



Can electro-cleaning solve the problem of membrane biofouling for membrane bioreactor?

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ABSTRACT

Membrane bioreactor (MBR) is a trustworthy and encouraging technology for wastewater treatment and reuse. However, membrane fouling limits the sustainability of this technology. Electro-membrane bioreactor (E-MBR) is a promising technology for the control of membrane fouling which prevents the adhesion of the charged pollutants to the membrane surface by several mechanisms. In this study, an E-MBR system with a platinum-plated titanium anode and an activated carbon cloth membrane cathode was used for the control of membrane fouling by electro-cleaning. The effects of applied voltage (5–50 V) and direct current exposure time (1–5 min) on permeate flux and, extracellular polymeric substances, soluble microbial products (SMPs) and chemical oxygen demand (COD) removal efficiencies were examined. Also, the impact of electro-cleaning on reversible and irreversible membrane fouling was investigated. The maximum removal efficiencies of SMP carbohydrate, SMP protein and COD were determined as 88.1%, 86.5%, and 83.8% for 5 V and 3 min electro-cleaning time while it was about 18.8%, 80.1%, and 30.3% for without electro-cleaning, respectively. Also, the reversible membrane fouling was enhanced from 52.5% to 98.5% and the irreversible membrane fouling was decreased from 47.4% to 1.5% when the electro-cleaning was applied for 5 V and 3 min. This study showed that E-MBR provides promising results for the control of membrane fouling in MBR.

Keywords: Electro-membrane cleaning; Membrane biofouling; EPS; Domestic wastewater

1. Introduction

Membrane bioreactor (MBR), which is the combination of a biological system with a membrane filtration technology, has gained many advantages compared to conventional activated sludge process due to the better effluent quality, higher disinfection efficiency, less footprint and sludge formation and flexible operating possibility against variable influent characterization [1,2]. However, besides all these advantages, membrane biofouling is the main issue

that limiting the sustainability of this technology which caused a decrease in permeate flux, an increment in membrane pressure, and a higher operational cost. In general, membrane fouling can occur as reversibly which can be removed by physical cleaning methods and irreversibly that can be removed by only chemical cleaning and maintenance cleaning or it cannot be recovered [5]. The main contributors that enhance the membrane fouling are the membrane properties, characterization of the wastewater (size of the flocs, presence of the colloids), biofoulants such as

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extracellular polymeric substances (EPSs), soluble microbial products (SMPs or soluble EPS), operating parameters like mixed liquor suspended solids (MLSS), sludge retention time (SRT), hydraulic retention time (HRT) [6,7]. Among these, EPS and SMP are the major components of biofouling which are the metabolism products of the microorganisms and they have mainly negative charges [8]. EPSs are mostly comprised of polysaccharides and proteins which are the major construction components for flocs, sludge, and biofilms. SMPs, which are similarly composed of proteins and carbohydrates, are formed during microbial activities like biomass growth and decay [9].

There are many methods to reduce and control membrane fouling includes appropriate pre-treatment technologies, backwashing of permeate water or filtration relaxation, chemical cleaning of the membranes or chemical added backwashing, optimization of the operating conditions such as SRT or backwashing frequency, improvement of the reactor shape, aeration intensity and membrane module configuration, use of adsorbents and coagulants, fabrication and modification of fouling resisted membranes by the addition of nanoparticles [10–13]. Among these, electro-membrane bioreactor (E-MBR) is a promising technology for the control of membrane fouling in which a direct electrical field is applied into MBR during membrane filtration to avoid the adhesion of the charged pollutants (SMPs, EPSs, flocs, etc.) to the membrane surface [14,15]. In that mechanism, also called electrophoresis, the repulsion force was independent of the anode material while in an electro-coagulation mechanism, the hydrated oxide metal ions were released from the iron or aluminum anodes and acted as coagulants [16]. The efficiency of the E-MBR depends on the material and design of the electrodes, and the applied direct current field [17].

In recent years, many researchers have studied E-MBR to reduce the membrane fouling. For instance, Bani-Melhem and Elektorowicz [17] investigated the treatment efficiency of the submerged E-MBR for the treatment of synthetic wastewater and they obtained 96% chemical oxygen demand (COD) and 98% $\text{PO}_4\text{-P}$ removal efficiencies when the iron electrode was used. The operational mode of direct current (DC) was adjusted as 15 min on–45 min off. Ibeid et al. [10] studied the E-MBR to observe the effect of electro-conditioning on the membrane fouling dewatering efficiency of the activated sludge. Several current densities between 5 and 35 A/m^2 and five electrical exposure modes (time on/time off) were tested. It was obtained that the filterability of the sludge was improved for the DC of 15 to 35 A/m^2 and the membrane fouling rate was reduced by 6 times. The effect of the short time electrical field usage on the membrane fouling and filtration performance was investigated by Liu et al. [18] and it was observed that the applied DC was importantly improved the permeate flux of the membrane, reduced the irreversible membrane fouling as well as sludge EPS formation.

The specific aim of this study was to investigate the effect of electro-cleaning on membrane filtration performance and to decrease membrane fouling for the treatment of municipal wastewater by E-MBR. Therefore, the effect of DC exposure times (1–3–5 min) and applied voltage (5–50 V) on flux performance, EPS, SMP fractions and COD removal

efficiencies, reversible and irreversible membrane fouling were investigated.

2. Materials and methods

2.1. Experimental setup

The continuously operated E-MBR consisted of an aerobic biological reactor with a volume of 25 L and a submerged membrane module. The membrane module comprised platinum-plated titanium as anode and an activated carbon cloth membrane (ACCM) as a cathode. The surface area of the membrane module and electrodes were 25 and 17.6 cm^2 , respectively. The distance of the anode was adjusted as 1 cm away from the membrane surface. All the experiments were carried out by different membrane sheets. The ACCM was used as a dynamic membrane during the experiments due to its high electrical conductivity. The constant vacuum pressure was adjusted as 200 mbar.

The wastewater was taken from an aeration basin of a real municipal wastewater treatment plant and fed to the laboratory scale E-MBR system. Then, synthetic wastewater was added continuously to the system according to the level sensors and the synthetic wastewater was prepared as in the study by [19]. The HRT (θ_h) and sludge age were adjusted as 12 h and 15 d, respectively. The reactor mixing was provided with the diffusers placed at the bottom of the reactor. A schematic diagram of the E-MBR is given in Fig. 1. The details of the module assembly were given elsewhere [19].

Three different electro-cleaning times (1, 3, and 5 min) and four different voltages (5, 10, 30, and 50 V) were tested to the E-MBR and the operation parameters were optimized. The SMP and COD concentrations of the permeate and the SMP and EPS concentrations of the sludge obtained from the membrane surface were analyzed for all experiments. The COD and MLSS concentrations of the activated sludge were measured daily.

2.2. Analytical methods

In the EPS and SMP analyzes, the sample was prepared by a physical–chemical extraction method as stated by Li et al. [20]. The polysaccharides and protein contents in SMP and EPS were analyzed as DuBois [21] and Lowry [22] methods, respectively. COD and MLSS concentrations are determined according to Standard Methods [23]. The COD analysis of the activated sludge was performed in the filtrate through a 0.45 μm membrane. Each analysis was repeated three times.

2.3. Characterization methods

The surface morphologies and the chemical compositions of the pristine and fouled membranes were analyzed with scanning electron microscopy (SEM, Zeiss Supra 55, Germany), and energy-dispersive X-ray spectroscopy (EDX attached to the SEM) (Quorum, Q150R ES, UK), respectively. Fourier-transform infrared spectroscopy (FTIR) spectrums of the pristine and fouled membranes were analyzed with the Perkin Elmer FTIR spectrometer (USA) using a spectral range of 4,000–400 cm^{-1} .

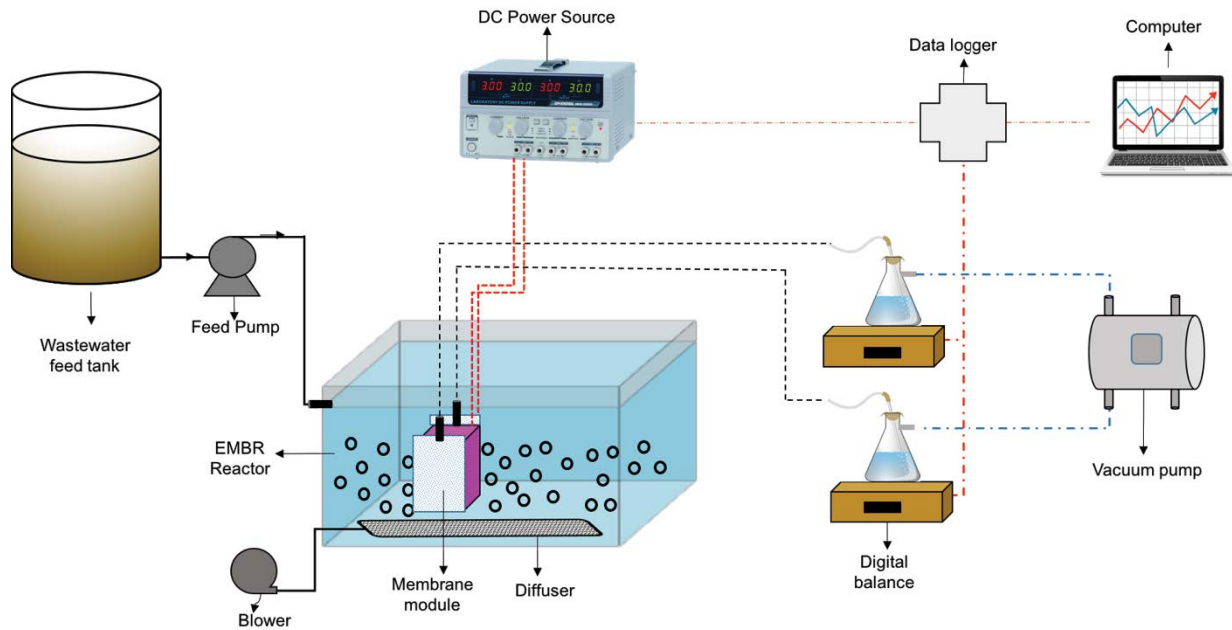


Fig. 1. Schematic diagram of the E-MBR.

3. Results and discussion

3.1. Effect of time and electric current on membrane cleaning

The effect of cleaning time was investigated on membrane cleaning. The MBR system was operated 30 min vacuum on mode. After then, the permeate valve was switched off and electro-cleaning was started during 1, 3, 5 min at 30 V. The flux data vs. time for electro cleaning time and the applied voltage is shown in Fig. 2. A rapid flux decline from 316 to 8 L/m²/h was observed in the absence of electro cleaning for 450 min filtration (Fig. 2a). However, steady-state fluxes were 17.2, 18.6, 13.6 L/m²/h for 1, 3, 5 min at 30 V, respectively. The results showed that 3 min electro-cleaning enhanced better results than 1 and 5 min operation. The flux decreased gradually when the applied voltage increased from 5 to 50 V (Fig. 2b). The steady-state fluxes were 62.7, 38.6, 18.6, 13.7 L/m²/h at 5, 10, 30, 50 V for 3 min electro cleaning and 450 min filtration, respectively. The results showed that low electric current (5 V) supplied the best flux compared to high electric current.

3.2. Comparison of permeate water quality

The effect of electro cleaning time and applied voltage on SMP fractions and COD concentrations are presented in Fig. 3. SMP carbohydrate (SMP_c) concentration was higher than SMP protein (SMP_p) concentration for all working conditions. SMP_c and SMP_p concentrations were the lowest for 3 min electro cleaning time (10.7 and 4.8 mg/L, respectively) (Fig. 3a). SMP_c and SMP_p removal efficiencies without electro cleaning were 18.7% and 80.1%, respectively. When electro cleaning was applied throughout the dynamic membrane, SMP_c removal efficiencies increased and were measured as 36.1%, 88.1%, and 23.8% for 1, 3, and 5 min, respectively. SMP_p removal efficiencies decreased and were

measured as 69.5%, 86.5%, and 79.8% for 1, 3, and 5 min electro cleaning, respectively. COD removal efficiencies improved when electro cleaning was applied. The highest COD removal efficiency was obtained as 83.8% when 3 min electro cleaning was applied at 5 V (Fig. 3b). It could be concluded that electro cleaning time was a very important parameter for SMP fractions in the permeate and cleaning under the electric field for 5 V and 3 min could enhance better soluble biological molecules oxidation without cell disruption.

The microorganism will deposit on the dynamic membrane depend on filtration time and biofilm formation will occur with time. In our study, we also measured EPS fractions by scraping the biofilm layer on the dynamic membrane at the end of the filtration. The effect of electro cleaning time and applied voltage on EPS concentrations are presented in Fig. 4. In the bioreactor, SMP_c and EPS_c concentrations were higher than SMP_p and EPS_p concentrations. However, SMP_p and EPS_p concentrations were higher than SMP_c and EPS_c concentrations when the biofilm layer deposited on the dynamic membrane was analyzed. The concentration of EPS and SMP fractions were the lowest for 3 min electro cleaning time and 5 V applied voltage. SMP_c and SMP_p concentrations increased from 70.3 and 203.4 mg/L to 79.0 to 138.5 mg/L, respectively, when electro cleaning time increased from 1 to 5 min (Fig. 4a). Moreover, SMP_c and SMP_p concentrations increased from 6.2 and 5.9 mg/L to 65.9 to 125.9 mg/L, respectively, when applied voltage increased from 5 to 50 V (Fig. 4b).

3.3. Comparison of physical and electro cleaning on fouling resistance

The fouling resistance was also determined for activated sludge filtration depends on electro-cleaning time

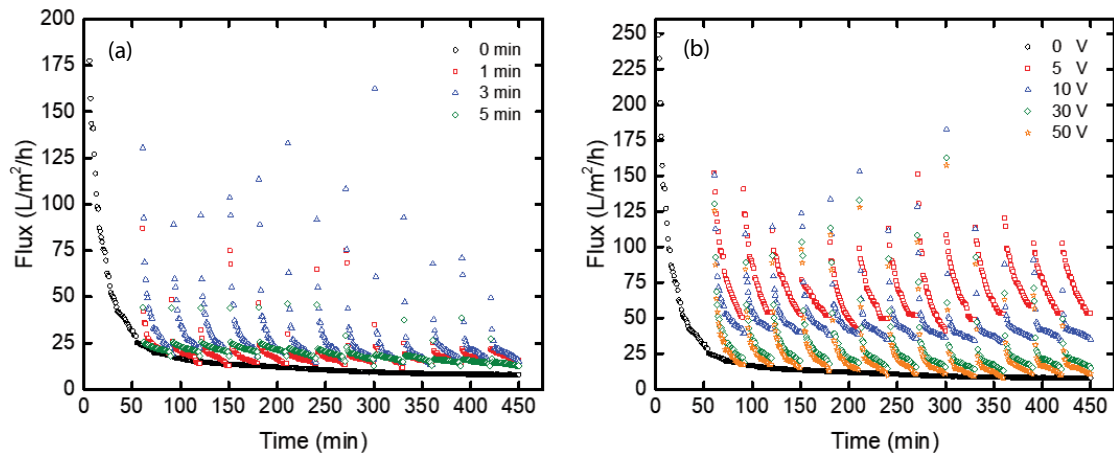


Fig. 2. Effect of (a) electro-cleaning time (experimental conditions: ΔP : 200 mbar; voltage: 30 V) and (b) applied voltage on permeate flux (experimental conditions: ΔP : 200 mbar; electro-cleaning time: 3 min).

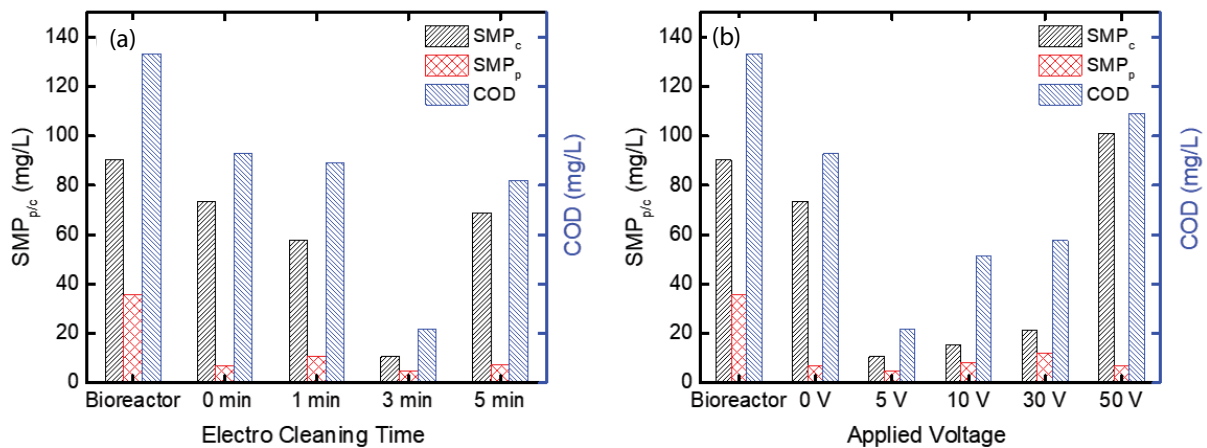


Fig. 3. Effect of (a) electro cleaning time (experimental conditions: ΔP : 200 mbar; 30 min vacuum on/1, 3, 5 min vacuum off; voltage: 5 V; filtration time: 450 min) and (b) applied voltage on SMP fractions (SMP_c and SMP_p) and COD removal efficiencies using the dynamic membrane (experimental conditions: ΔP : 200 mbar; 30 min vacuum on/3 min vacuum off; filtration time: 450 min).

and applied electric current. Moreover, physical and electro-cleaning were also compared and the values are shown in Fig. 5. First, electro-cleaning time on fouling resistance was investigated. The results showed that reversible (R_r) and irreversible (R_{ir}) resistance of the membrane were 52.5% and 47.4%, respectively, when electro-cleaning did not apply throughout the membrane (0 min) (Fig. 5a). R_{ir} decreased from 31.5% to 1.5% when electro-cleaning time increased from 1 to 3 min. However, R_{ir} increased dramatically from 1.5% to 48.8% when cleaning time increased from 3 to 5 min. Thus, 3 min electro-cleaning was chosen for further study. Second, the applied voltage on fouling resistance was optimized. The results showed that R_{ir} increased from 1.5% to 41.7% and R_r decreased from 98.3% to 57.8% when applied voltage increased from 5 to 50 V (Fig. 5b). Thus, 5 V was chosen as the optimum voltage for electro-cleaning. Third, the membrane was backwashed with permeate water for 3 min in every 30 min and the fouling resistance values were compared to electro cleaning values. The results clearly

showed that electro-cleaning decreased R_{ir} of the dynamic membrane (Fig. 5c). Physical cleaning was not as good as electro-cleaning.

3.4. Characterization of the dynamic membrane

The SEM images and SEM-EDX analyses of the pristine and fouled dynamic membranes operated with Ti/ACCM anode/cathode electron pairs were investigated at different electro cleaning time and voltage. First, the effect of electro cleaning time from 1 to 5 min was investigated on dynamic membrane fouling at 30 V. The surface morphology of the membranes was affected under the electrical field (Fig. 6). The clean membrane consists of large fibers and it contains mostly C and O due to its carbon structure (Fig. 6a). The dynamic membrane suffered from foulants without an electric field (Fig. 6b). However, some foulants were deposited when electro cleaning was applied between 1–5 min (Figs. 6c–e). Long-term electro cleaning (5 min) caused

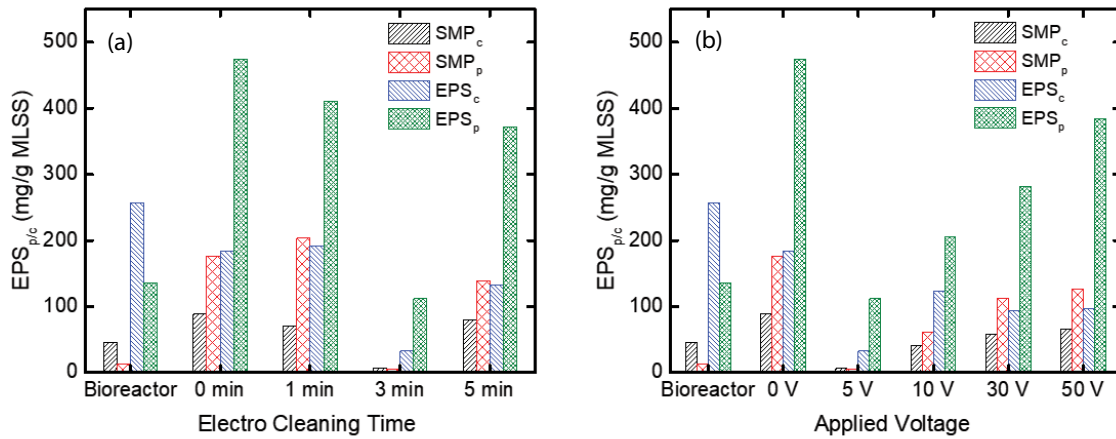


Fig. 4. Effect of (a) electro cleaning time (experimental conditions: ΔP : 200 mbar; 30 min vacuum on/1, 3, 5 min vacuum off; voltage: 5 V; filtration time: 450 min) and (b) applied voltage on SMP (SMP_c and SMP_p) and EPS (EPS_c and EPS_p) fractions deposited on the dynamic membrane (experimental conditions: ΔP : 200 mbar; 30 min vacuum on/3 min vacuum off; filtration time: 450 min).

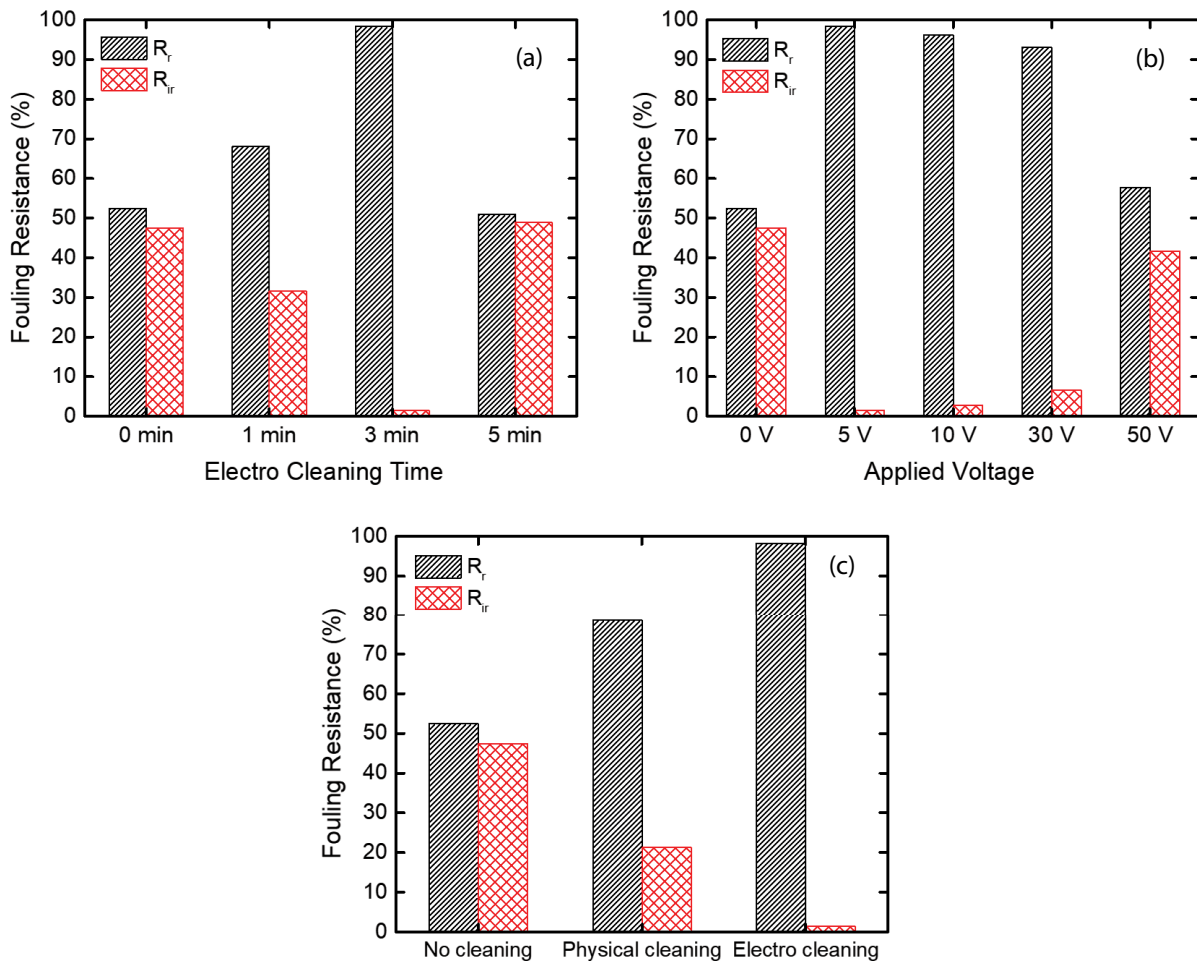


Fig. 5. Change of fouling resistance with (a) electro-cleaning time (experimental conditions: ΔP : 200 mbar; 30 min vacuum on/1, 3, 5 min vacuum off; voltage: 30 V; filtration time: 450 min), (b) applied voltage (experimental conditions: ΔP : 200 mbar; 30 min vacuum on/3 min vacuum off; filtration time: 450 min) and (c) comparing of physical and electro-cleaning (experimental conditions: ΔP : 200 mbar; 30 min vacuum on/3 min vacuum off; voltage: 5 V; filtration time: 450 min).

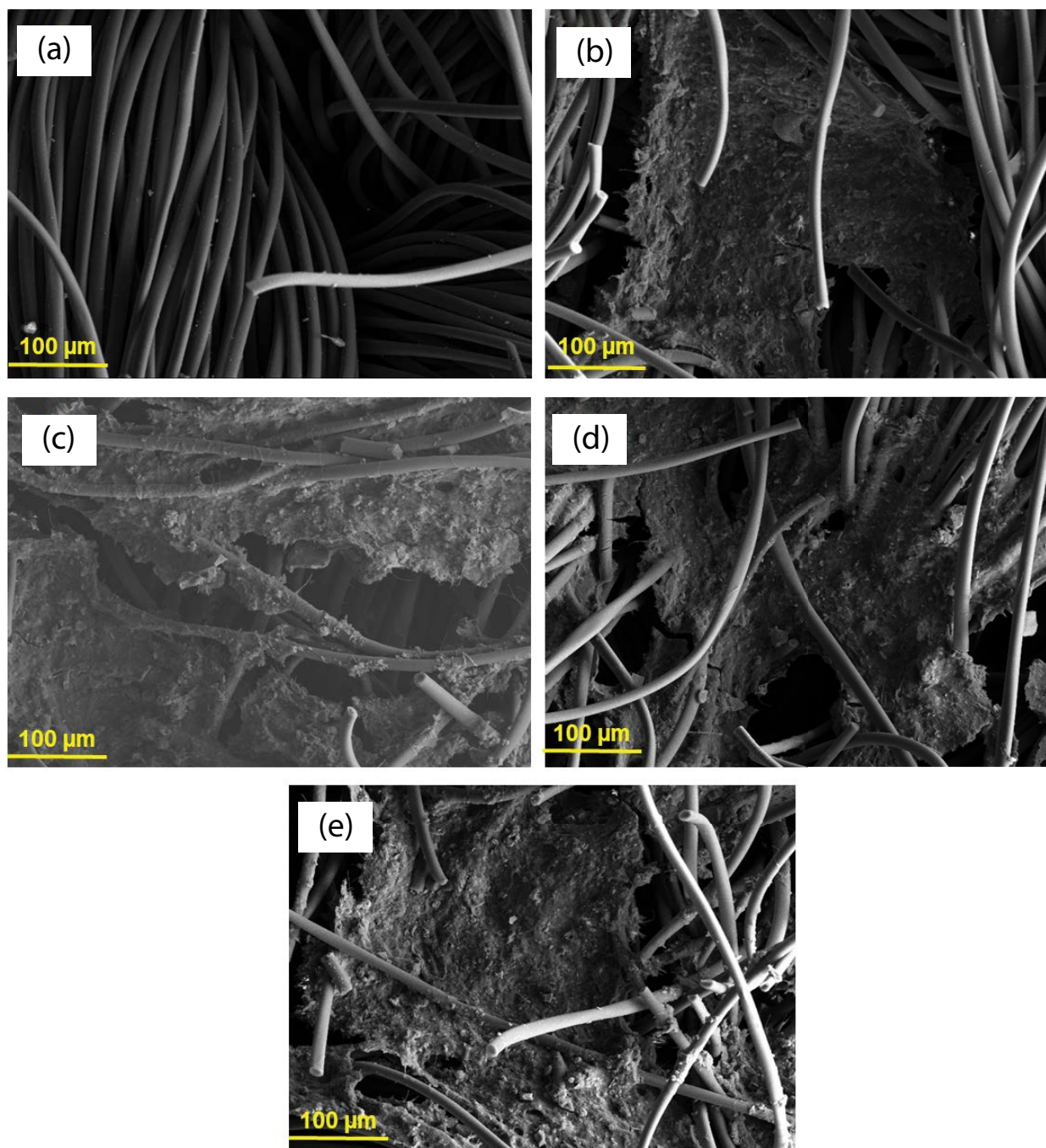


Fig. 6. SEM images of the membranes cleaned with and without electric field (a) clean pristine membrane, (b) without electro-cleaning, (c) 1 min electro-cleaning, (d) 3 min electro-cleaning and (e) 5 min electro-cleaning (SEM images were taken the experimental conditions of ΔP : 200 mbar; 30 min vacuum on; ΔE : 30 V during vacuum off mode for 1, 3, 5 min; filtration time: 450 min).

more membrane fouling due to the disruption of the cells (Fig. 6e).

Second, the effect of applied voltage from 5 to 50 V was investigated on dynamic membrane fouling for 3 min electro-cleaning time. The SEM images clearly showed that low voltage protected the membrane surface against fouling (Fig. 7a). When applied voltage increased from 10 to 50 V, the deposition of foulants on the membrane surface also increased (Figs. 7b–d).

Third, physical cleaning with permeate and electro-cleaning performance was also compared for applying 3 min backwash and electric current at 5 V. The SEM images clearly showed that physical cleaning was not enough to remove foulants from the membrane surface (Fig. 8a); however, the cake formation on the membrane surface removed more efficiently compared to physical cleaning (Fig. 8b).

FTIR spectroscopic study of the pristine and fouled dynamic ACCM was conducted to ascertain the active

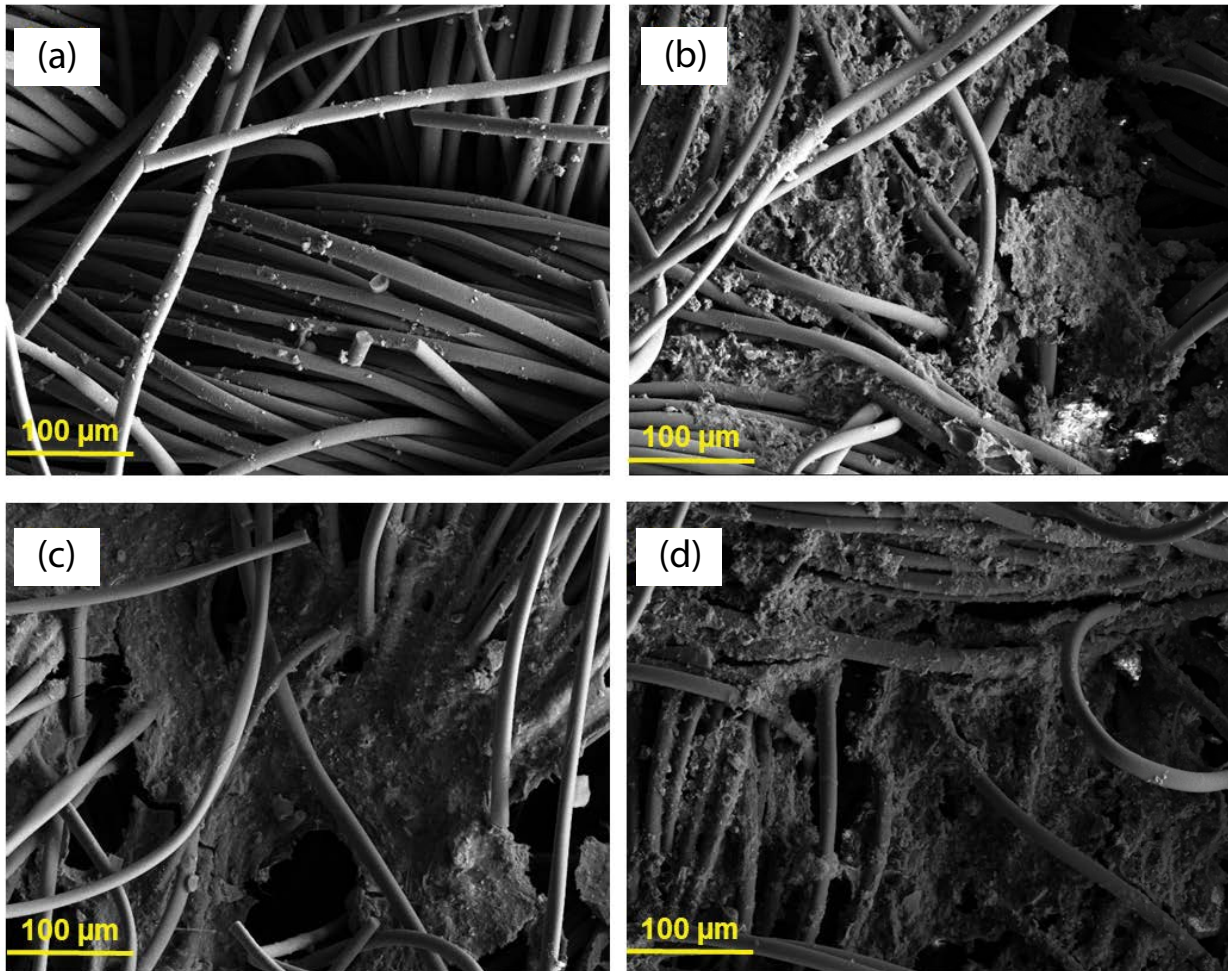


Fig. 7. SEM images of the membranes cleaned with an electric field for 3 min (a) ΔE : 5 V, (b) ΔE : 10 V, (c) ΔE : 30 V and (d) ΔE : 50 V (SEM images were taken the experimental conditions of ΔP : 200 mbar; 30 min vacuum on/3 min vacuum off; filtration time: 450 min).

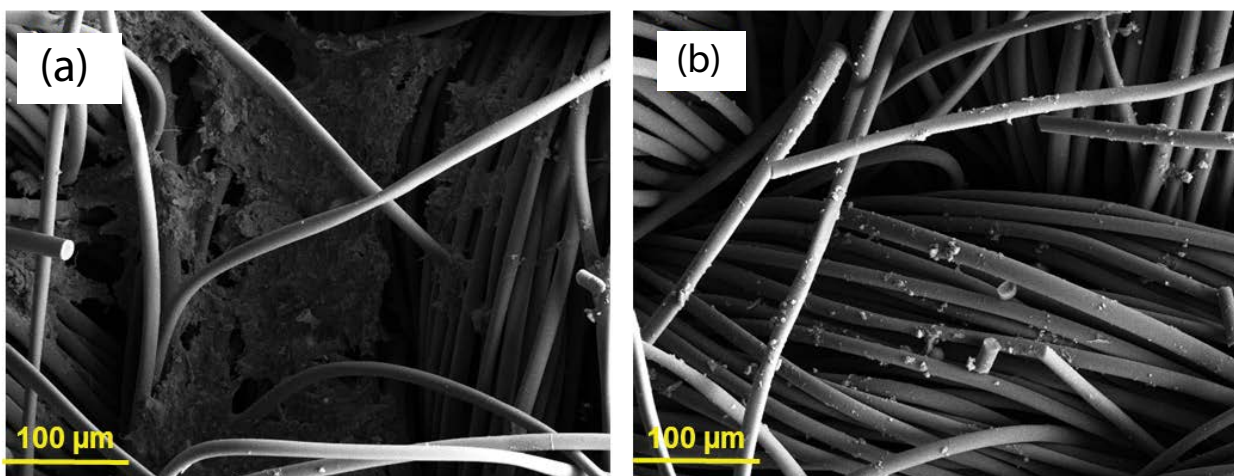


Fig. 8. SEM images of the membranes applied (a) physical and (b) electro cleaning for 3 min (SEM images were taken the experimental conditions of ΔP : 200 mbar; ΔE : 5 V for electro cleaning; Q_{backwash} : 1,000 mL/h for physical cleaning; 30 min vacuum on/3 min vacuum off; filtration time: 450 min).

functional groups between material and foulants (Fig. 9). The absorption band at $1,743\text{ cm}^{-1}$ is a C–O stretching vibration of the pristine membrane. This peak decreased when electro cleaning was applied due to the accumulation of foulants. The bands at $2,974\text{ cm}^{-1}$ (C–H aliphatic stretching) and $1,365\text{ cm}^{-1}$ (C–H aliphatic bending) are more intense in pristine ACCM.

The photos of the membranes before and after cleaning are shown in Fig. 10. As could be seen from the figure that the membrane without electro-cleaning was heavily coated with contaminants. In contrast, a thinner and drier layer was observed on the membrane surface for 1 min electro-cleaning. During the 3 min electro-cleaning, membrane surfaces were not covered by foulants and uncovered areas with foulants were still observed on the membrane surface. However, the thickest and slimy layer was observed at 5 min electro-cleaning.

The economical comparison of the electro cleaning with physical cleaning was performed according to optimum electro cleaning operating conditions in which ΔP was 200 mbar, the vacuum was on for 30 min and off for 3 min, the voltage was 5 V for 450 min filtration time. Electro-cleaning was performed for a total of 39 min during 450 min. of operation. In electro-cleaning, when the current was 0.3 A and the voltage supply was 5 V, the power consumption was calculated as 0.975 Watt-h for 450 min. of operation while it was 1.950 Watt-h for physical cleaning in which a peristaltic pump was used for backwashing.

4. Conclusion

In this study, we investigated the effectiveness of electro-cleaning to solve the problem of membrane biofouling for MBR and compared to physical cleaning. The results showed that electro cleaning was very effective at 5 V

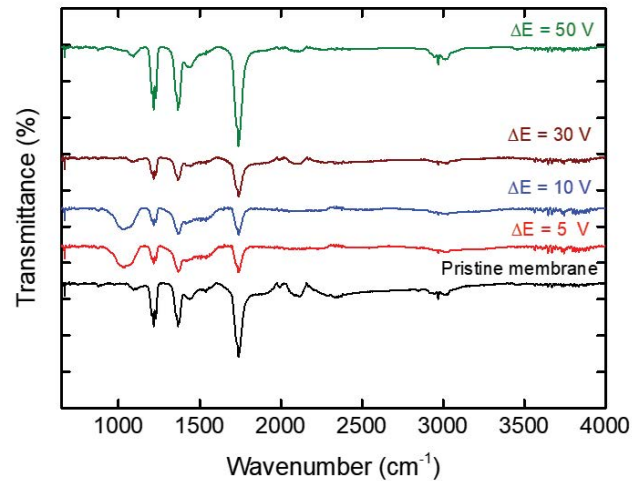


Fig. 9. Fourier-transform infrared (FTIR) spectrum of pristine and fouled ACCM applying 5, 10, 30, 50 V electric current for 3 min electro cleaning time.



Fig. 10. The photos of the membranes before and after cleaning.

for 3 min. When the cleaning time and applied voltage increased, flux was seriously affected negatively. The maximum removal efficiencies of SMP_{c} , SMP_{p} , and COD were determined as 88.1%, 86.5%, and 83.8% for 5 V and 3 min electro-cleaning time while it was about 18.8%, 80.1% and 30.3% for without electro-cleaning, respectively. The flux also increased when electro cleaning was applied on a dynamic membrane. Moreover, R_{ir} decreased under electric current up to 30 V. The results depicted that electro cleaning could control biofilm formation on the dynamic membrane at low voltage and short time.

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