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# Treatment of Dairy Industry Cleaning-in-Place Wastewater by Electrocoagulation Supported with Immersed Membrane Process

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This study aims to develop an easy and economic technique for dairy cleaning-in-place wastewater treatment to obtain high-quality treated wastewater effluent. A batch electrocoagulation (EC) study is conducted using iron and aluminum electrodes. The effects of various operational parameters are investigated on the treatment performance. Maximum soluble chemical oxygen demand (COD) removal efficiencies of 81% for an Fe–Fe electrode and 53% for an Al–Al electrode are obtained at an applied voltage of 10 V, an electrode spacing of 2 cm, pH of 7.2, for a reaction time of 60 min. Moreover, protein removal efficiencies of 72.6 and 79.4% are obtained for Fe–Fe and Al–Al electrodes, respectively. An immersed membrane is integrated into the EC reactor and the reactor is operated in continuous mode. COD removal efficiency is determined as 86.4 and 84.5% for Fe–Fe and Al–Al electrodes, respectively, in continuous mode. The protein removal efficiency is also calculated as 80.9 and 83.8% for Fe–Fe and Al–Al electrodes, respectively. The Fe–Fe electrode shows the maximum COD removal efficiency, whereas Al–Al provides the maximum protein removal efficiency. Additionally, field emission scanning electron microscopy (FE-SEM) and Fourier transform IR spectroscopy (FT-IR) are applied to describe fouling phenomena.

## 1. Introduction

The dairy industry uses too much water and also produces too much wastewater. This sector has a high organic load content which is basically composed of milk (raw material and dairy products). However, large amounts of effluent from dairy industry (generating from 0.2 to 10 L of effluent per liter of processed milk) contains high levels of chemical oxygen demand (COD), biochemical oxygen demand (BOD), oils and grease, nitrogen, and phosphorus.<sup>[1]</sup>

The dairy industry has a wide range of dairy products such as milk, cheese, butter, milk powder etc. After processing these products, too much wastewater is generated even though almost all the milk processing units use the automatic cleaning system – cleaning in place (CIP). Cleaning chemicals such as caustic and

acidic solutions are discharged as rinse water which pH value usually varies between 1 and 13.<sup>[1,2]</sup> CIP wastewater can also contain surfactants, base, acid, salts, sanitizers, and organic matters.<sup>[3–5]</sup> The high and varied pollution loads of this wastewater cause significant environmental, economic, and societal problems.<sup>[2,6]</sup>

Conducted studies indicated that a typical European dairy factory produced approximately 50 m<sup>3</sup> day<sup>-1</sup> wastewater with high concentration of organic matter and nutrients.<sup>[7,8]</sup> It is difficult to develop a single and an effective treatment method for dairy CIP wastewater due to a wide diversity of organic matters. Until now, anaerobic treatment,<sup>[9]</sup> chemical precipitation,<sup>[10]</sup> and membrane processes<sup>[11,12]</sup> were tested for the treatment of dairy CIP wastewater. However, intense research is ongoing to develop cheap and easy treatment methods.

Electrocoagulation (EC) process removes suspended or dissolved contaminants by dissolving either aluminum or iron ions from aluminum or iron electrodes applying electric current.<sup>[13,14]</sup> Easy operation, simple equipment, and little sludge formation are the advantages of the EC process.<sup>[15]</sup> EC is an intensively studied process and it has been successfully used to treat biodiesel wastewater,<sup>[16]</sup> urban wastewater,<sup>[17]</sup> chips manufacturing wastewater,<sup>[18]</sup> landfill leachate,<sup>[19]</sup> and dye stuff.<sup>[20,21]</sup> Moreover, it has also been used for the removal of boron from aqueous solution,<sup>[22]</sup> remediation of fluoride from pretreated photovoltaic wastewater,<sup>[23]</sup> elimination of the malathion pesticide from polluted solutions,<sup>[24]</sup> and simultaneous removal of hexavalent chromium and COD from industrial wastewater.<sup>[25]</sup>

Membrane processes for wastewater treatment to produce purified water have been applied in the food industry, especially the dairy industry. Membrane filtration of wastewater serves two purposes. First, membrane plays a major role for water reuse which decreases total water consumption. Second, it provides a requirement for control of the discharge of industrial wastewater. The purified water produced by membrane filtration can be reused in the dairy factory as heating or cooling water, boiler make-up water or cleaning purposes.<sup>[26]</sup> However, the proteinaceous materials in the dairy wastewater have severe fouling for the membrane materials.<sup>[27]</sup> Up to now, use of coagulant as a

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pretreatment for membrane application were used to control membrane fouling and flux decline.<sup>[10,28]</sup>

The objective of this study was to investigate EC process supported with immersed membrane for treatment of dairy industry wastewater containing moderate soluble organic carbon and protein. Batch and continuous EC processes for treating CIP wastewater were investigated and compared. The first purpose was to determine the effects of electrode material, applied voltage, pH, and electrode spacing to detect the optimum conditions on CIP wastewater treatment by the EC process. The second purpose was to integrate the membrane system to EC reactor which operated in optimum conditions to supply continuous treatment.

## 2. Experimental Section

### 2.1. Dairy CIP Wastewater Characterization

Wastewater samples from dairy CIP originate out of rinse and washing of tanks, bottles, cans, pipelines, and different equipment, and thus wastewater samples contain milk drippings and chemicals used for cleaning containers and equipment. CIP wastewater from dairy industry was kindly provided from a local factory located in Mersin, Turkey. Wastewater was stored in a closed plastic cans at +4 °C. The average pH values, COD concentrations, electrical conductivities, suspended solids, and total protein for the raw wastewater are shown in Table 1.

### 2.2. Electrochemical Reactor Setup

EC experiments were carried out in a cylindrical glass reactor (10.5 cm [radius] × 14.5 cm [height]). The iron and aluminum electrodes (one serving as a cathode and the other as anode) were used (6 cm width × 9 cm in height) and both electrodes had the same dimensions with 1 mm as thickness. For each electrode, the active surface area was 54 cm<sup>2</sup> and electrode distance was variable between 2 and 6 cm. Mixing in the reactor was achieved by a Teflon-covered stirring bar installed at the bottom of the reactor. The volume of the EC reactor was 1000 mL, and the net reaction volume was 800 mL. New electrodes were used for each experiment. The anode and cathode ends were linked to the positive and negative outlets of a DC power source (AATech ADC-3303D) with a maximum current rating of 30 A. However, several experiments were performed to obtain the best COD and total protein removal efficiency. The electrodes were prepared as follows in order to avoid any interference: (i) rinse with distilled

**Table 1.** Characteristics of the raw CIP wastewater from dairy industry.

Parameter	Value
pH	7.2 ± 0.1
Conductivity (μS/cm)	1060 ± 25
Suspended solid (mg/L)	415 ± 52
COD (mg/L)	800 ± 112
Total protein (mg/L)	80 ± 5

water (500 mL); (ii) clean in sodium hydroxide solution (500 mL, 0.5 wt.%); (iii) rinse with distilled water (500 mL); (iv) dry with absorptive paper. Selected initial pH value was adjusted in the reactor using HCl or NaOH (1 N) solutions.

### 2.3. Continuous Process Setup

A flat sheet MP005 polyethersulfone membrane (with nominal pore sizes of 0.05 μm) supplied by Microdyn-Nadir was used and the membranes were wetted in distilled water overnight. EC reactor was operated in the optimum conditions with an immersed membrane module to obtain continuous treatment. Process scheme is shown in Figure 1. The wastewater was continuously pumped with a peristaltic pump (Cole Parmer Masterflex) into the reactor at a constant rate (800 mL h<sup>-1</sup>). The flat sheet membrane modules, made of polypropylene, had a total area of 80 cm<sup>2</sup>. Short-term (180 min) filtration tests were conducted in constant pressure mode (-150 ± 5 mm Hg). Filtration tests were also conducted with raw wastewater. The permeate flow rate was measured every 5 min by an electronic balance and recorded. The permeate obtained from membrane was analyzed directly for COD and total protein removal. Excess sludge was withdrawn continuously out of the reactor.

### 2.4. Analytical Method

EC supported with immersed membrane process efficiency was measured by monitoring the COD and total protein values. COD was analyzed by closed reflux method with a thermo-reactor (Giant QBH2) using potassium dichromate as oxidant according to the standard methods.<sup>[29]</sup> The analysis for COD was performed in duplicates and average value was presented.

Determination of total protein concentration was measured according to Bradford method.<sup>[30]</sup> Protein concentration was determined using bovine serum albumin (BSA) standard curve (595 nm). The COD and total protein removal efficiencies R(%) were calculated as follows:

$$R(\%) = \frac{(L_o - L_t)}{L_o} \times 100 \quad (1)$$

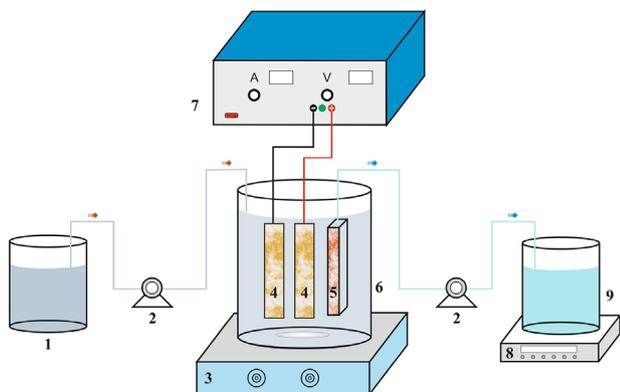
where  $L_o$  is the initial level and  $L_t$  is the level measured at time  $t$ .

Measurement of suspended solids was performed according to procedures described in APHA Standard Methods.<sup>[29]</sup> pH and conductivity values were monitored by a multi meter (WTW inoLab<sup>®</sup> pH 7110).

The permeation flux ( $J$ ) was measured by collecting the filtered water in the determined intervals and calculated using the following equation:

$$J = \frac{V}{A \times \Delta t} \quad (2)$$

where  $J$  is permeate flux (L m<sup>-2</sup> h<sup>-1</sup>),  $V$  the volume of permeate pure water (L),  $A$  the effective area of the membrane (m<sup>2</sup>), and  $\Delta t$  the permeation time (h).



**Figure 1.** Schematic diagram of the integrated treatment system. 1. Wastewater storage tank, 2. Peristaltic pump, 3. Magnetic stirrer, 4. Electrodes, 5. Membrane modules, 6. Electrocoagulation reactor, 7. DC power supply, 8. Balance, 9. Permeate storage tank.

The surface morphology of the clean and contaminated membranes was characterized by field emission scanning electron microscopy (FE-SEM), using a Zeiss/Supra 55 FE-SEM model operating at 10 kV. All samples were dried and coated under vacuum with a thin layer of platinum-palladium by a sputtering system. Fourier transform infrared (FTIR) (Perkin Elmer) spectroscopy was obtained to analyze the functional groups on contaminated membrane surfaces.

### 3. Results and Discussion

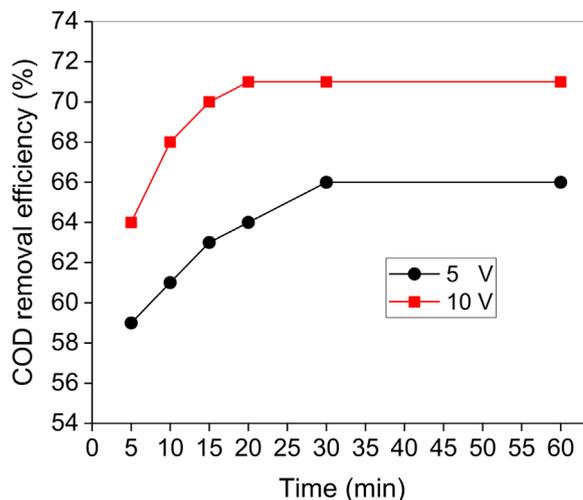
In this study, the effect of the basic operating parameters (type of electrode material, pH, applied voltage, electrode spacing, and reaction time) on the system performance was tested for treatment of the dairy industry CIP wastewater. The results are discussed below in the relevant sections.

#### 3.1. Batch Treatment Experiments by EC Process

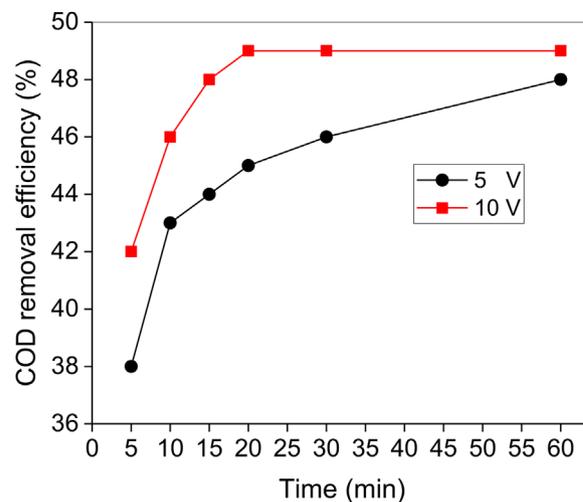
It was reported that electrode materials, applied voltage, pH, electrode spacing, and operating time have an important influence for the EC process.<sup>[31,32]</sup> In this study, applied voltage, pH, and electrode spacing were optimized. Treatment studies were conducted in 5, 10, 15, 20, 30, and 60 min for each experiment.

##### 3.1.1. The Effect of Applied Voltage on COD Removal Efficiency

Two different voltages (5 and 10 V) were applied at original pH (7.2) with 6 cm electrode spacing for 60 min to remove COD and proteins. According to **Figure 2**, increasing of applied voltage from 5 to 10 V, the removal efficiency of COD increased from 66 to 71% when Fe-Fe was used as electrode material (**Figure 2a**). Whereas, the increase in voltage did not affect significantly the COD removal efficiency when Al-Al was used as electrode material and COD removal efficiency was 48 and 49% for 5 and



(a)



(b)

**Figure 2.** Effect of applied voltage on COD removal efficiency for a) Fe-Fe electrode and b) Al-Al electrode (pH 7.2; electrode spacing, 6 cm)

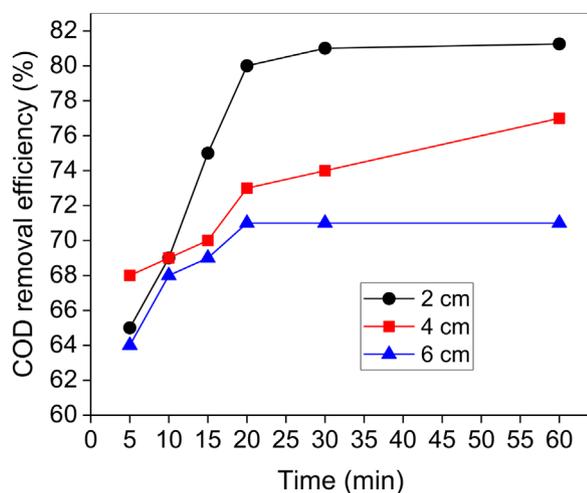
10 V, respectively (**Figure 2b**). The results showed that the Fe-Fe electrode provided better COD removal efficiency than the Al-Al electrode. The Fe-Fe electrode has relatively high solubility at acidic or neutral conditions and the dissolving rate of the electrode increased with increasing current density at this pH (7.2). Consequently, organic compound removal increased with increasing applied voltage due to releasing of more  $\text{Fe}^{3+}$  ions into wastewater at higher voltage and formation rate of iron hydroxide ( $\text{Fe}(\text{OH})_3$ ). The COD removal efficiency depended on the applied voltage. For this reason, the optimum applied voltage was chosen as 10 V for the following experiments.

Linares-Hernandez et al.<sup>[33]</sup> reported that high COD removal efficiency (99%) was obtained by two-step process EC with an iron electrode for industrial wastewater treatment. COD removal of palm oil mill wastewater by EC was studied by Sontaya et al.<sup>[34]</sup> Maximum COD removal (89.2%) was achieved at pH 4, current voltage of 12 V, and reaction time of 15 min. In another study,

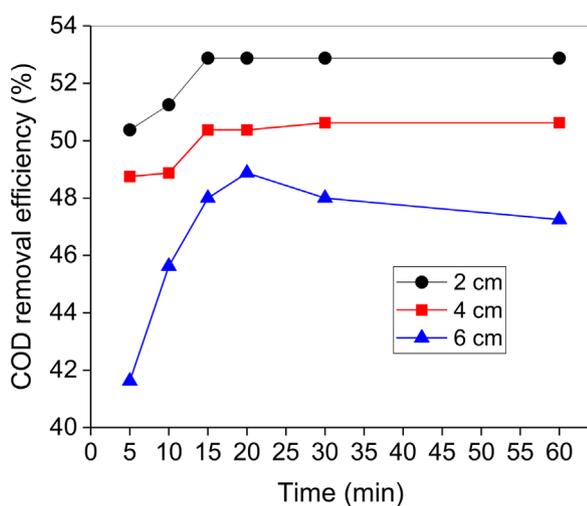
Jotin et al. showed that 74.1% COD removal efficiency was obtained at a voltage of 10 V using an aluminum electrode for the treatment of landfill leachate.<sup>[35]</sup> Chou et al.<sup>[36]</sup> investigated the relationship between COD removal efficiency and applied voltage during EC of real oxide-chemical mechanical polishing wastewater with an Fe/Al electrode pair. It was reported that COD removal efficiency reached 62.3, 85.3, and 91.5% after 20 min of electrolysis for 10, 20, and 30 V, respectively.

### 3.1.2. The Effect of Electrode Spacing on COD Removal Efficiency

The electrode spacing was optimized at 2–6 cm for further treatment. Three different spaces (2, 4, 6 cm) were applied for 60 min at 10 V and pH 7.2 (Figure 3). When the electrode spacing increased from 2 to 6 cm, the COD removal efficiency decreased



(a)



(b)

**Figure 3.** Effect of electrode spacing on COD removal efficiency for a) Fe-Fe electrode and b) Al-Al electrode (applied voltage, 10V; pH 7.2).

from 81 to 71% for the Fe-Fe electrode (Figure 3a) and from 53 to 47% for the Al-Al electrode (Fig. 3b). A decrease in the treatment efficiency was observed when the electrode spacing increased. It can be explained as molecule interactions with both oxidants and coagulants become weak when the electrode distance becomes >1 cm.<sup>[37]</sup> Similar results were also obtained by Naje et al.<sup>[37]</sup> during the treatment of textile wastewater using the EC process. The impact of electrode distance on pollutant removal was tested at 0.5, 1.0, and 1.5 cm. The COD removal efficiency was increased up to 90% at an electrode distance of 1 cm using titanium plates. Hussien et al.<sup>[38]</sup> found that a 3 cm electrode gap distance supplied effective treatment of sewage wastewater via the EC process using iron electrode to obtain high quality irrigation water or disinfected drinking water. In addition, Saleem et al.<sup>[39]</sup> reported that electrode spacing of 5 cm provided 77.2% COD removal efficiency within 30 min of EC treatment.

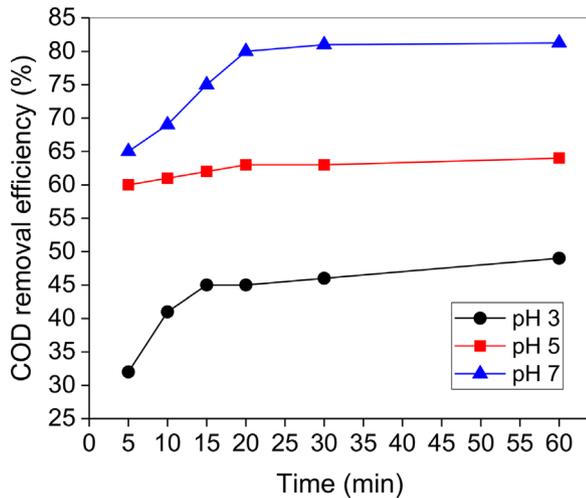
### 3.1.3. The Effect of pH on COD Removal Efficiency

The electrochemical reaction of Fe and Al electrode material for treatment process is well described in the literature.<sup>[14,40]</sup> When Fe is used as electrode material, iron ions are generated by electrochemical reactions and iron hydroxide,  $\text{Fe}(\text{OH})_{3(s)}$ , is produced depending on the pH.<sup>[41]</sup> However, aluminum ions form various monomeric species, and finally react to  $\text{Al}(\text{OH})_{3(s)}$  according to complex precipitation kinetics when an Al electrode is used.<sup>[42,43]</sup> Formed  $\text{Fe}(\text{OH})_{3(s)}$  and  $\text{Al}(\text{OH})_{3(s)}$  behave as “sweep flocs” and clutch soluble organic compounds.

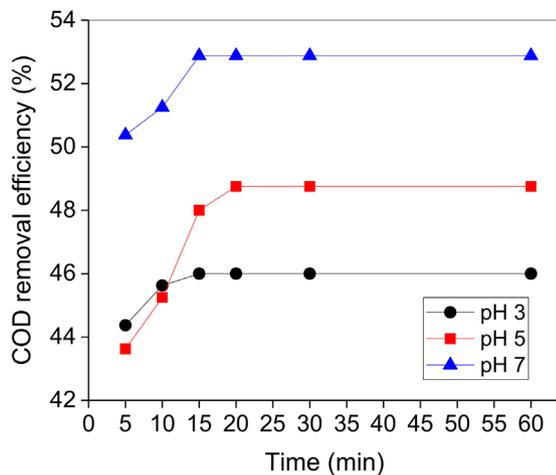
It is well known that the pH has a remarkable influence on the performance of the EC process. The effect of pH on the COD removal was examined at pH 3.0, 5.0, and 7.2 with iron and aluminum electrodes. Applied voltage of 10V and electrode spacing of 2 cm was kept constant in the experiments. It can be seen from Figure 4a and b that the COD removal efficiency of 81 and 53% was determined at original pH (7.2) for Fe-Fe and Al-Al electrodes, respectively. However, a lower COD removal efficiency was obtained at pH 3 as 32 and 49% for Fe-Fe and Al-Al electrodes, respectively. For pH < 6, reducing the protons to  $\text{H}_2$  and producing hydroxide ions in solution caused less COD removal efficiency.<sup>[44]</sup> Khandegar and Saroh reported that the maximum COD removal efficiency of 84.6 and 76.9% was determined at pH 7.2 for Al-Al and Fe-Fe electrodes, respectively.<sup>[45]</sup> Wang et al.<sup>[46]</sup> also investigated the efficiency of COD removal at different pH values, ranging from 2.5 to 9.5. The optimal pH of their study was found to be 5.1 and the COD removal efficiency was about 66% using Al-Al electrodes. Kobya et al.<sup>[18]</sup> and Linares-Hernandez et al.<sup>[47]</sup> reported that highest treatment efficiency was obtained with pH < 8 using an aluminum electrode.

### 3.1.4. Protein Removal Efficiency

Typical protein content in milk is 3.5% (w/w). Milk proteins exist in two distinct groups; caseins (80%) and whey proteins (20%). The whey proteins constitute about 18% of the protein content of milk. Casein, a protein found only in milk, contains all the essential amino acids. It accounts for 82% of the total proteins in milk and is used as a standard for evaluating proteins of other



(a)



(b)

**Figure 4.** Effect of pH on COD removal efficiency for a) Fe-Fe electrode and b) Al-Al electrode (applied voltage, 10 V; electrode spacing, 2 cm).

foods.<sup>[48]</sup> In this study, protein removal efficiency was measured from dairy industry CIP wastewater. The results are shown in Table 2. The Al-Al electrode was more effective than the Fe-Fe electrode for protein removal at all conditions. The maximum protein removal efficiency was obtained as 72.6% for the Fe-Fe electrode and as 79.4% for the Al-Al electrode at applied voltage of 10 V, electrode spacing of 2 cm, and pH 7.2.

### 3.2. Continuous Treatment Experiments by EC and Immersed Membrane Process

The continuous treatment studies were carried out after determining the optimum conditions in batch studies as applied voltage of 10 V, electrode spacing of 2 cm, pH of 7.2, and reaction time of 60 min. The average removal efficiency for COD and total protein at these experimental conditions in batch study were

**Table 2.** Protein removal efficiency for Fe-Fe and Al-Al electrodes (Operating time: 60 min).

Condition	Protein removal efficiency (%)	
	Fe-Fe electrode	Al-Al electrode
Applied voltage (V)		
5	59.9	71.1
10	69.2	73.6
Electrode spacing (cm)		
2	72.6	79.4
4	71.3	78.2
6	69.2	73.6
pH		
3.0	61.9	76.6
5.0	63.3	77.4
7.2	72.6	79.4

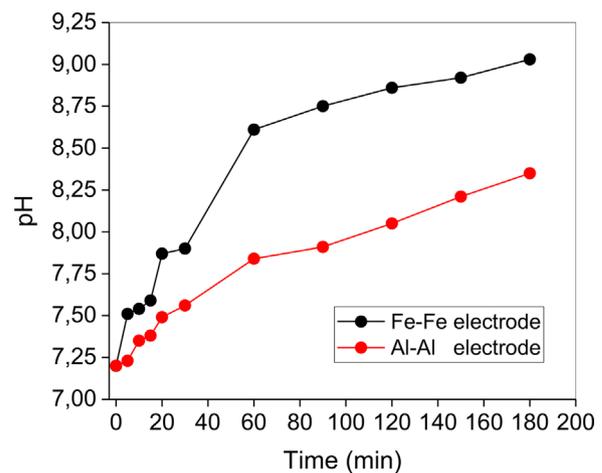
determined as 81.0 and 72.6% for the Fe-Fe electrode, and 53.0 and 79.4% for the Al-Al electrode, respectively. During the continuous experiments, wastewater was pumped to the EC reactor with a flow rate of 800 mL h<sup>-1</sup>. pH values, COD, and total protein values were measured every 30 min for a period of 3 h.

#### 3.2.1. Change of pH

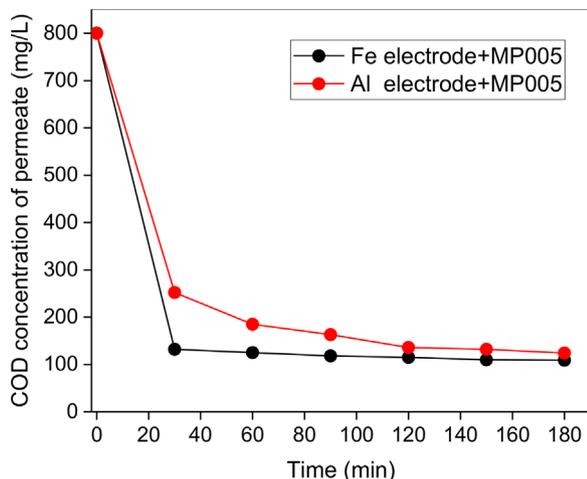
The change of pH over time is shown in Figure 5. When Fe-Fe and Al-Al electrodes were used, pH value increased to 9.0 and 8.3, respectively, after 180 min reaction time (initial pH 7.2).

#### 3.2.2. Change of COD and Protein Concentration

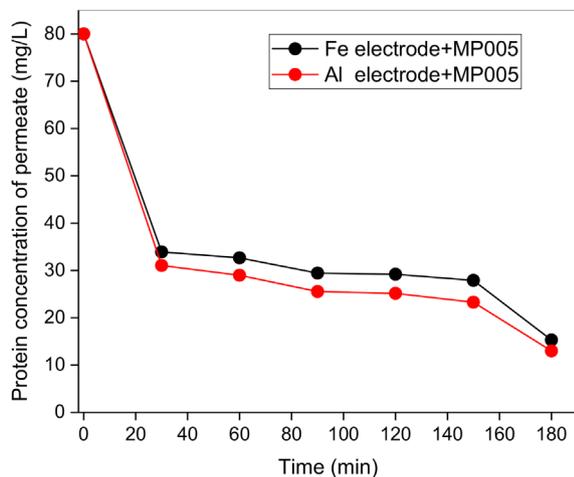
The change of COD and protein concentration is shown in Figure 6. The inlet COD concentration was 800 mg L<sup>-1</sup>. Fe-Fe and Al-Al electrodes were used in the EC reactor and immersed



**Figure 5.** pH changes during continuous treatment (applied voltage, 10 V; electrode spacing, 2 cm; pH 7.2; time, 180 min).



(a)



(b)

**Figure 6.** a) COD and b) protein changes during continuous treatment (applied voltage, 10V; electrode spacing, 2 cm; pH 7.2; time, 180 min).

membrane was integrated in the reactor. Membrane filtration was operated for 180 min and COD was measured both in the EC reactor and membrane permeate. The COD concentration inside the reactor was measured as 152 mg L<sup>-1</sup> for the Fe-Fe electrode and 376 mg L<sup>-1</sup> for the Al-Al electrode. However, the COD concentration of the membrane permeate was measured as 109 mg L<sup>-1</sup> for the Fe-Fe electrode and 124 mg L<sup>-1</sup> for the Al-Al electrode at the end of 180 min operating time. Consequently, total COD removal efficiency was 86.4% for the Fe-Fe electrode and 84.5% for the Al-Al electrode in continuous operation (Figure 6a).

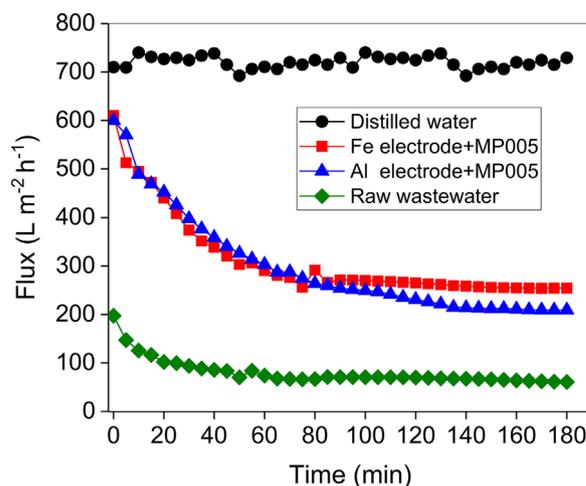
The change of the protein concentration is shown in Figure 6b. The total protein concentration of the wastewater was 80 mg L<sup>-1</sup>. The protein concentration inside the reactor was measured as 21.9 mg L<sup>-1</sup> for the Fe-Fe electrode and 16.5 mg L<sup>-1</sup> for the Al-Al electrode. However, the protein concentration of the membrane permeate was measured as 15.3 mg L<sup>-1</sup> for the Fe-Fe electrode and 12.9 mg L<sup>-1</sup> for the Al-Al electrode at the end of

180 min operating time. As a result, the total protein removal efficiency was 80.9% for the Fe-Fe electrode and 83.8% for the Al-Al electrode in continuous operation modus (Figure 6b).

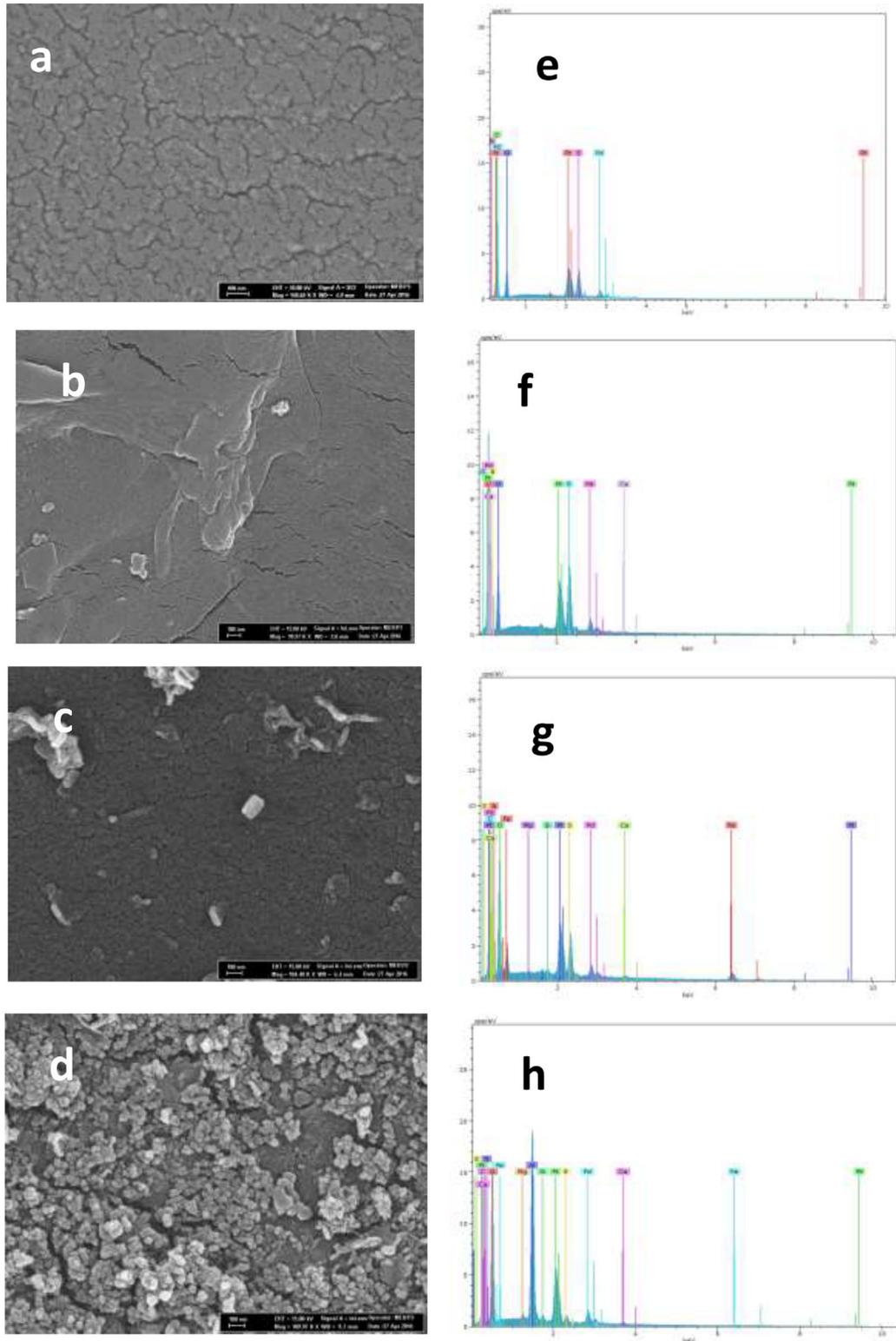
### 3.2.3. Change of Flux

The wastewater containing soluble organic carbon and proteins was filtered with a MP005 membrane and the permeate flux was calculated. The membrane experiments were carried out before and after EC treatment. As shown in Figure 7, the lowest permeate flux was obtained for raw wastewater filtration. The initial flux was 197 L m<sup>-2</sup> h<sup>-1</sup> and the flux reached a pseudo steady-state condition within 30 min. The steady-state permeate flux value was 61 L m<sup>-2</sup> h<sup>-1</sup>. However, the initial flux was 610 L m<sup>-2</sup> h<sup>-1</sup> using the Fe-Fe electrode and immersed membrane simultaneously. The flux reached a pseudo steady-state condition within 90 min and the steady-state permeate flux value was 254 L m<sup>-2</sup> h<sup>-1</sup>. On the other hand, the initial flux was 600 L m<sup>-2</sup> h<sup>-1</sup> using the Al-Al electrode. The flux reached a pseudo steady-state condition within 90 min and the steady-state permeate flux value was 209 L m<sup>-2</sup> h<sup>-1</sup>. It can be clearly seen from the results that the Fe-Fe electrode and immersed membrane resulted in a cleaner permeate and higher flux than the Al-Al electrode.

The surface morphologies of clean and contaminated membranes were investigated by FE-SEM to observe changes on the membrane morphology. Figure 8a–d shows the SEM images of the top layer surfaces of clean and contaminated membranes. Figure 8a show a clean membrane structure and it can be seen that the membrane surface shows a fractured surface. However, membrane pores were covered by fouling matter which could be organic and inorganic when raw wastewater was filtered through the membrane without being subjected to any treatment process (Figure 8b). It can be seen from Figure 8c and d that the membrane had a cleaner surface when the immersed membrane was integrated within the EC process using the Fe-Fe electrode compared to the Al-Al electrode. Figure 8e–h shows the FE-SEM images of the top



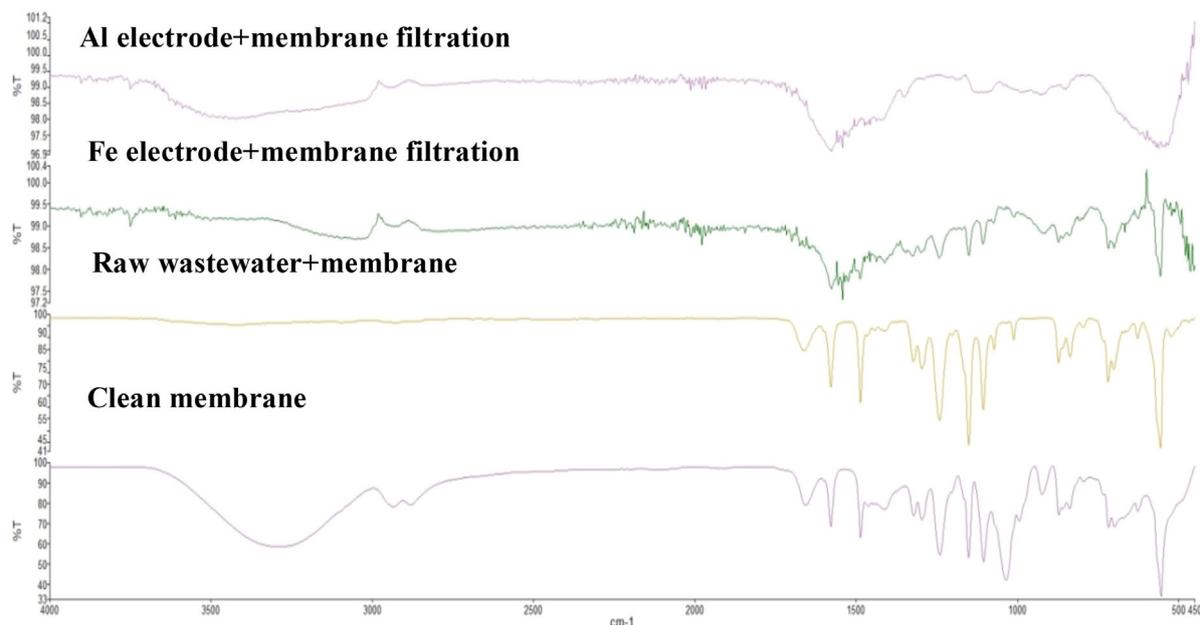
**Figure 7.** Flux changes during continuous treatment (10V, 2 cm, pH 7.2, and 180 min).



**Figure 8.** a–d) SEM and e–h) FE-SEM images of clean and contaminated membranes.

layer surfaces of clean and contaminated membranes. As expected, C, O, H, S were observed for the clean MP005 membrane (Figure 8e). However, Ca and N were observed on the membrane surface when raw wastewater filtration was used

(Figure 8f). The membrane surface shown in Figure 8g and h indicated that Fe and Al metals, released from the electrode surface, deposited on the membrane surface and some metals formed aggregates (metal hydroxide). The



**Figure 9.** FTIR spectra of clean membrane, raw wastewater filtrated membrane, and EC integrated membranes

membrane surface was cleaner when the Fe-Fe electrode was used in the EC process. Moreover, some inorganic contaminants such as Ca, Mg, Al, Si, N contained in wastewater were determined on the membrane surface. Consequently, the immersed membrane supported with EC process supplied an efficient result to control fouling of the membrane surface.

FT-IR images of the clean and contaminated MP005 membranes are shown in **Figure 9**. In the FT-IR spectrum of the clean membrane, the bands at 3293 and 1655  $\text{cm}^{-1}$  were due to the OH group and C=O stretching, respectively.<sup>[49]</sup> The C=O band decreased after EC treatment using Fe-Fe and Al-Al electrodes. Aromatic bands at 1578 and 1486  $\text{cm}^{-1}$  decreased after fouling with metal hydroxides. The band at 1542  $\text{cm}^{-1}$  is characteristic for N-H and was visible after EC treatment. This peak was not observed during the filtration of the raw wastewater. However, protein-metal complexes can be formed during treatment by the EC process and the complexes can be deposited on the membrane surface. The clean membrane exhibited bands at 1241 and 1151  $\text{cm}^{-1}$  that were repressed by the foulants. However, the band at 1105  $\text{cm}^{-1}$  disappeared after the EC treatment. A band at 1072 and 1035  $\text{cm}^{-1}$  indicated the existence of organic matter and carbohydrates (lactose), disappeared during the EC treatment. These organic substances were degraded by EC treatment and prevented accumulation of foulants on the membrane surface. The peaks at 1035  $\text{cm}^{-1}$  represent the sulfone group (S=O) stretching for the clean membrane. This band disappeared after membrane fouling (Figure 9).

#### 4. Conclusion

EC of dairy industry CIP wastewater was carried out using Fe-Fe and Al-Al electrodes in batch mode of operation. Moreover, the EC process was operated in continuous mode supported with

immersed membrane in the reactor. The optimum value for batch mode of operation was found to be applied voltage of 10 V, electrode spacing of 2 cm, and pH of 7.2. In batch mode, the average removal efficiency for COD was obtained as 81% for the Fe-Fe electrode and 53% for the Al-Al electrode, respectively. The average removal efficiency for proteins was obtained as 72.6% for the Fe-Fe electrode and 79.4% for the Al-Al electrode, respectively. In continuous mode, 86.4 and 84.5% COD removal efficiency was obtained for Fe-Fe and for Al-Al electrodes, respectively. However, 80.9 and 83.8% protein removal efficiency was calculated for Fe-Fe and Al-Al electrodes, respectively. Membrane study results also showed that the Fe-Fe electrode resulted in a higher flux and cleaner permeate than the Al-Al electrode. The proposed process using EC supported with immersed membrane can be performed for the treatment of different kinds of CIP wastewater.

#### Abbreviations

BOD, biochemical oxygen demand; BSA, bovine serum albumin; CIP, cleaning in place; COD, chemical oxygen demand; EC, electrocoagulation; FE-SEM, field emission scanning electron microscopy; FT-IR, Fourier transform IR spectroscopy.

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#### Conflict of Interest

The authors have declared no conflict of interest.

## Keywords

Al–Al electrode, dairy industry wastewater, Fe–Fe electrode, membrane fouling, wastewater treatment

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