixture of 2-naphthoic acid and 2-naphthol)	N_e : two Al electrodes (used as anode and cathode), connection mode: MP-P, $S_e = 192 \text{ cm}^2$, $C_{\text{dye}} = 25\text{--}200 \text{ mg/L}$ (625–5000 mg/L COD), pH_i 3.2–9.01, $j = 20.8\text{--}62.5 \text{ mA/cm}^2$, $C_{\text{NaCl}} = 1500\text{--}5000 \text{ mg/L}$ ($\kappa = 2.4\text{--}8 \text{ mS/cm}$), $d = 1 \text{ cm}$, $Q = 25.2\text{--}78 \text{ L/h}$	$R_{e,\text{COD}} = 95\text{--}85\%$; $R_{e,\text{dye}} = 95\text{--}85\%$; $\tau = 14 \text{ min}$, $j = 31.25 \text{ mA/cm}^2$, $Q = 37.2 \text{ L/h}$, $\tau = 14 \text{ min}$, pH_i 6.1, $\kappa = 2.4 \text{ mS/cm}$, $\text{ENC} = 3.5\text{--}3.2 \text{ kWh/kg dye}$, $\text{ELC} = 0.155\text{--}0.180 \text{ kg Al/kg dye}$, $\text{OC} = 0.52\text{--}0.56 \text{ \$/kg dye}$ ($C_{\text{dye}} = 100\text{--}200 \text{ mg/L}$)	[24,25]
Synthetic dye wastewater (Reactive Blue 49 (RB-49), Reactive Yellow-84 (RY-84), Disperse Blue 106 (DB-106), Disperse Yellow 54 (DY-54) and real textile wastewater)	N_e : 4–7 pair as Al, Fe and stainless plate (SS) (as both anodes and cathodes), connection mode: MP-P, $S_e = 640\text{--}1120 \text{ cm}^2$, $C_{\text{dye}} = 200\text{--}1000 \text{ mg/L}$, $C_{\text{COD}} = 870 \text{ mg/L}$ (wastewater), pH_i 4–12, $C_{\text{NaCl}} = 0\text{--}2000 \text{ mg/L}$, $j = 1.01\text{--}4.5 \text{ mA/cm}^2$; $d = 0.5\text{--}3 \text{ cm}$, $Q = 50\text{--}200 \text{ mL/min}$, $\tau = 26\text{--}6.5 \text{ min}$	$R_{e,\text{color}} = 99.5\%$ (DB-106), 98% (DY-54), 90% (RY-84) and 72% (RB-49) $R_{e,\text{COD}} = 62\%$ (DB-106), 50% (DY-54), 60% (RB-49) and 88% (RY-84), $R_{e,\text{wastewater}} = 51.4\%$ (COD) and 41.4% (color) $j = 4.45 \text{ mA/cm}^2$, $C_{\text{NaCl}} = 1000 \text{ mg/L}$, $d = 1 \text{ cm}$, $C_{\text{dye}} = 500 \text{ mg/L}$, $\tau = 13 \text{ min}$, $N_e = 7$ pairs Al anodes, $Q = 100 \text{ mL/min}$, the influence of the anode: $\text{Al} > \text{SS} > \text{Fe}$	[29]
Synthetic textile wastewater (Direct Red 81)	N_e : six Al plate anodes and cathodes, connection mode: BP-S, $S_e = 1200 \text{ cm}^2$, $C_{\text{dye}} = 50 \text{ mg/L}$, $C_{\text{COD}} = 120 \text{ mg/L}$, Turbidity = 380 NTU, pH_i 7.5, $j = 100\text{--}200 \text{ A/m}^2$, $\kappa = 2575 \pm 25 \mu\text{S/cm}$, $d = 1 \text{ cm}$, $Q = 10\text{--}28 \text{ L/h}$, $\tau = 1.6\text{--}0.57 \text{ h}$	$R_{e,\text{dye}} = 71.5\text{--}90.2\%$, $R_{e,\text{COD}} = 52.3\text{--}76.1\%$ and $R_{e,\text{NTU}} = 99.9\text{--}95.6\%$, $\tau = 1.6 \text{ h}$, $j = 150 \text{ A/m}^2$ and $Q = 10 \text{ L/h}$ (97.3% NTU, 69.4% COD and 80.5% dye), $\text{ENC} = 52\text{--}66 \text{ kWh/kg Al}$ (0.07–0.19 kWh/mg removed dye)	[54]
Synthetic textile wastewater (Remazol Red RB 133)	N_e : Al anode and two stainless plate cathodes, connection mode: MP-P, $S_e = 235 \text{ cm}^2$, $C_{\text{dye}} = 250 \text{ mg/L}$, pH_i 6, $j = 5\text{--}17 \text{ mA/cm}^2$; $\kappa = 249\text{--}1966 \mu\text{S/cm}$ (NaCl), $d = 0.75 \text{ cm}$, $Q = 0.6\text{--}2 \text{ L/h}$, $\tau = 23\text{--}6.8 \text{ min}$	$R_{e,\text{dye}} = 99.3\text{--}93\%$ (optimum 96.9%) $\tau = 11.5 \text{ min}$, $j = 10 \text{ mA/cm}^2$, $Q = 1.2 \text{ L/h}$, $\kappa = 500 \mu\text{S/cm}$, $\text{ELC} = 1.3\text{--}0.4 \text{ kg Al/m}^3$ (theoretical)	[55]
Synthetic textile wastewater (Reactive Blue 21)	N_e : five pairs Fedisk electrodes (diameter = 12.2 cm), connection mode: MP-P, $S_e = 1011 \text{ cm}^2$, $C_{\text{dye}} = 500 \text{ mg/L}$, $\kappa = 2055 \mu\text{S/cm}$, $d = 0.75 \text{ cm}$, $Q = 0.75\text{--}0.1 \text{ L/min}$, $\tau = 2\text{--}20 \text{ min}$	$R_{e,\text{color}} = 99\%$, $R_{e,\text{COD}} = 90\%$, $j = 30 \text{ A/m}^2$, $Q = 0.75\text{--}0.1 \text{ L/min}$, $\tau = 10\text{--}20 \text{ min}$, $\text{ENC} = 0.495 \text{ kWh/m}^3$ (1 kWh/kg dye)	[56]
Synthetic textile wastewater (Basic Red 5001 B)	N_e : one Fe anode and one Fe cathode, connection mode: MP-P, $S_e = 124.4 \text{ cm}^2$, $C_{\text{COD}} = 300 \text{ mg/L}$, pH_i 9, $j = 7.55\text{--}23 \text{ mA/cm}^2$; $\kappa = 4.7 \text{ mS/cm}$, $d = 4 \text{ cm}$, $Q = 75 \text{ mL/min}$, $\tau = 20\text{--}40 \text{ min}$	$R_{e,\text{dye}} = 95\%$ and $R_{e,\text{COD}} = 76\%$, $\tau = 20 \text{ min}$, $j = 14\text{--}17 \text{ mA/cm}^2$, $Q = 75 \text{ mL/min}$,	[57]
Synthetic textile wastewater (Reactive Blue 140: RB 140; Reactive Red 23: RR 23)	N_e : 25 circular shape Fe electrodes (diameter: 12.2 cm), connection mode: MP-P, $S_e = 5625 \text{ cm}^2$, $C_{\text{dye}} = 100 \text{ mg/L}$, $j = 30\text{--}40 \text{ A/m}^2$, $d = 0.8 \text{ cm}$, pH_i 9.6, $\kappa = 875 \pm 25 \mu\text{S/cm}$ (NaCl), $Q = 0.07\text{--}1140 \text{ mL/min}$, $\tau = 5\text{--}10 \text{ min}$	$R_{e,\text{dye}} = 96\%$ (RB 140), $R_{e,\text{dye}} = 98\%$ (RR 23) $j = 40 \text{ A/m}^2$ (RB 140), $j = 30 \text{ A/m}^2$ (RR 23), $Q = 884 \text{ mL/min}$, $\tau = 5 \text{ min}$, $\text{ENC} = 1.42 \text{ kWh/m}^3$ (RB 140), $\text{ENC} = 0.69 \text{ kWh/m}^3$ (RR 23), $\text{OC} = 0.152 \text{ \$/m}^3$	[58]
Synthetic textile wastewater (Orange II)	N_e : five Al plate electrodes, connection mode: BP-S, $S_e = 1254 \text{ cm}^2$, $C_{\text{dye}} = 10\text{--}50 \text{ mg/L}$, $j = 120\text{--}215 \text{ A/m}^2$, pH_i 2–12.6, $\kappa = 3.9\text{--}14.07 \text{ mS/cm}$ (2–8 g/L NaCl), $d = 0.6 \text{ cm}$, $Q = 300\text{--}575 \text{ mL/min}$, $\tau = 7.4\text{--}4.2 \text{ min}$	$R_{e,\text{dye}} = 94.5\%$ $j = 120 \text{ A/m}^2$, $\tau = 7.4 \text{ min}$, $C_{\text{dye}} = 10 \text{ mg/L}$, pH_i 6.5, $\kappa = 7.1 \text{ mS/cm}$ (4 g/L NaCl), $Q = 350 \text{ mL/min}$	[59]
Synthetic textile wastewater (Acid Orange 6)	N_e : six Fe plate electrodes, connection mode: BP-S, $S_e = 38.88 \text{ m}^2$, $C_{\text{dye}} = 50\text{--}200 \text{ mg/L}$, $j = 0.06\text{--}150 \text{ A/cm}^2$, pH_i 4–10, $\kappa = 200 \mu\text{S/dm}$, $d = 0.5 \text{ cm}$, $Q = 0.3\text{--}0.9 \text{ L/min}$, $\tau = 2.7\text{--}0.9 \text{ min}$	$R_{e,\text{color}} = 98\%$, $R_{e,\text{TOC}} = 40\%$, $j = 68.3 \text{ A/dm}^2$, $C_{\text{dye}} = 50 \text{ mg/L}$, pH_i 4, $Q = 0.3\text{--}0.5 \text{ L/min}$, $\tau = 2.7\text{--}1.6 \text{ min}$	[60]
Synthetic textile wastewater (Orange II)	N_e : five carbon steel plate electrodes, connection mode: BP-S, $S_e = 1254 \text{ cm}^2$, $C_{\text{dye}} = 10\text{--}50 \text{ mg/L}$, $j = 159.5\text{--}398.7 \text{ A/m}^2$, pH_i 4.7–11, $\kappa = 3.76\text{--}10.53 \text{ mS/cm}$ (1.99–5.97 g/L NaCl), $d = 0.6 \text{ cm}$, $Q = 350\text{--}600 \text{ mL/min}$, $\tau = 1.32\text{--}0.75 \text{ min}$	$R_{e,\text{dye}} = 98.5\%$ $j = 319 \text{ A/m}^2$, $\tau = 1.32 \text{ min}$, $C_{\text{dye}} = 10 \text{ mg/L}$, pH_i 7.3, $\kappa = 3.81 \text{ mS/cm}$ (2.02 g/L NaCl), $Q = 350 \text{ mL/min}$	[61]
Real textile wastewater	Semi-continuous flow packed bed electrochemical reactor, cylindrical Fe cathode (height = 50 cm and diameter = 4.3 cm), Fe hexagonal wire netting sheet anode (height = 50 cm and diameter = 1.6 cm), $C_{\text{COD}} = 1953 \text{ mg/L}$, pH 9.01, $\kappa = 739 \mu\text{S/cm}$, $j = 20\text{--}50 \text{ mA/cm}^2$, pH_i 3–9.01, 0.05 M Na_2SO_4 , $t_{\text{EC}} = 10\text{--}90 \text{ min}$, $Q = 240\text{--}930 \text{ mL/min}$	$R_{e,\text{COD}} = 90.84\%$, $R_{e,\text{color}} = 99\%$ $j = 20 \text{ mA/cm}^2$, pH_i 9.01, $t_{\text{EC}} = 60 \text{ min}$, $Q = 540 \text{ mL/min}$ and $\text{ENC} = 69.1 \text{ kWh/kg COD}$ (for semi-continuous flow) $R_{e,\text{COD}} = 98.5\%$, $R_{e,\text{color}} = 93.8\%$ $j = 20 \text{ mA/cm}^2$, pH_i 9.01, $t_{\text{EC}} = 60 \text{ min}$ $Q = 20 \text{ mL/min}$, steady-state time = 30 min and $\text{ENC} = 51.8 \text{ kWh/m}^3$ (for continuous flow).	[62]
Real textile wastewater	N_e : one carbon steel anode and one stainless steel cathode (tube-type electrodes), connection mode: MP-P, $S_e = 300.5 \text{ cm}^2$ (anode) and 497.4 cm^2 (cathode), $C_{\text{COD}} = 450\text{--}766 \text{ mg/L}$, Turbidity = 33.2–78.1 NTU, $\text{SS} = 28\text{--}146 \text{ mg/L}$, pH_i 7.7–10.8, $j = 142\text{--}426 \text{ A/m}^2$; $\kappa = 1551\text{--}2052 \mu\text{S/cm}$, $Q = 1.2\text{--}277 \text{ cm}^3/\text{s}$, $\tau = 0.8\text{--}1.7 \text{ min}$	$R_{e,\text{color}} = 96\%$ $R_{e,\text{COD}} = 81\%$, $R_{e,\text{SS}} = 99\%$ $\tau = 0.8 \text{ min}$, $j = 426 \text{ A/m}^2$, $Q = 1.2 \text{ cm}^3/\text{s}$	[63]
Real textile wastewater (dyeing and finishing mill)	N_e : eight cast Fe anodes and eight stainless steel cathodes, connection mode: MP-P, $S_e = 3744 \text{ cm}^2$, pH_i 4–9.4, $j = 13.35\text{--}106.8 \text{ mA/cm}^2$, $\kappa = 3300 \mu\text{S/cm}$, $C_{\text{COD}} = 1012 \text{ mg/L}$, $Q = 0.5\text{--}2 \text{ L/min}$, $\tau = 35.36\text{--}8.84 \text{ min}$	$R_{e,\text{COD}} = 63\%$ pH_i 7, $j = 53.4 \text{ mA/cm}^2$, $Q = 1 \text{ L/min}$, $\tau = 18 \text{ min}$, $\text{OC} = 0.34 \text{ \$/m}^3$	[64]

* j : current density, $R_{e,p}$: removal efficiency, Q : flow rate, τ : residence time in EC reactor, S_e : effective electrode surface area, N_e : electrode number, κ : conductivity, ENC: energy consumption, OC: operating cost, t_{EC} : operating time, pH_i : initial pH, MP-P: monopolar parallel, BP-S: bipolar series.

Table 3
Comparisons of operating costs and treatment performances of textile wastewaters by the treatment processes except for the EC process.

Treatment process	Experimental and optimum conditions	Performance of treatment and operating cost	Ref.
Photo-Fenton process	<i>Characterization of wastewater:</i> real dyeing wastewater (pH 12.5, Turbidity = 310 NTU, $C_{\text{COD}} = 1636 \pm 47$ mg/L, $C_{\text{TOC}} = 638 \pm 8$ mg/L). <i>Experimental conditions:</i> pH 2–8, $[\text{Fe}^{2+}] = 0.01\text{--}0.09$ g/L, $[\text{H}_2\text{O}_2] = 1\text{--}7$ g/L, $T = 35^\circ\text{C}$ and mean daily solar irradiation = 13.6 MJ/m ² . <i>Optimum conditions:</i> pH 3, $[\text{Fe}^{2+}] = 0.05$ g/L, $[\text{H}_2\text{O}_2] = 6$ g/L, irradiation time = 360 min	$R_{\text{e,color}} > 99\%$, $R_{\text{e,COD}} = 94\%$, $R_{\text{e,TOC}} = 85\%$ OC = 6.85 \$/m ³ (solar irradiation) OC = 17.95 \$/m ³ (artificial irradiation)	[65]
Solar photo-Fenton process	<i>Characterization of wastewater:</i> synthetic polyester dyeing wastewater (pH 8.3, $\kappa = 2.9$ mS/cm, $C_{\text{COD}} = 517.9$ mg/L, $C_{\text{DOC}} = 143.1$ mg/L and $C_{\text{BOD}} = 130.7$ mg/L), Synthetic acrylic dyeing wastewater (pH 6.8, $\kappa = 1503$ mS/cm, $C_{\text{COD}} = 828.1$ mg/L, $C_{\text{DOC}} = 334.1$ mg/L and $C_{\text{BOD}} < 1$ mg/L), Synthetic cotton dyeing wastewater (pH 11.4, $\kappa = 23.2$ mS/cm, $C_{\text{COD}} = 350$ mg/L, $C_{\text{DOC}} = 117.5$ mg/L and $C_{\text{BOD}} = 77.5$ mg/L). <i>Experimental conditions:</i> pH 2–8, $[\text{Fe}^{2+}] = 0\text{--}350$ mg/L, $[\text{H}_2\text{O}_2] = 0.625\text{--}20$ g/L, light flux = 7–500 W/m ² and irradiation time = 120 min, pH 3.5 and $T = 50^\circ\text{C}$. <i>Optimum conditions:</i> $[\text{H}_2\text{O}_2] = 6.5, 3.75$ and 0.938 g/L for acrylic, cotton and polyester dyeing; $[\text{Fe}^{2+}] = 113.8, 112.5$ and 131.2 mg/L for acrylic, cotton and polyester dyeing; light flux = 500 W/m ²	$R_{\text{e,color}} = 98\text{--}99\%$ (for all wastewaters) $R_{\text{e,COD}} = 72\%$ (acrylic), 55.4% (polyester), and 30.1% (cotton) OC = 5.8 €/m ³ (for acrylic dyeing) OC = 3.5 €/m ³ (for cotton dyeing) OC = 1 €/m ³ (for polyester dyeing)	[66]
Coagulation (P-1: FeSO ₄ and P-2: FeCl ₃) Fenton oxidation (P-3) Fenton-like oxidation (P-4) Ozone oxidation (P-5) Peroxon process (O ₃ + H ₂ O ₂) (P-6)	Real textile wastewater (biologically pre-treated textile industry effluent), pH 7.8, $\kappa = 6.14$ mS/cm, $C_{\text{COD}} = 160$ mg/L, $C_{\text{color}} = 0.193$ (per cm at 600 nm). <i>Experimental conditions:</i> coagulant dosage: 50–700 mg/L (P-1 and P-2), H ₂ O ₂ and FeSO ₄ or FeCl ₃ dosages: 100–500 mg/L and 100–500 mg/L and pH 2–7 (P-3 and P-4), time = 5–90 min, pH 3–11, O ₃ dosage = 20 mg/min (P-5), H ₂ O ₂ dosage = 5–25 mg/L and time 2–14 min (P-6). <i>Optimum conditions:</i> 450 mg/L FeSO ₄ and 350 mg/L FeCl ₃ (P-1 and P-2), FeSO ₄ (400 mg/L H ₂ O ₂) (P-3), 350 mg/L FeCl ₃ (200 mg/L H ₂ O ₂) (P-4), 360 mg/L O ₃ (P-5), 360 mg/L O ₃ and 5 mg/L H ₂ O ₂ (P-6)	P-1: $R_{\text{e,COD}} = 62\%$, $R_{\text{e,color}} = 99\%$ OC = 5.02 \$/m ³ P-2: $R_{\text{e,COD}} = 64\%$, $R_{\text{e,color}} = 91\%$ OC = 0.19 \$/m ³ P-3: $R_{\text{e,COD}} = 78\%$, $R_{\text{e,color}} = 95\%$ OC = 0.59 \$/m ³ P-4: $R_{\text{e,COD}} = 64\%$, $R_{\text{e,color}} = 71\%$ OC = 0.57 \$/m ³ P-5: $R_{\text{e,COD}} = 43\%$, $R_{\text{e,color}} = 97\%$ OC = 4.94 \$/m ³ P-6: $R_{\text{e,COD}} = 54\%$, $R_{\text{e,color}} = 99\%$ OC = 5.02 \$/m ³	[67]
P: Process Fenton (P-1) Coagulation/ Flocculation + Fenton (P-2) Fenton + Coagulation/ Flocculation (P-3)	<i>Characterization of wastewater:</i> synthetic polyester dyeing wastewater (pH 8.3, $C_{\text{COD}} = 517.9 \pm 7.3$ mg/L, $C_{\text{DOC}} = 143.1 \pm 0.7$ mg/L, $C_{\text{BOD}} = 130.7 \pm 2.8$ mg/L). <i>Experimental conditions:</i> For P-1 (operating time = 0–120 min, $[\text{Fe}^{2+}] = 275\text{--}400$ mg/L and $[\text{H}_2\text{O}_2] = 1\text{--}10$ g/L), for P-2 ($T = 10\text{--}70^\circ\text{C}$, $[\text{Fe}^{2+}] = 0\text{--}241.3$ mg/L, operating time = 0–120 min and $[\text{H}_2\text{O}_2] = 500\text{--}1500$ mg/L), for P-3 ($[\text{Fe}^{2+}] = 0\text{--}241.3$ mg/L, operating time = 0–120 min and $[\text{H}_2\text{O}_2] = 500\text{--}1500$ mg/L). <i>Optimum conditions:</i> $[\text{H}_2\text{O}_2]$ and $[\text{Fe}^{2+}] = 2.5$ g/L and 350 mg/L (P-1), 1 g/L and 191.3 mg/L (P-2), 1.25 g/L and 350 mg/L (P-3), pH 3.5 and $T = 50^\circ\text{C}$	$R_{\text{e,COD}} = 57.3\%$, $R_{\text{e,BOD5}} = 52\%$, $R_{\text{e,DOC}} = 55.7\%$, OC = 2.7 €/m ³ (for the P-1). $R_{\text{e,COD}} = 79.2\%$, $R_{\text{e,BOD5}} = 61.9\%$, $R_{\text{e,DOC}} = 75.7\%$, OC = 3.1 €/m ³ (for the P-2). $R_{\text{e,COD}} = 78.9\%$, $R_{\text{e,BOD5}} = 73.3\%$, $R_{\text{e,DOC}} = 82.9\%$, OC = 1.3 €/m ³ (P-3).	[68]
Fenton (P-1) Coagulation/ Flocculation + Fenton (P-2) Fenton + Coagulation/ Flocculation (P-3) Biological oxidation + Fenton (P-4)	<i>Characterization of wastewater:</i> real cotton dyeing wastewater (pH 10, $\kappa = 11.5$ mS/cm, $C_{\text{COD}} = 495$ mg/L, $C_{\text{DOC}} = 174.7$ mg/L, $C_{\text{BOD}} = 127.5$ mg/L) <i>Experimental conditions:</i> For P-1: (operating time = 60 min, $T = 50^\circ\text{C}$, pH 3.5, $[\text{Fe}^{2+}] = 37.5\text{--}300$ mg/L and $[\text{H}_2\text{O}_2] = 0.938\text{--}10$ g/L). For P-2: (200 mg/L Fe^{2+} and flocculant: 0.5 mg/L Magnafloc 155 at coagulation/flocculation stage; 98.3 mg/L Fe^{2+} , 500 mg/L H ₂ O ₂ , pH 3.5, operating time = 60 min and $T = 50^\circ\text{C}$ at Fenton stage). For P-3: (18.8–37.5 mg/L Fe^{2+} and 2.5 mg/L Magnafloc 155 at coagulation/flocculation stage, 0.625–1.25 mg/L H ₂ O ₂ , pH 3.5, $T = 50^\circ\text{C}$, operating time = 60 min at Fenton stage). For P-4: (pH 3.5, $T = 50^\circ\text{C}$, 1.25 g/L H ₂ O ₂ and 37.5 mg/L Fe^{2+}) <i>Optimum conditions:</i> $[\text{H}_2\text{O}_2] = 1.25$ g/L and $[\text{Fe}^{2+}] = 37.5$ mg/L for P-1 Above experimental conditions for P-2 $[\text{H}_2\text{O}_2] = 0.938$ g/L and $[\text{Fe}^{2+}] = 37.5$ mg/L for Fenton stage for P-3 $[\text{H}_2\text{O}_2] = 0.938$ g/L and $[\text{Fe}^{2+}] = 37.5$ mg/L, last four cycles for SBR (sequential batch reactor) for P-4:	For P-1: $R_{\text{e,COD}} = 53\%$, $R_{\text{e,BOD5}} = 33.6\%$, $R_{\text{e,DOC}} = 52.8\%$, $R_{\text{e,color}} = 94.3\%$, OC = 1.4 €/m ³ . For P-2: $R_{\text{e,COD}} = 70.8\%$, $R_{\text{e,BOD5}} = 47.6\%$, $R_{\text{e,DOC}} = 66.1\%$, $R_{\text{e,color}} = 99.5\%$, OC = 1 €/m ³ . For P-3: $R_{\text{e,COD}} = 62.6\%$, $R_{\text{e,BOD5}} = 47.5\%$, $R_{\text{e,DOC}} = 64.4\%$, $R_{\text{e,color}} = 96.6\%$, OC = 1 €/m ³ . For P-4: $R_{\text{e,COD}} = 63\%$, $R_{\text{e,BOD5}} = 49.5\%$, $R_{\text{e,DOC}} = 64.6\%$, $R_{\text{e,color}} = 96.5\%$, OC = 2.3 €/m ³ .	[69]
Electrochemical Fenton (EF) Chemical Fenton (CF)	<i>Characterization of wastewater:</i> real textile wastewater (pH 6.5, $\kappa = 0.5$ mS/cm, $C_{\text{COD}} = 1800$ mg/L, $C_{\text{color}} = 1080$ ADMI, $C_{\text{BOD5}} = 320$ mg/L) <i>Experimental conditions:</i> For EF: (Fe anode and cathode ($S_e = 40$ cm ² , $d = 3$ cm)), operating time = 0–60 min, $[\text{H}_2\text{O}_2] = 989\text{--}2472$ mg/L, $j = 3.75\text{--}11.25$ mA/cm ² and pH = 3. For CF: $[\text{Fe}^{2+}] = 50\text{--}350$ mg/L, operating time = 0–150 min and $[\text{H}_2\text{O}_2] = 989\text{--}2472$ mg/L. <i>Optimum conditions:</i> $[\text{H}_2\text{O}_2] = 1978$ mg/L (for CF and EF), $[\text{Fe}^{2+}] = 250$ mg/L (CF), $j = 8.75$ mA/m ² (EF), operating time = 60 min (EF) and 120 min (CF)	$R_{\text{e,color}} = 72.9\%$ and $R_{\text{e,COD}} = 70.6\%$, OC = 17.56 \$/kg COD (for EF). $R_{\text{e,color}} = 52.3\%$ and $R_{\text{e,COD}} = 51.2\%$, OC = 8.6 \$/kg COD (for CF)	[70]
Electro-oxidation process	<i>Characterization of wastewater:</i> real textile wastewater (pH 8.84, $\kappa = 1.94$ mS/cm, 2175 color (ADMI unit), 1354 mg/L COD, 183.4 mg/L). <i>Experimental conditions:</i> $j = 20\text{--}80$ mA/cm ² , NaCl = 0–20 g/L, operating time = 0–240 min, pH 8–11, $T = 25\text{--}40^\circ\text{C}$ <i>Optimum conditions:</i> $j = 80$ mA/cm ² , NaCl = 20 g/L, pH 8, $T = 40^\circ\text{C}$, operating time = 240 min	$R_{\text{e,color}} = 98.5\%$ $R_{\text{e,COD}} = 51.5\%$ OC = 643 \$/kg COD	[71]

COD, TOC and turbidity remained in the treated wastewater were under the permissible limit of discharge standard. However, the legal discharge limit to receiving environment (i.e. streams and rivers) for the textiles industry is COD of 250 mg/L for 2 h composite sample according to the Turkish legislation. From results of the above experiments, direct dischargeable

effluents (the national water pollution regulations) were not obtained by reducing the initial COD concentration of 2000 mg/L to <250 mg/L. To comply this, it was needed additional process(es) (i.e. biological oxidation, Fenton, photo-Fenton, photoelectro-Fenton, and electrooxidation processes) along with the CEC process.

3.4. Comparison of economic and treatment performance of the results

Many studies were reported for the treatment of synthetic and real textile wastewater by electrocoagulation in the literature [10,15,53]. In most cases, each paper mentioned in Table 2 was focused on one specific dye dissolved in pure water and the effects of various operating parameters (e.g. pH, initial concentration, current density, electrolyte concentration, electrode type (Fe, Al), temperature, electrode gap) on the removal efficiency were evaluated, in particular regarding the COD level and the absorbance (color) at the maximum wavelength. Very satisfactory results were usually reported in most cases with color removal efficiency ranging between 95% and 100%. Although, there have been many studies on the treatments of real textile and dyeing wastewater in a batch mode of operation by the EC process in the literature, few similar studies were carried out in a batch mode of operation with the CEC process. Therefore, the present study was performed for the treatment of the real textile wastewater in the CEC process. Treatments of textile wastewaters with the CEC process in the literature are summarised in Table 2 [24,25,29,54–64]. As seen in Table 2, most of the studies were dealt with the synthetic dyeing wastewater and electrodes such as Fe and stainless steel plate anodes. Of 94% was related to removal of color efficiency except for [29] and over 50% was mainly concerned with removal of COD. In addition, color removal efficiency of 94% and COD removal efficiency of 63% were achieved with the real textile wastewater in the CEC process. Other processes for treatment of textile wastewater expect for EC process with respect to removal performances and operating cost are presented in Table 3 [65–71]. High COD (60%) and color (95%) removal efficiencies were obtained for synthetic and real dyeing wastewaters (Table 3). Removals of color (94%) and COD (62%) were better with coagulation/flocculation plus Fenton, Fenton plus coagulation/flocculation, photo-Fenton processes and satisfied the limit values of discharge standards for treated textile wastewaters. Fenton plus coagulation/flocculation or coagulation/flocculation plus Fenton (color of $\geq 97\%$ and COD of $\geq 63\%$), and photo-Fenton (color of $\geq 99\%$ and COD of $\geq 94\%$) processes were found to be more effective for the treatment of real textile wastewaters in a batch mode of operation. Operating costs were varied between 1 $\$/\text{m}^3$ [69] and 6.85 $\$/\text{m}^3$ [65] (Table 3). When the results in Table 3 for Al and Fe anodes were compared to the present study in terms of removal efficiencies and cost, this study with the CEC process gave better performances (85% and 1.562 $\$/\text{m}^3$ for Fe and 77% and 1.851 $\$/\text{m}^3$ for Al).

4. Conclusions

In this study, the effects of operating parameters (current density, inlet flow rate, operating time, and loading COD or TOC) from the real textile wastewater by the CEC process on COD, TOC and turbidity removal efficiencies were investigated. The optimum operating conditions were determined to be inlet flow rate of 0.010 L/min, operating time of 80 min (hydraulic retention time of 350 min), and current density of 65 A/m² for both Fe and Al electrodes. COD, TOC and turbidity removal efficiencies at the optimum conditions were 85%, 76% and 95% for Fe electrode, and 77%, 72% and 95% for Al electrode. Values of COD, TOC and turbidity after the treatment at the optimum operating conditions reduced to 260 mg/L, 99 mg/L and 153 NTU for Fe electrode and 440 mg/L, 117 mg/L and 214 NTU for Al electrode and these values were under the permissible limit to discharge into the sewage. Operating costs at the optimum conditions were calculated as 1.562 $\$/\text{m}^3$ (7.282 $\$/\text{kg COD}$) for Fe electrode and 1.851 $\$/\text{m}^3$ (14.257 $\$/\text{kg COD}$) for Al electrode. According to the results, treatment of the wastewater with Fe electrode in the CEC process was 1.95 times more efficient in terms of removed $\$/\text{kg COD}$ than that of Al electrode.

Removals of COD (1000–8000 mg/L), TOC (243–1942 mg/L) and turbidity (1113–8895 NTU) from spent dyeing bath wastewater were obtained as 80–20%, 74–16%, 96–36% for Al electrode and 90–21%, 83–18%, and 93–30% for Fe electrode, respectively. The removal efficiencies were decreased significantly in the wastewater containing COD of >2000 mg/L since dye molecules and polymeric chemical species in the wastewater adsorbing the electrode surfaces caused passivation or deactivating of the electrodes. The operating cost for both electrodes increased with increased flow rate per m³ treated wastewater but the cost decreased with per removed kg COD. It can be concluded that the CEC process was found to be an efficient alternative method with respect to the other treatment methods employed for the treatment of the similar dyehouse effluents.

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