

Sequential Treatment of Food Industry Wastewater by Electro-Fenton and Electrocoagulation Processes

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The food industry is one of industrial activity that use large amounts of water and as a consequence of various consumption stages in the process, the quantity and the composition of wastewater can be significantly ranged. In this study, high strength of food industry wastewater was treated by electro-Fenton (EF) and electrocoagulation (EC) process was sequentially applied to remove total organic carbon (TOC) from wastewater. During EF process, H₂O₂ amount was periodically added based on the calculated amount of released iron content from anode electro-dissolution. Then, EC process was further carried out to finalize the sequential treatment process. Optimum reaction time for EF process was initially investigated then the ideal current density value was determined for EC process with iron plate electrodes. This tandem sequential treatment processes resulted in 58.7 % TOC, 93.9 % total phosphate, 82.8 % TSS and 74.4 % turbidity reduction at 120 min EF (5mA/cm²) and 180 min EC (15mA/cm²) by applying optimum operation conditions. The electrode and energy consumptions were calculated as 13.43 kg/m³ and 31.26 kWh/m³, respectively in EF+EC processes.

Keywords: Food industry wastewater, electrocoagulation, electro-Fenton, sequential treatment.

1. INTRODUCTION

Wastewater streams are generally discharged to receiving media and this situation reduces not only water quality of discharge channel but also has a harmful effect on aquatic organisms, ecosystem and human's life. Therefore, recent studies are concerned with treatment solutions for wastewaters originating from food, textile, chemical, pharmaceutical etc. industries. Food industry has large amounts of water consumption for many purposes like production, cleaning, transportation, and refrigeration. According to the product variety; the source, quantity and the composition of wastewater range significantly. The main streams of wastewater in pastry industry are from washing the egg-crusher, blender, filler and wooden box [1]. The pastry wastewater effluent's characteristic consists in

large amounts of total suspended solids (TSS), different nitrogen compounds, fats, proteins, oils, and resistant organic pollutants, phosphorus, chlorine and other chemicals used in washing and sanitizing purposes [2]. Depending on the type of pollutants present in wastewater, several treatment methods have been applied to treat wastewater as adsorption, electrochemical, biological processes and advanced oxidation processes etc. [3]. Biological processes are commonly used for the treatment of the wastewater containing high concentrations of biodegradable organic matter. Although biological processes are effective, they may not be feasible due to long hydraulic retention time and large area requirement [4, 5]. Since resistant organic pollutants are fragmented, it is difficult and not feasible to apply traditional treatment methods. Therefore, the methods generally require modification with other processes.

EC process has been successfully conducted for the treatment of food effluents at industrial scale but was not sufficient meeting the discharge limits without applying co-treatment [6]. Some studies have been reported as combination of EC processes with membrane technologies, thermolysis and chemical coagulation [7-9].

Advanced oxidation processes supported by electrochemical processes (as EF, photo-EC and photo-EF) have also been recently considered as an alternative methods for wastewater treatment [10-12]. Using a synergetic effect, removal of resistant organic pollutants efficiency was maximized with minimal operating costs, versatility, high energy efficiency and simple equipment design. Electrochemical advanced oxidation processes can also be performed in room temperature and pressure. Combination of electrochemical and advanced oxidation processes easily oxidize and lead to mineralize most organic and inorganic pollutants to produce H_2O , CO_2 and inorganic ions by production of hydroxyl radicals [13]. Hydroxyl radicals can be produced by various methods i.e. chemical, electrochemical, photo assisted electrochemical, photocatalysis, Fenton, ozonation [10].

1.1. Electrocoagulation

Metal electrodes connected a direct current (DC) power source are dipped in a solution and the electrical current passes through the electrodes in an electrochemical reactor. Thus, various processes occur in electrochemical reactor: formation of anodic metal and metal hydroxide cations in aqueous phase by electrolytic reactions at the electrode surface; adsorption of colloidal or soluble pollutants onto the surface of metal hydroxides; removal of pollutants, adhesion to bubbles and eventually sedimentation [14].

On the other hand, when pH is acidic, the electrode is attacked by H^+ ions which lead to electrodes' dissolution, then oxygen in the aqueous phase oxidizes Ferrous ions (Fe^{2+}) to ferric ions. Meanwhile, the oxygen evolution reaction occurs at anode. The Fe^{2+} and Fe^{3+} ions are released in aqueous phase, such as hydrated and hydrolyzed monomeric and polymeric iron forms i.e. $Fe(OH)_2^+$, $FeOH^{2+}$, $Fe_2(OH)_2^{4+}$, $Fe(OH)_4^-$, $Fe(H_2O)_2^+$, $Fe(H_2O)_5OH^{2+}$, and $Fe(H_2O)_4(OH)_2^+$ [15]. Negative charged colloidal particles in effluent get neutralize with hydroxyl species and settle due to the agglomeration of heavy molecules. Organic species are also eliminated through sweeping process by mingling in the way of settling heavily mass chemicals [14].

1.2. Electro-Fenton

When H_2O_2 is added into the aqueous phase during the electrocoagulation process by using Fe anode as the Fe^{2+} source, several competing reactions which contain Fe^{2+} , Fe^{3+} , H_2O_2 are involved and eventually forming hydroxyl radicals and this process is defined as electro-Fenton [16]. The hydroxyl radicals are capable of quickly decomposing organic substrates (RH) and cause chemical degradation of these organic compounds. Meanwhile, ferrous ions are depleted as rapidly as they are produced. On the other hand, ferrous ions can also react with hydroxyl radicals in aqueous solution [17]. For that reason, more ferrous ion amount is required to maintain hydroxyl radical production or this process should be carried out intermittently.

Several integrated or sequential treatment processes were applied for different wastewater resources. Kumar et al. (2009) have applied catalytic thermal treatment and coagulation to treat desizing wastewater [8]. Ghanbari and Moradi (2015) have carried out the comparative study of electrocoagulation, electrochemical Fenton, electro-Fenton and peroxi-coagulation for decolorization of real textile wastewater [18]. Gerek et al. (2017) also studied the energy and removal efficiency of electrochemical wastewater treatment for leather industry [19].

In this study, sequential and integrated processes including EF and EC were applied for the treatment of pastry industry wastewater. Effect of process period of EF and current density of EC process using iron plate electrodes were examined in terms of TOC removal, electrode and energy consumption.

2. MATERIALS AND METHODS

2.1. Materials

The cake industry wastewater used for experiments was provided from a food industry complex in Gebze, TURKEY. The wastewater was stored in a cold room (+4 °C) to avoid of any decomposition in wastewater quality. The raw wastewater was initially filtrated through two sized screen filters and then filtrated using coarse filtration as pretreatment processes, since it included large size solids. After applying screen filtrating, total suspended solids (TSS) were found to be 410 mg/L. After coarse filtrating, this value was dropped to 87 mg/L. After the removal of large particles with pretreatment steps, the main characteristics of the wastewater were analyzed and given in Table 1. Wastewater conductivity level allowed the process to be performed without addition any electrolyte chemical all through experiments. H_2O_2 (35% w/w) were used as the oxidizing reagent and pH values were adjusted with H_2SO_4 and NaOH in the experiments.

Table 1. Characteristic of cake industry wastewater

| Parameters | Quantity |
|------------|---------------------|
| pH | 5.1 ± 0.1 |
| COD | 3600 ± 100 mg/L |

| | |
|------------------|--------------------|
| BOD ₅ | 1100 ± 50 mg/L |
| TOC | 1100 ± 10 mg/L |
| TSS | 87 ± 1 mg/L |
| Conductivity | 1.41 ± 0.01 mS/cm |
| Turbidity | 67.2 ± 0.2 NTU |
| TP | 5.1 ± 0.05 mg/L |
| TN | 10.6 ± 0.1 mg/L |
| Ca ²⁺ | 108.05 ± 0.05 mg/L |
| Fe ²⁺ | 6.3 ± 0.1 mg/L |
| Mg ²⁺ | 8.3 ± 0.1 mg/L |

2.2. Experimental

EF and EC processes were implemented in the batch electrochemical reactor which made-up of polypropylene (PP) with a capacity of 1 L and the working volume of the effluent was 0.5 L as shown in Fig. 1.

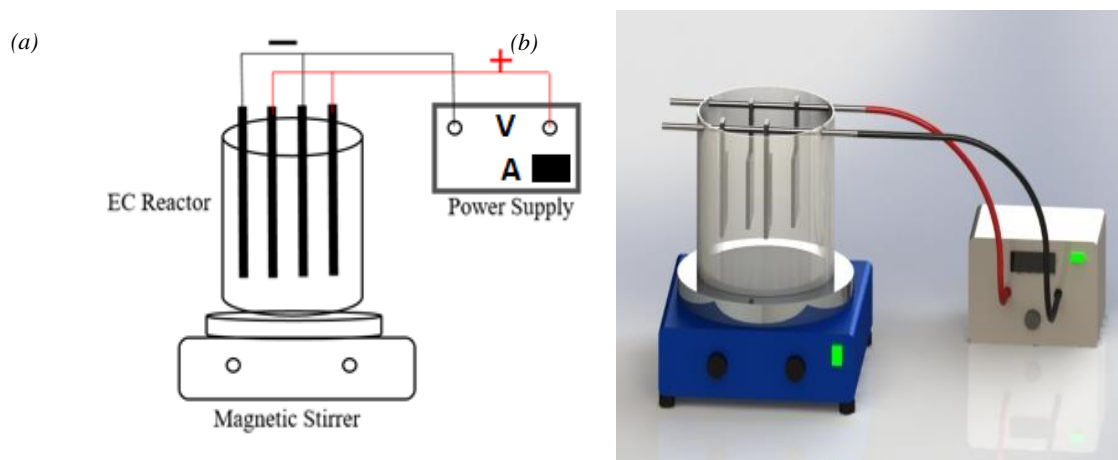


Figure 1. (a) Schematic diagram, (b) 3D view of experimental set-up.

Table 2. Properties of electrochemical reactor (ECR)

| ELECTRODE | | REACTOR | | POWER SUPPLY | |
|---|-------------|-----------------|---------------|---------------|--------|
| Material | Iron | Material | Polypropylene | Voltage range | 0-20 V |
| Shape | Rectangular | Operating mode | Batch | Current range | 0-2 A |
| Size (mm) | 45 *70 | Shape | Cylinder | | |
| Thickness (mm) | 3 | Volume (L) | 1 | | |
| Electrode gap (mm) | 20 | Dimensions (mm) | 100*110 | | |
| Plate plan | Parallel | | | | |
| Effective electrode surface area (mm ²) | 13500 | | | | |
| Number of electrodes | 4 | | | | |

Both anode and cathode electrodes made from iron were connected with direct current (DC) power supply source (NETES 6303D) in parallel mode. Uniform distribution in the reactor was sustained by magnetic stirrer. The pH of the influent was adjusted to the desired value for EF process using H_2SO_4 solutions. The electrodes used in the processes were cleaned manually with 35% HCl solution followed by washing with distilled water and dried after each experiment prior to their use. The properties of electrochemical reactor (ECR) were given in Table 2.

EF process was initially performed in order to determine the amount of released iron ions into the solution by different time intervals using 5 mA/cm^2 current density and at initial pH 2. Iron concentration of raw wastewater was measured as 6.3 mg/L . The iron content of wastewater increased to 32.4, 45.4 and 296.5 mg/L after applying the current in the reactor 0.5, 1 and 30 minutes, respectively.

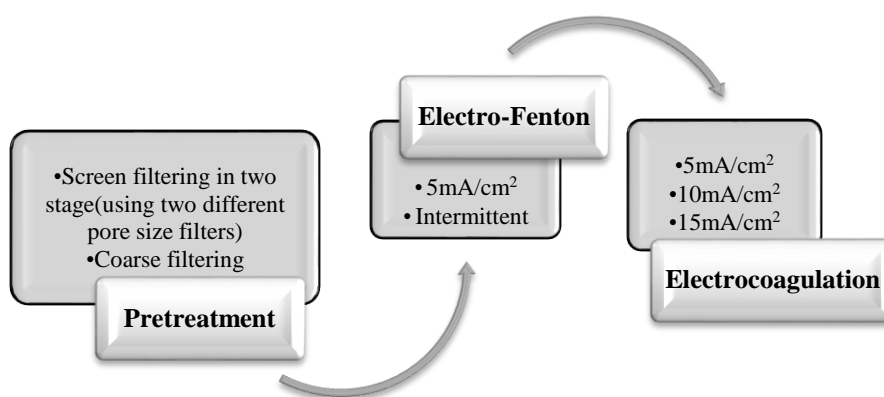


Figure 2. Experimental steps of cake wastewater treatment

The results indicated that the released iron content was sufficient for performing Fenton process in 1 min electro-dissolution time. In response to this iron value, H_2O_2 concentration was adjusted considering optimum Fe: H_2O_2 ratio as 1:10 based on the literature studies [20, 21]. EF process was then maintained in a pulsed current feeding by performing 1 min current on and 5 minutes current off cycles. These cycles were repeated 10 and 20 times for 60 and 120 min EF time, respectively. Necessary amount of H_2O_2 was added at the beginning of each current off cycles by adjusting the amount in stoichiometric ratio (1:10). EF process was applied during 60 and 120 min periods at pH 2. Prior to EC process, the pH was adjusted to the value of 7.5 ± 0.3 to get effective coagulation process. The complete removal sequential process was maintained by either “60 min EF followed by 240 min EC” or “120 min EF followed by 180 min EC” periods. On the other hand, EC process was performed in 3 different current densities such as 5, 10 and 15 mA/cm^2 . The treatment procedures used in the experiments are summarized in Fig. 2.

2.3. Methods

Samples were periodically taken from the EF and EC reactor at certain intervals. Each sample taken was filtered using $0.45 \mu\text{m}$ pore size filters and centrifuged at 6000 rpm for 10 min to separate

flocs (Hettich/EBA 20) for EF and EC processes, respectively. pH and conductivity were also measured by a multimeter (Mettler Toledo/Seven Go Duo). COD, TSS and BOD₅ values were determined by according to the Standard Methods 5220C, 2540D and 5210B, respectively [22]. Total organic carbon (TOC) of the samples was measured by a TOC analyzer (Shimadzu TOC-L). The concentrations of the metal ions in samples were analyzed by using Optical Emission Spectroscopy (Perkin Elmer Optima 7000 DV). The absorption spectrums of EF effluent samples were scanned by a UV–Vis spectrophotometer (Hach Lange, DR 6000). Turbidity was measured using a Turbidimeter (Hach/2100 P). TN values were measured by photometric method using Laton LCK 138 (Hach-Lange).

3. RESULTS AND DISCUSSION

3.1. Effect of EF process period on degradation of organic material

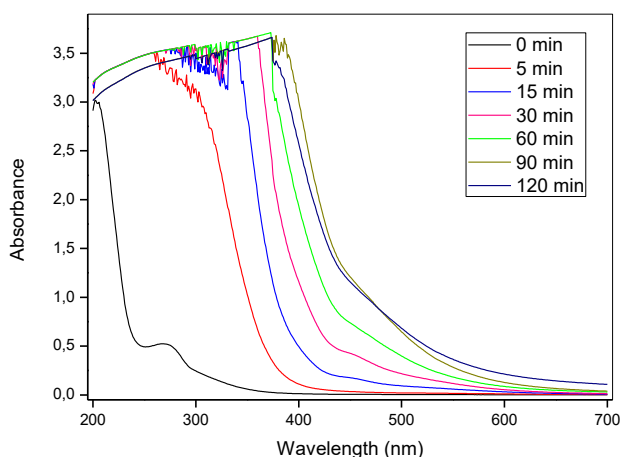


Figure 3. UV-Vis spectrum of EF process effluent (conditions; CD: 5 mA/cm², initial TOC: 1107 mg/L)

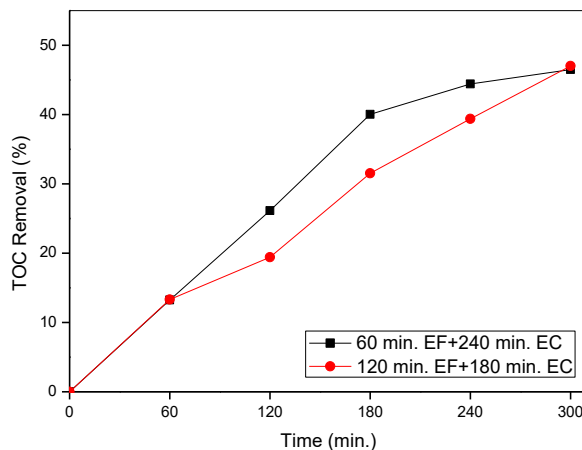


Figure 4. Implementation of sequential EF + EC process on the TOC removal efficiency (conditions; EF current density: 5 mA/cm², EC current density: 10 mA/cm², pH: 7)

EF process was initially performed in order to determine organic degradation in 60 min and 120 min cycles. 0.5 mL H₂O₂ (35% w/w) was added according to calculated Fe:H₂O₂ ratio (1:10) at the beginning of each current off cycles. The degradation efficiency of EF process in 120 min was implemented and the UV–Vis absorption spectrums of effluent samples were shown in Fig. 3.

The results showed in Fig. 3 that influent wastewater's spectrum stayed under visible range however, spectrums were observed shifting to visible range during the reaction. This indicates that organic matter degrades to molecules, which can be detected under visible range. Degradation was effectively progressed till 60 min and slowed down at final 120 min and no significant difference of organic material degradation spectrum was observed between 60 and 120 min. Meanwhile TOC removal efficiency was compared using sequential implementation of EF and EC processes (Fig. 4). TOC results clearly indicated that implementation of EC process after EF in 60 min increased the removal efficiency. However, EF process during 120 min had economic benefits because of pulsed current feeding. Consequently, “120 min EF and 180 min EC” process implementation was preferred in the benefit of process economy due to high current values used in EF process done for 60 minutes. The cost calculations were given in the following section.

3.2. Effect of current density on EC process efficiency

Current density (CD) is one of the important parameter for EC process. It increases the metal hydroxide dosage ratio and the bubble production rate, size and floc growth, which can affect the efficiency of the EC process [14, 23]. Afterward EF process was applied in 120 min, the influence of current density on the TOC removal of EC process was investigated. TOC removal was increased by increasing the current density (Fig. 5). The maximum 58.7 % TOC removal was obtained when CD was 15 mA/cm² in 120 min EF + 180 min EC process. However, 10 mA/cm² followed similar efficiency with 15 mA/cm².

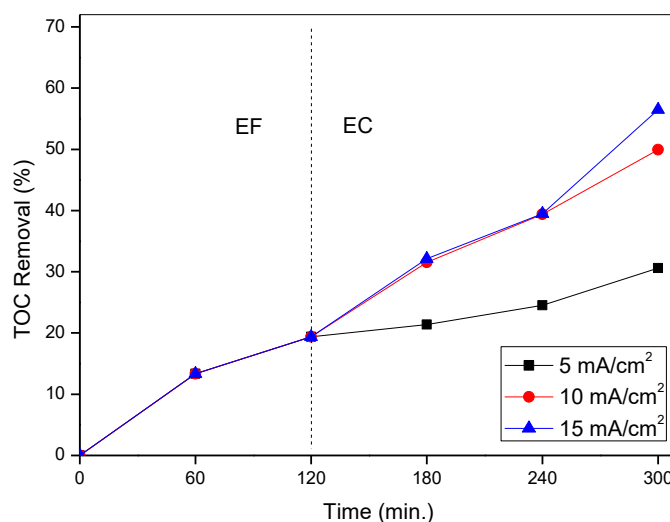


Figure 5. Effect of current density on the TOC removal efficiency (conditions; 120min EF (5mA/cm²) + 180min EC (pH:7))

Moreover, the main characteristics of effluent samples were determined after implementation of optimum conditions and initial-final values of these parameters were presented in Table 3. Based on obtained values, BOD₅/COD ratio was increased from 0.3 (slowly biodegradable) to 0.5 (easily biodegradable) in sequential processes, showing that the content of wastewater could easily be treated by biological treatment. This phenomenon was also proved in many research papers that advanced oxidation processes such as Fenton improved biodegradability of high strength or recalcitrant pollutants in wastewater [24, 18]. Baiju et al. (2018) and Chemlal et al. (2014) showed that biological treatment process was significantly achieved an improved removal rate for landfill leachate pretreated by AOP [25, 26]. Besides, the other parameters were also significantly decreased (Table 3).

Table 3. Process parameters before and after sequential treatment

| Parameters | Influent | Effluent |
|------------------|------------|------------|
| COD | 3700 mg/L | 1850 mg/L |
| BOD ₅ | 1100 mg/L | 900 mg/L |
| TOC | 1107 mg/L | 456.8 mg/L |
| TSS | 87 mg/L | 15 mg/L |
| Turbidity | 67.2 NTU | 17.2 NTU |
| TP | 5.1 mg/L | 0.3 mg/L |
| TN | 10.6 mg/L | 8.4 mg/L |
| Ca ²⁺ | 108.1 mg/L | 16.0 mg/L |
| Mg ²⁺ | 8.3 mg/L | 1.1 mg/L |

3.3. Energy and electrode consumption for cake industry wastewater

Mass of electrode material and energy consumption were calculated based on the optimum experimental conditions (120 min EF; 5mA/cm² and 180 min EC; 15mA/cm²). The electrode material usage and consumed electrical energy constitute significant percentage of operating cost of electrochemical processes. The following well-known Faraday Law equation was used to calculate theoretical electrode consumption for EF+EC processes [27, 28]:

$$m_{anode} = \frac{I \times t \times M_{electrode}}{z \times F} \quad (12)$$

m_{anode} : The amount of dissolved electrode material, g/L

I: Current intensity, A

t: Time, sec.

$M_{electrode}$: Molecular weight of electrode material, g/mol

z: Valence of metal ions, e⁻/mol

F: Faraday constant, 96500 Coulomb/mol

Energy consumption of the processes at the optimum experimental conditions was also calculated using the following equation [29, 30];

$$E = \frac{I \times U \times t}{V} \tag{13}$$

E: Consumed energy, kWh/m³

I: Current intensity, A

U: Voltage, V

t: Time, h

V: Volume of treated solution, m³

The theoretical iron electrode and energy consumption for first (EF) and second part (EC) of tandem treatment process were calculated using Eq. 12-13 and presented in Table 4. In EF process, electrode and energy consumptions were lower than EC process due to lower current density and pulsed time were used at EF reaction. Since both electrode and energy consumptions were calculated higher in EC process, each process cost efficiency was then solely calculated to estimate the total cost of efficiency. The results showed that total electrode and energy cost was calculated as 12 \$ per treated m³ wastewater in this study.

Table 4. Electrode and energy consumption for treatment of cake industry wastewater

| | | Electrode consumption | Energy consumption |
|------------------------------|--|------------------------|-------------------------|
| Sequential Treatment Process | EF (5 mA/cm ² , 120 min.) | 0.5 kg/m ³ | 6.1 kWh/m ³ |
| | EC (15 mA/cm ² , 180 min.) | 13.0 kg/m ³ | 25.2 kWh/m ³ |
| Total Consumption | | 13.5 kg/m ³ | 31.3 kWh/m ³ |
| Cost | | 9.5 \$/m ³ | 2.5 \$/m ³ |

The electrode and energy consumptions were calculated as 13.5 kg/m³ and 31.3 kWh/m³, respectively for sequential treatment process. The results found in this study were compared with some literature values (Table 5). It can easily be said that our results are compatible with values from the literature.

Table 5. The comparison of the results with some literature values

| Ref. | Type of solution | COD of the solution (mg O ₂ /L) | Process | Time (min) | Current Density (mA/cm ²) | Energy Consumption (kWh/m ³) | Electrode Consumption (kg/m ³) | COD removal % |
|------|--------------------|--|---------|------------|---------------------------------------|--|--|---------------|
| [18] | Textile wastewater | 1310 | EC | 40 | 4.76 | 3.58 | 0.464 (Fe) | 78.6 |
| | | | EF | 160 | 4.76 | 83.37 | 0.168 (Fe) | 64.2 |

| | | | | | | | | |
|------------|--------------------------|------|----|-----|----|------|----------|------|
| [19] | Tannery wastewater | 1024 | EC | 50 | 20 | 8.33 | -- (Al) | 82.0 |
| | | | EF | 40 | 20 | 6.92 | -- (Al) | 91.7 |
| This study | Food industry wastewater | 3700 | EC | 180 | 15 | 25.2 | 13 (Fe) | 50 |
| | | | EF | 120 | 5 | 6.1 | 0.5 (Fe) | |

4. CONCLUSION

In this study, high strength of cake industry wastewater was sequentially treated by electro-Fenton (EF) and electrocoagulation (EC) processes. The performance of sequential implementation of EF and EC processes was investigated by using different current densities and reaction times. The most effective and economic degradation were observed in the 120 min EF using 5mA/cm² pulsed current density and then 180 min EC using 15mA/cm² of current density.

Results of tandem sequential treatment processes indicated 58.7 % TOC, 93.9 % total phosphate, 82.8 % TSS and 74.4 % turbidity reduction at 120 min EF (pulsed 5mA/cm²) and 180 min EC (15mA/cm²). Additionally, BOD₅ /COD ratio was improved from 0.3 to 0.5 showing good biodegradability of cake wastewater. The electrode and energy consumptions were calculated as 13.5 kg/m³ and 31.3 kWh/m³, respectively.

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