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Fouling control, using various cleaning methods, applied on an MBR system through continuous TMP monitoring

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ABSTRACT

Membrane fouling is the major disadvantage of MBRs, which leads to decreased membrane performance and increased operating expenses. In this study, fouling was monitored in a pilot-scale submerged MBR system fed with municipal wastewater and operated under intermittently aerated conditions. Transmembrane pressure (TMP) was measured online on the membrane module during the whole operating period; permeability and resistance were estimated on a daily basis as well. The most common fouling prevention processes were systematically assessed and optimized. To control TMP increase owing to biosolids accumulation on membrane surface, successive backflushing cycles, backwash volume increase, air-cross flow velocity increase and in/ex situ mechanical cleaning were applied. Hydraulic cleaning resulted in TMP improvement and flux recovery of 40% and 32%, respectively. Ex situ and in situ mechanical cleaning led to TMP improvement of 25% and 39%, corresponding to flux recovery of 63% and 189%, respectively. Increased aeration intensity improved TMP and increased permeate flux by 63% and 56%, respectively. In the case of fouling that was caused by pore blocking and cake layer formation, chemical cleaning was implemented on the membrane module. Extensive chemical cleaning with NaOCl solution led to permeate flux increase of 90%, corresponding to TMP improvement of 44%.

Keywords: Membrane bioreactor; Fouling; Monitoring; Transmembrane pressure

1. Introduction

Membrane fouling in membrane bioreactors (MBRs) decreases permeate yield (flux) and increases energy consumption [1,2]. Flux can be affected by concentration polarization and external and internal membrane deposits [3]. Membrane fouling can be biological, organic and inorganic, as a result of biological and physicochemical properties of membrane foulants [1,2]. The nature of such foulants determines the choice of membrane cleaners. A screening of literature databases has showed that the number of research papers focusing on membrane cleaning has been rapidly increased in the last decade [4–9]. Membrane cleaning is typically classified into in situ and ex situ cleaning based on membrane module cleaning within the MBR or outside the bioreactor [4]. It is divided into physical, mechanical and chemical cleaning based on foulants' removal mechanisms or cleaning agents used. Physical cleaning is applied to remove reversible fouling (e.g., biosolids coats and cake layer) and is usually attained by backflushing (performed under the appropriate frequency and volume), providing an additional decrease of the internal fouling [5], and/or relaxation, which pauses permeation and permits air bubbles to remove solids from membrane surface [10,11], aeration velocity regulation [12,13] and sonication [14,15]. Mechanical cleaning with sponge is an effective way to mitigate on site

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the membrane fouling in MBR systems [6,16]. In addition, in situ and/or ex situ mechanical cleaning practices can regulate the cleaning efficacy [7]. On the other hand, chemical cleaning is applied in emergency membrane fouling cases to remove strongly and absorbed deposits referred as intrapore fouling [4]. The common cleaning agents applied for organic or inorganic fouling (scaling) prevention are sodium hypochlorite (NaOCl) and citric acid ($C_6H_8O_7$), respectively, which offer high cleaning performance. Organic fouling refers to proteins, polysaccharides, humic acids and other organic contaminants that are attached on membrane surface [9] and scaling refers to the formation of salt deposits of inverse solubility onto membrane surface, such as silica, calcium salts and other metal ions [4,15]. Chemical cleaning is mainly performed in situ as maintenance cleaning or intensive cleaning on site [9]. However, the frequency of chemical cleaning and thereby the consumption of chemical cleaning agents is reduced through the use of the aforementioned physical techniques.

The monitoring and controlling of the membrane fouling in MBRs is important since such flux obstacles can affect operating costs [17]. Most of the monitoring approaches are based on sludge filterability estimation and fouling potential, thus, on membrane fouling rate [2]. However, they do not predict the membrane behavior under the applied operating conditions. By measuring the transmembrane pressure (TMP) directly on membrane modules, monitoring of the fouling trend is permitted under transient and steady operating conditions [18].

In the current work, monitoring and recording of TMP pattern were carried out to estimate membrane fouling and to prevent permeate decrease under various cleaning actions. Specifically, strategies, such as application of various aeration velocities, the optimization of backwashing (focusing on both intensity and frequency) and performance of on-site/off-site mechanical and chemical cleaning practices, were examined to evaluate membrane fouling recovery.

2. Materials and methods

2.1. Membrane bioreactor set up

A schematic layout of the pilot-scale MBR system is shown in Fig. 1. A bioreactor tank (100 L) and an external membrane tank (80 L) were the main parts of the pilot-scale MBR system. Biomass recirculation was achieved from the main bioreactor to the external tank at a rate of 187 L/h to keep stable the biosolids concentration. The MBR system was inoculated by activated sludge from a domestic sewage treatment plant. A diffuser plate was installed at the bottom of the aeration tank and the dissolved oxygen was maintained at 2–3 mg/L in the aeration period. An overhead axial blade stirrer was used to mix the activated sludge in the reactor and an air compressor supplied air through a pipeline into a fine bubble diffuser plate at a rate of 1.5 L/min.

The membrane tank was equipped with the flat sheet module BIO-CEL®-LAB of Microdyn Nadir GmbH, which acts as an artificial barrier for retaining effluent biosolids [19]. A fine bubble aeration system (porous tube) was placed just beneath the membrane module to provide air crossflow and prevent clogging. Such approach offers a high oxygen transfer capacity and an enhanced distribution of the air bubbles in comparison with coarse bubble systems [20]. A submersible pump was set in the membrane tank, mixing the activated sludge at a rate of 2,500 L/h. Permeate was obtained by operating a multistage peristaltic pump through a solenoid valve installed over the membrane sheets. On the other hand, the backflow water stream was achieved by using a similar pump and valve. Furthermore, a ball valve was installed in

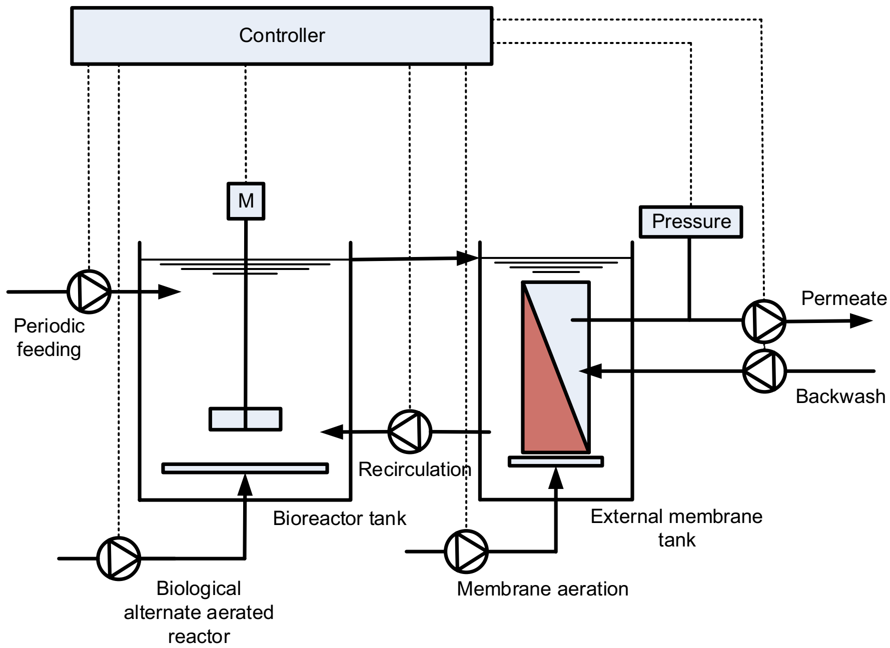

Fig. 1. Schematic illustration of the pilot-scale MBR system.

the flow stream to replace the solenoid valves in the case of damage.

The membrane unit had an effective surface area of 0.34 m^2, consisting of a UP150-flat sheet membrane made by hydrophilic polyethersulfone (PES) sheets of 2 mm thickness and 0.04 μm nominal pore size (cut-off, 150 kDa). The membrane could operate under a wide range of pH and temperature, that is, 2–10 and 5°C–40°C, respectively. Several details on MBR system characteristics and operation conditions are reported in the study by Azis et al. [19].

2.2. Control system operation

A real-time control approach was implemented to oversee and control the membrane filtration efficiency. The programmable logic controller (PLC) program was developed to regulate the permeate flow, the relaxation mode (by turning on/off the permeate pump and the respective solenoid valve) and the backwash flow rate and to record pressure transmitter data. Filtration was performed during the oxic phase, where filtration cycle program consisted of a permeate phase of 480 s, a relaxation phase of 30 s, a backwash period of 60 s and a second relaxation phase of 30 s. The TMP data were online logged (every minute) via the implementation of the Modcan32 software, which interacted with the PLC. To sustain efficient filtration performance, suction duration phase, crossflow aeration, backwash and chemical cleaning (above –200 mbar) were controlled by the CODESYS software, evaluating the on-line acquisition data. The lower and upper pressure limits were –400 and +150 mbar during the filtration and backwash period, respectively. It is noteworthy that emergency backwash cycles (1 min duration) automatically begun by the controller at TMP values above –300 mbar in order to cease filterability and ensure membrane integrity. The monitoring of TMP during each filtration cycle consisted of a permeate phase (ca –134 mbar), an intermediate backwash period (ca +59 mbar) and a no pumping period (ca 0 mbar; Fig. 2a). The increased membrane fouling rate is shown in Fig. 2b, indicating the need for cleaning.

2.3. Wastewater characteristics

The MBR system was fed with sewage obtained from the University Campus of Xanthi. The influent traits were determined as follows: pH, 7.31 ± 0.23; EC, 1,229 ± 170 μS/cm; suspended solids (SS), 201 ± 88.5 mg/L; BOD_5, 149 ± 37.2 mg/L; total COD, 388 ± 196 mg/L; total Kjeldahl nitrogen (TKN), 73.8 ± 12.9 mg/L; ammonium nitrogen (NH_4^+–N), 57.3 ± 15.8 mg/L; orthophosphates, 7.1 ± 2.2 mg/L. All the physicochemical parameters were measured twice per week for a period of 1 year.

2.4. Cleaning processes

A specialized, innovative handheld flexible oblong (length, 60 cm; width, 10 cm) microfiber sponge with thin folds was used for in situ mechanical cleaning without draining the membrane tank. Taking advantage of the specific geometry and the kind of sponge material, the built-up cake layer of the membrane sheets was successfully removed. Regarding the air cross velocities, aeration intensities of 0.06 (4.5 L/min)

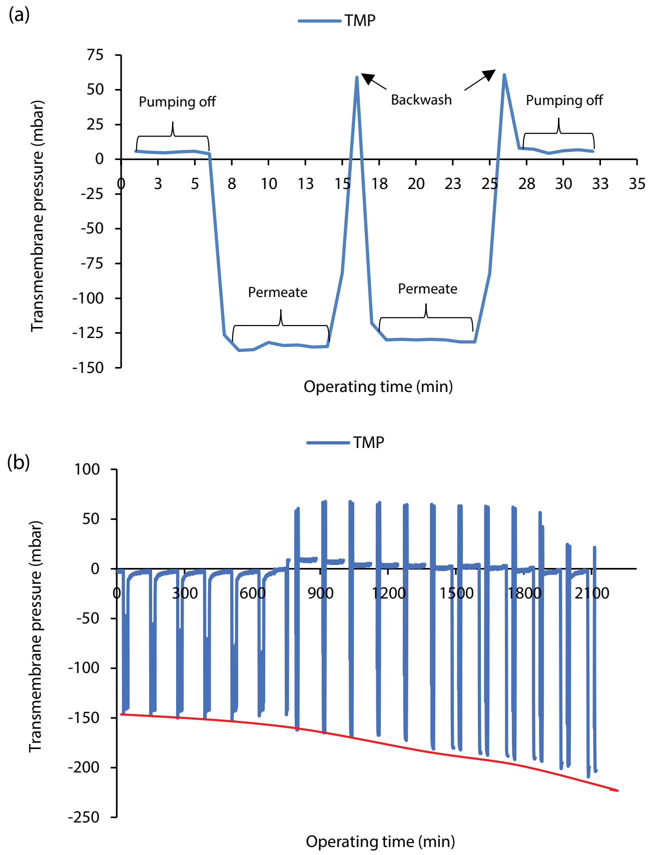

Fig. 2. (a) TMP profile during two successive filtration cycles and (b) TMP profile at increased membrane fouling rate.

m^3/m^2 h, 0.08 (6 L/min) m^3/m^2 h, 0.11 (8 L/min) m^3/m^2 h, 0.14 (10 L/min) m^3/m^2 h and 0.16 (12 L/min) m^3/m^2 h were provided in the membrane tank during the aeration phase. Various backwash fluxes (150, 200 and 400 mL/min) and successive backwash cycles were applied for hydraulic membrane cleaning to improve permeability. The membrane module was chemically cleaned by employing two cleaning methods. An automated cleaning method consisted of two successive steps of (i) an initial aeration phase of 15 min, (ii) a backflush stage, where 500 ppm sodium hypochlorite (NaOCl) solution was added in the backflush water stream for a defined period of 5–10 min, (iii) a soaking time period of 60 min. These steps were performed immediately each, one after the other. In addition, in situ chemical cleaning was applied when the TMP exceeded –200 mbar in order to recover flux and expand membrane lifetime duration. Ex situ intensive cleaning using 500–750 ppm NaOCl was applied for full flux recovery. After trials, the optimal cleaning soaking time was found to be 24 and 12 h for NaOCl and citric acid solution, respectively. Thus, the fouled membranes were soaked in the NaOCl solution for 24 h to remove biofouling, whereas they were rinsed in 0.2% w/v citric acid for 12 h to remove the precipitated salts (inorganic scaling). The membrane tank remained inactive during this cleaning period, where no feeding was performed, so biomass synthesis was restricted. The proposed times of chemical cleaning were in accordance to those reported previously [9,21].

2.5. PLC operation and chemical analysis

An ABB PLC (ASEA Brown Boveri PLC) was programmed through the implementation of the Controller Functionality Software (CODESYS) and the use of Continuous Function Charts as the target language for PLC environments. The dissolved oxygen (DO) concentration was online measured by an optical dissolved oxygen sensor (WTW FDO 700 IQ sensor). NO_3^-–N and NH_4^+–N concentrations were determined by ion chromatographic analysis and the steam distillation method, respectively. All the physicochemical parameters measured were based on protocols reported in APHA manual [22].

3. Results and discussion

The submerged MBR was initially inoculated with activated sludge from the wastewater treatment plant of Xanthi. Regarding loading characteristics, the MBR was operated under an F/M ratio of 0.27 ± 0.1 g BOD_5/g VSS d, an organic loading rate (L_{ORG}) of 0.9 ± 0.2 g BOD_5/L d and a nitrogen loading rate ($L_{N,V}$) of 0.024 g TKN/L d. During the whole experimental period, sludge was not wasted out and the corresponding sludge age (SRT) was identical with the exact day of operation. Permeate flux was ranged between 13.1 and 32.6 L/m² h, while MLSS concentration was ranged from 5 to 9 g/L. The permeability was determined as 275 ± 67 L/m² h bar, which corresponded to a membrane resistance of 2.4 ± 0.5 m^{-1}.

In the effluent of the MBR system, the BOD_5, COD and PO_4^{3-}–P concentrations were 3.4 ± 1.5, 21 ± 12 and 0.7 ± 0.29 mg/L, respectively. Regarding nitrogen removal process, TKN, NH_4^+–N and NO_3^-–N concentrations in the effluent of the MBR were 6.76 ± 1.39, 1.70 ± 0.65 and 0.7 ± 0.5 mg/L, respectively. No differences in the effluent parameters were observed after the performance of the cleaning procedure. In all cases, the effluent characteristics of the MBR met the discharge limits for unrestricted irrigation. The removal efficiencies of BOD_5, COD, TN and PO_4^{3-}–P were 97.9%, 93.2%, 90.5% and 91%, respectively.

3.1. Fouling monitoring by TMP

Membrane fouling phenomena were prevented through the implementation of physicochemical practices for TMP control. Such fouling prevention methods applied were the hydraulic cleaning performed via backwashing, which was achieved either by increasing the water supply rate or the frequency of the backwashes, mechanical cleaning (flushing and scrubbing), aeration capacity increase and in situ or ex situ chemical cleaning, as appropriate.

3.1.1. Hydraulic cleaning (backflushing)

According to Hwang et al. [8] and Raffin et al. [23], solids accumulation on membrane surface can be removed by increasing backflush flux and backwashing replications, improving permeate recovery. To improve transmembrane pressure, the intensity of backwashing was increased from 150 to 200 mL/min (Fig. 3). Thus, the TMP was improved from –282 ± 2.1 to –175 ± 4.5 mbar and the permeate flux was increased from 13.2 to 17.4 L/m² h. The respective permeability was increased from 46.8 to 99.4 L/m² h bar. To sustain TMP lower than –180 mbar, backwash rate was then increased from 200 to 400 mL/min, improving the TMP by 40% from 175 ± 4.5 to 105 ± 1.4 mbar and increasing permeability from 99.4 to 126 L/m² h bar. In addition, three successive backwash cycles (1 min duration each) were applied at a maximum pump rotating speed to improve TMP. TMP was improved from –105 ± 1.4 mbar to –71 ± 1.6 mbar, resulting in a significant permeability increase (from 126 to 221 L/m² h bar) and resistance decrease (from 3.6 to 2.7 m^{-1}). Indeed, the reversible fouling can be eliminated by frequent backwashing and this practice was also confirmed by Yigit et al. [5].

3.1.2. Mechanical cleaning

Another cleaning approach included the removal of membrane module and its intensive wash by tap water flushing. This resulted in a TMP reduction from –173 ± 3.6

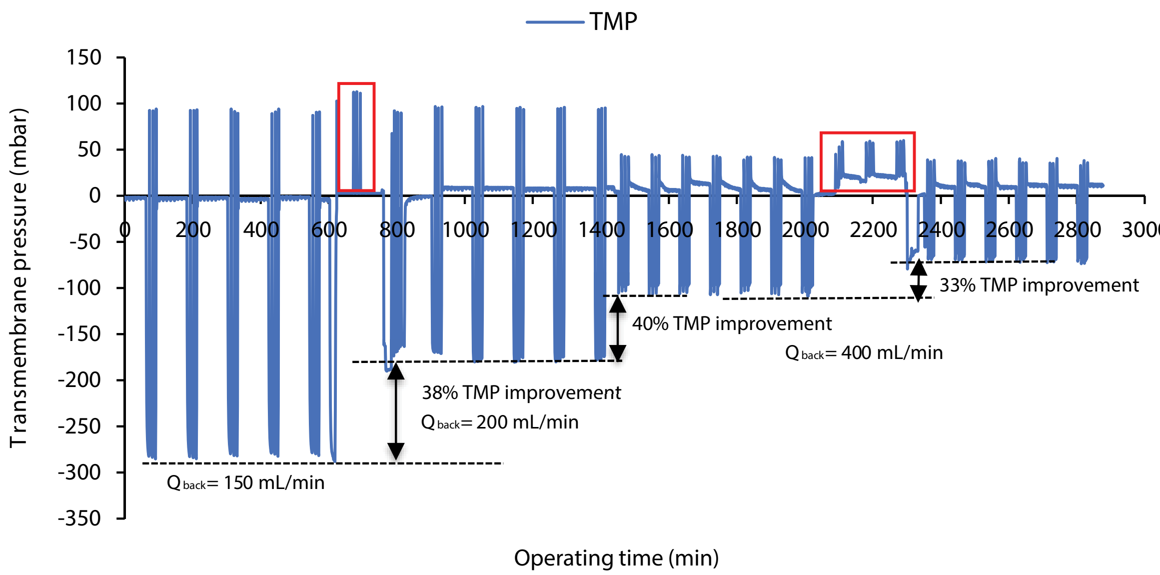

Fig. 3. TMP profile after gradual backwash volume increase and successive backflushing applications.

to −131 ± 3.7 mbar (decreased by 24.5%), improving permeate flux from 13.1 to 21.4 L/m² h (Fig. 4). The respective permeability was increased from 83.5 to 164 L/m² h bar, whereas the corresponding resistance was dropped from 6.8 to 3.5 m^{-1}. This cleaning step resulted in sustaining membrane integrity for about 3 months. According to Van den Brink et al. [24], fouling cannot be removed completely by harsh mechanical cleaning, but partially by superficial external clogging of pores.

A second mechanical effort to reduce TMP and improve permeate flux was the in situ cleaning of the membrane surface with a flexible brush without washing, which resulted in scrapping the fouling 'cake' layer. The efficacy of this cleaning method was ensured by the high TMP decline as illustrated in Fig. 5. After membrane layer scrapping, TMP was decreased (by 39%) from −167 ± 3.9 to −102 ± 3.8 mbar and permeability was fivefold increased (from 55 to 245 L/m² h bar), while the membrane resistance was decreased from 6.7 to 1.5 m^{-1}. In comparison with intensive wash with tap water, this kind of mechanical cleaning led to higher filterability, resulting in flux increase by 63% (from 9.4 to 25.3 L/m² h).

3.1.3. Air scouring

Air scouring is used to prevent cake layer formation on the membrane surface [25], avoiding concentration polarization and fouling [26]. In the current work, when TMP reached −350 mbar (Fig. 6), a cross-flow bubble strategy, corresponding to a specific aeration capacity increase from 4.5 to 6.0 L/min, was employed to maintain a stable flux, improving the TMP from −343 ± 4 to −289 ± 18.1 mbar.

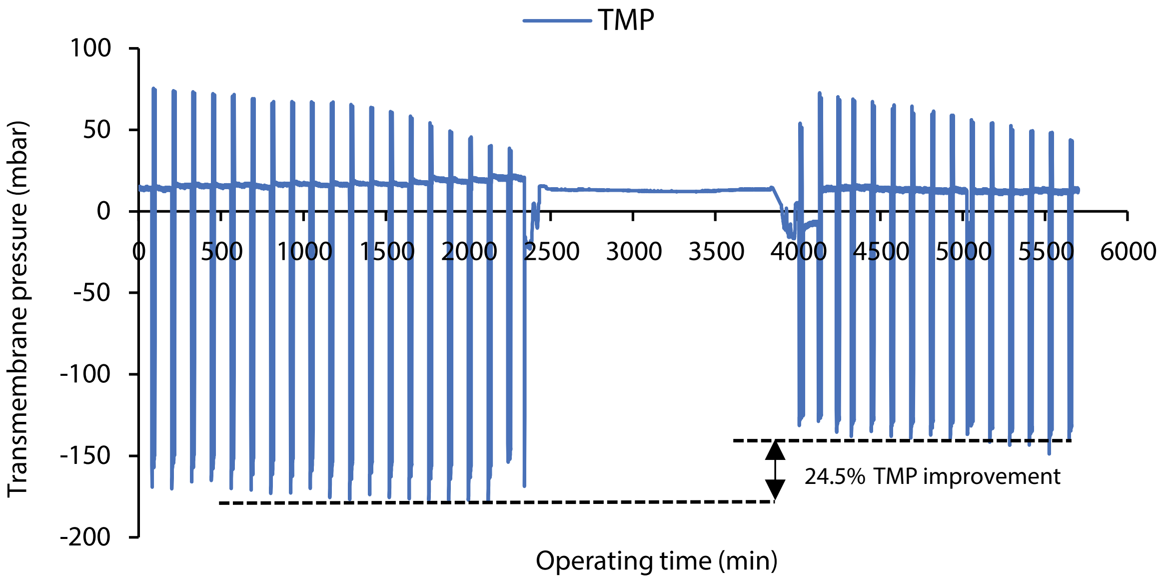

Fig. 4. TMP profile after intensive wash with tap water.

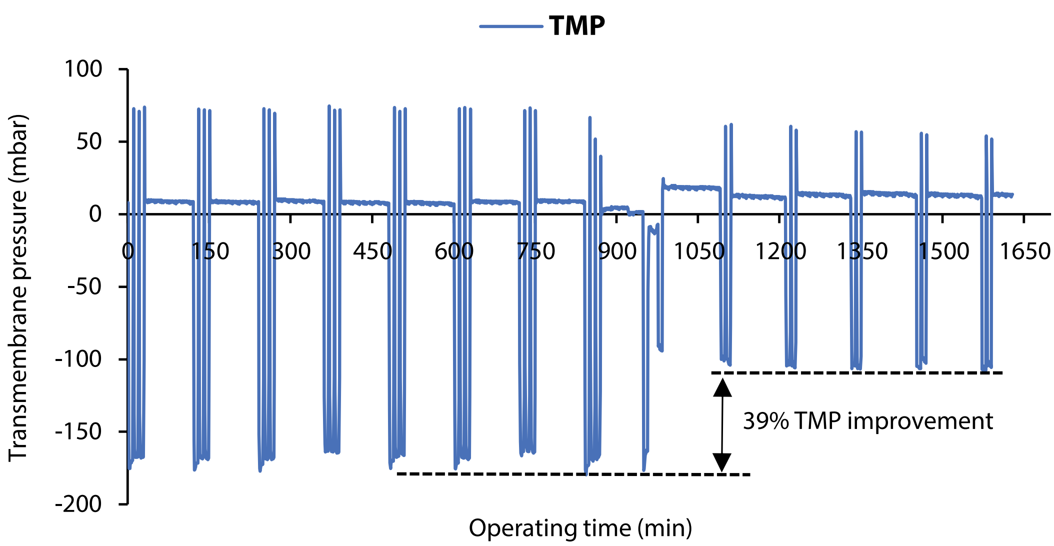

Fig. 5. TMP profile after in situ membrane layer scrapping.

The permeability was then increased from 38.6 to 47.7 L/m² h bar. Aeration intensity increase to 8.0 L/min improved the TMP from –289 ± 18.1 to –215 ± 1.8 mbar, the permeability from 47.7 to 81 L/m² h bar and the membrane resistance from 12 to 6.3 m⁻¹. A further aeration velocity increase to 10 L/min resulted in TMP decrease from –215 ± 1.8 to –134 ± 2.9 mbar, in permeability increase from 81 to 151 L/m² h bar and in membrane resistance decrease from 6.3 to 2.5 m⁻¹. Lastly, aeration velocity increase to 12 L/min decreased TMP from –134 ± 2.9 to –122 ± 2.3 mbar, increased permeability from 151 to 181 L/m² h bar and decreased membrane resistance from 2.54 to 2.11 m⁻¹. Thus, the optimal aeration velocity was 12 L/min since the deposition of large particles on the membrane surface was prevented [20,27].

3.1.4. Chemical cleaning

Chemical cleaning by using sodium hypochlorite or/and citric acid can effectively control the irreversible fouling [4]. Thus, a membrane that is chemically and biologically clean, free of organic foulants, can provide adequate flux [7]. In the current study, in situ chemical cleaning with 500 ppm NaOCl solution only slightly improved TMP (from –197 ± 3.4 mbar to –180 ± 3.1 mbar) and permeability (from 31 to 36 L/m² h bar; Fig. 7). The membrane resistance was decreased from 18.3 to 14.5 m⁻¹.

Acids are mainly used to dissolve calcium precipitates on membrane surface in order to recover flux [9]. Some organic acids, such as citric acids, are less corrosive than inorganic acids and are applied to remove effectively inorganic salts and metal oxides from the membrane [4,7]. In this work, ex situ chemical cleaning with 0.2% w/v citric acid solution was performed, resulting in TMP decrease from –181 ± 4.2 to –127 ± 2.3 mbar and permeability increase from 36 to 180 L/m² h bar. The membrane resistance was dropped down from 14.5 to 3.19 m⁻¹ (Fig. 8).

In addition, ex situ chemical cleaning with 750 ppm NaOCl solution was also performed (Fig. 9). This led to

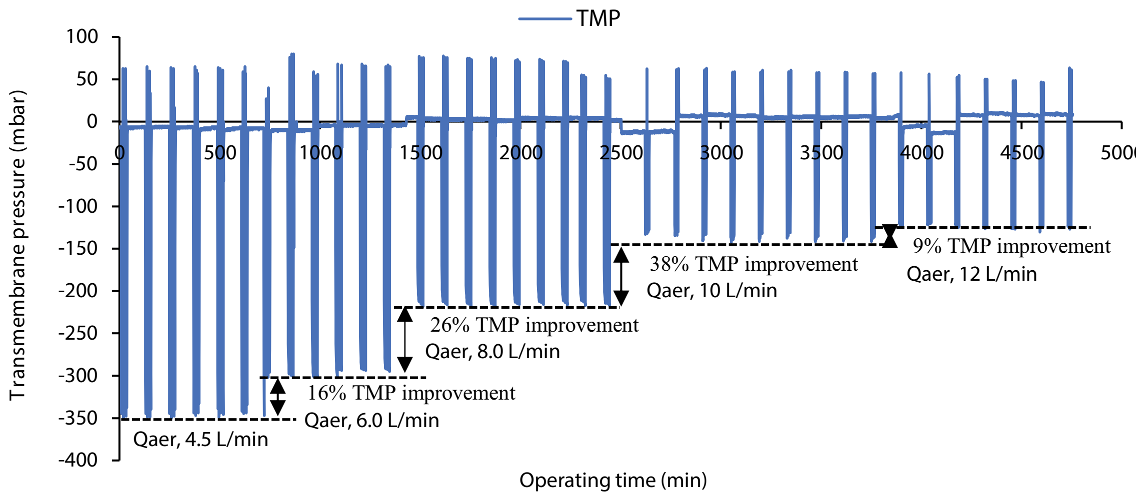

Fig. 6. TMP profile under various aeration velocities for air scouring.

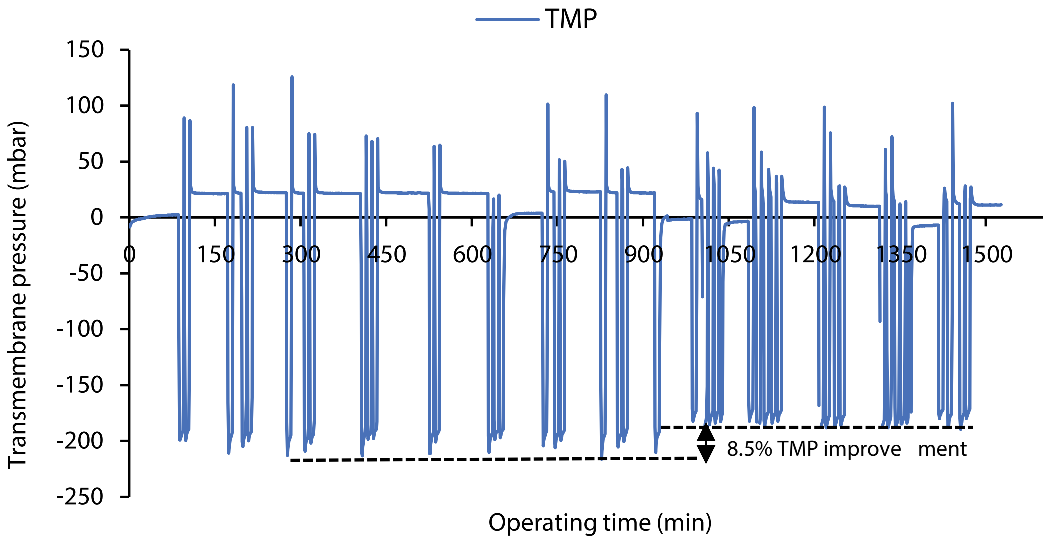

Fig. 7. TMP profile after in situ chemical cleaning with 500 ppm sodium hypochlorite solution.

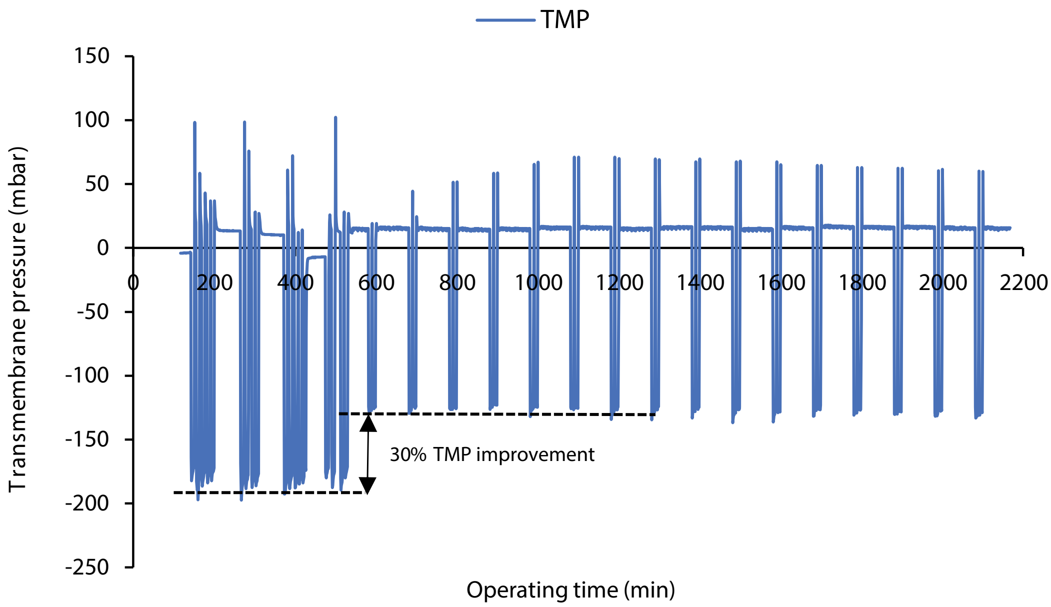

Fig. 8. TMP profile after ex situ chemical cleaning with 0.2 w/v citric acid solution.

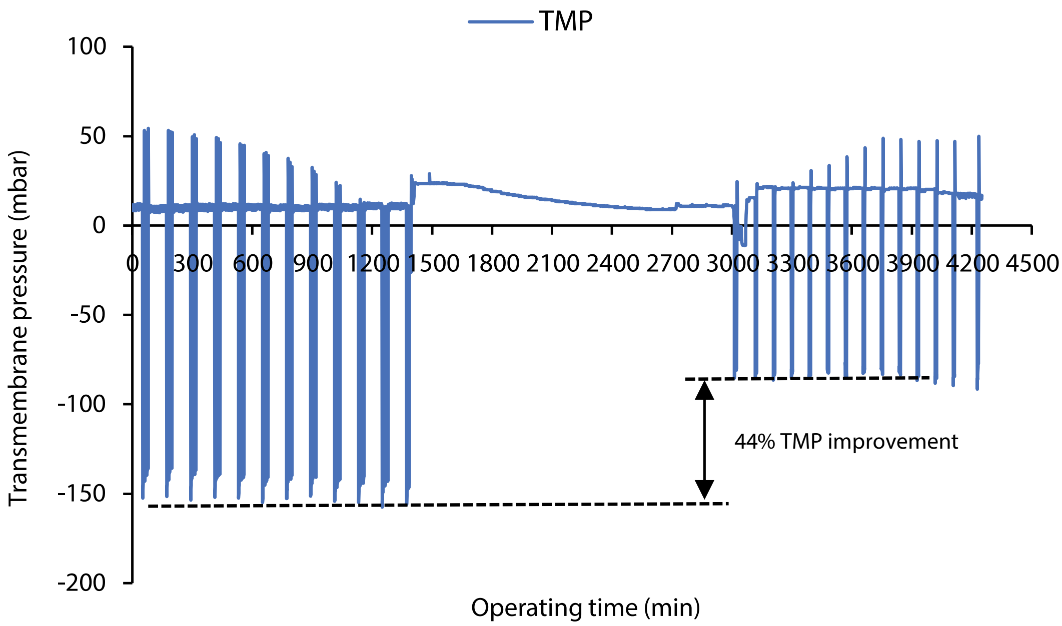

Fig. 9. TMP profile after ex situ chemical cleaning with 750 ppm NaOCl solution.

TMP reduction from –153 ± 1.5 to –85 ± 3.1 mbar, whereas membrane permeability was increased from 123 to 404 L/m^2 h bar. The membrane resistance was decreased from 3.79 to 1.17 m^{-1}. Interestingly, Puspitasari et al. [28] reported 20% resistance decrease during membrane treatment with 2% w/v NaOCl solution.

5. Conclusions

It is concluded that TMP monitoring is an effective tool to detect the membrane fouling grade in order to apply the appropriate cleaning method. Regarding reversible fouling, backflushing rate increase as well as mechanical cleaning with pressurized water and sponge was considered as effective approache to improve TMP by 25%–40%. Aeration intensity increase was also sufficient for reversible fouling prevention, resulting in TMP improvement by 61%. Indeed, combination of hydraulic and mechanical cleaning approaches can effectively control reversible fouling. Regarding irreversible fouling, extensive chemical cleaning with NaOCl and citric acid solution can restore membrane efficiency by 44% and 30%, respectively. In comparisons to previous studies [6,28,29], the optimization of the reversible and irreversible fouling prevention methods applied in the current study resulted in the recovery of TMP values.

Acknowledgments

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Author contributions

P. Melidis and K. Azis conceived and designed the experiments; K. Azis performed the experiments; P. Melidis, K. Azis and S. Ntougias analyzed the data; P. Melidis, K. Azis and S. Ntougias wrote the paper.

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