Efficient fouling control using outer-selective hollow fiber thin-film composite membranes for osmotic membrane bioreactor applications

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Abstract

This paper investigates the efficiency of fouling mitigation methods using a novel outer selective hollow fiber thin-film composite forward osmosis (OSHF TFC FO) membrane for osmosis membrane bioreactor (OMBR) system treating municipal wastewater. Two home-made membrane modules having similar transport properties were used. Two operation regimes with three different fouling mitigation strategies were utilized to test the easiness of membrane for fouling cleaning. These two membrane modules demonstrated high performance with high initial water flux of 14.4 LMH and 14.1 LMH and slow increase rate of mixed liquor's salinity in the bioreactor using 30 g/L NaCl as draw solution. OMBR system showed high removals of total organic carbon and NH4+-N (> 98%). High fouling cleaning efficiency was achieved using OSHF TFC FO membrane with different fouling control methods. These results showed that this membrane is suitable for OMBR applications due to its high performance and its simplicity for fouling mitigation.

Keywords: *Outer-selective hollow fiber; Thin-film composite; Osmotic membrane bioreactor; Municipal wastewater treatment; Membrane fouling control.*

1 Introduction

Water scarcity has been becoming a severe problem for over several decades all over the world due to the rapid increase of population and also because of the impact of climate change. Wastewater reclamation is considered one of the sustainable solutions for addressing the water shortage issue. Technologies for wastewater treatment and reuse are already available in the market especially using advanced membrane processes. Among the currently available technologies, membrane bioreactor (MBR) technology has been one of the most popular options for water reuse for improving water sustainability. Compared to traditional wastewater treatment systems such as conventional activated sludge (CAS), this technology has several advantages such as stable and high effluent quality, simple flow configuration, lower surplus sludge production, high volumetric loading, and smaller footprint (Guo et al., 2012). Albeit the advantages mentioned above, there are still challenges requiring MBR technology to overcome such as high power consumption, limited nutrient removal capacity and membrane fouling (Nguyen et al., 2012).

Since the regulations are getting more stringent on reclaimed water quality, membrane technologies are gaining more popularity in wastewater treatment and reuse applications. Forward osmosis (FO) - an emerging technology has gained growing interests due to a number of advantages such as high quality of product water, low fouling tendency, and low energy consumption (Cath et al., 2006; Lay et al., 2012). Achilli et al. (2009) proposed to integrate FO into conventional MBR which was then called osmotic membrane bioreactor (OMBR). In OMBR system, the pure water from the feed water is forced to pass through a selectively permeable membrane to draw solution side by osmotic driving force generated by draw solution. Due to the utilization of osmotic pressure, OMBR has lower fouling propensity than that of conventional MBR. In addition, due to high rejection of FO membranes, OMBR produces higher quality water compared to MBR (Achilli et al., 2009).

As one of the FO applications, OMBR suffers from one of the major hurdles of FO process such as lack of a suitable membrane (Yip et al., 2010). Over the last decade, researchers and industries have been attempting to develop an ideal membrane which should possess the following properties: (i) thin selective layer to provide high water permeability and high ion rejection, (ii) porous membrane substrate to lessen internal concentration polarization (ICP), and (iii) low fouling propensity (Lim et al., 2017). Furthermore, FO membrane must have the reasonable mechanical strength to retain to the harsh operating conditions with activated sludge in OMBR. With an optimal membrane, currently inherent drawbacks of OMBR including severe flux decline, high salinity build-up, and membrane fouling can be mitigated.

A plethora of studies has been conducted to investigate the performance of commercially available and home-made FO membranes applied into OMBR (Wang et al., 2016a). Among these investigated membranes, most of them were in the configuration of flat-sheet membranes including cellulose triacetate (CTA) membrane (Alturki et al., 2012; Holloway et al., 2014; Holloway et al., 2015; Luo et al., 2016a; Qiu and Ting, 2014; Qiu and Ting, 2013; Wang et al., 2014a; Wang et al., 2014b) and thin film composite (TFC) polyamide membrane (Blandin et al., 2018; Luo et al., 2016b; Morrow et al., 2018; Pathak et al., 2017; Pathak et al., 2018; Zhang et al., 2017) produced by Hydration Technology Innovations (HTI, Albany, OR) and Toray Industries, Inc. (Toray) respectively. There was only one of OMBR study that investigated the performance of an inner selective hollow-fiber (ISHF) TFC membrane made in-house at the Singapore Membrane Technology Centre (Zhang et al., 2012b). Zhang et al. (2012b) used inner-selective hollow fiber TFC membrane having polyamide selective layer on its lumen side under the orientation of active layer facing draw solution (AL-DS) while the support layer faced activated sludge. Although the performance of the membrane in terms of water flux was better due to the absence of internal concentration polarization (ICP) effects, membrane fouling was observed as severe issue. Foulants and particulates from the activated sludge easily deposit inside the pores of the support layer giving rise to pore clogging especially when a high suspended solid mixed liquor is used. This leads to sudden deterioration of the performance of the OMBR system and hence membrane fouling control becomes a significant challenge. If the inner selective hollow fiber TFC membrane is operated with the active layer facing feed solution (AL-FS) orientation, supplying mixed liquor containing particulates, foulants and flocculated sludge into the smaller bore side of hollow fiber membrane can be highly prone to clogging and hence fouling control could pose considerable challenges due to the small inner diameter of the membrane. However, the outer-selective hollow fiber (OSHF) TFC membranes are therefore expected to be the most suitable candidate for OMBR and in particular for submerged applications. OSHF TFC membranes have the edge over the ISHF TFC membranes because of (i) more substantial surface area per fiber, (ii) lower fouling propensity, and (iii) ease of fouling control (Cheng et al., 2016; Le et al., 2016).

The objectives of this study are to assess the efficiency of different fouling mitigation strategies using a novel OSHF TFC FO membrane developed in-house for the first time in OMBR application and to investigate the performance of these two membrane modules. The OMBR system, which used side-stream OSHF TFC FO membrane modules, was operated under two operation regimes. This system used 30 g/L NaCl as draw solution simulating the total dissolved solid concentration of future RO brine at Central Park Recycled Water Facility (CPRWF). Three fouling mitigation strategies were sequentially carried out during operation of each regime to restore water flux from fouling. The investigation focused on the performance of OSHF TFC membrane based on following key factors: (i) water flux, (ii) reverse solute flux or salinity buildup in the mixed liquor, (iii) removal of primary contaminants, and (iv) membrane fouling and efficiency of fouling control strategies. To the best of our knowledge, there has been no study investigating the performance of OSHF TFC FO membranes applied in OMBR.

2 Materials and methods

2.1 *Outer selective hollow fiber thin-film composite forward osmosis membrane and module.*

2.1.1 Membrane module

This study used two OSHF TFC FO membrane modules which were locally made in the laboratory of Centre of Technology for Water and Wastewater, University of Technology, Sydney (UTS), Australia. Each membrane module comprises of 8 fibers with the length of 11 cm, and the effective surface area of the membrane module was approximately 22.77 cm^2. Both ends of the module were carefully potted using epoxy resin to minimize deformation of the membrane fibers.

2.1.2 Determination of intrinsic transport properties and performance of membrane module

Intrinsic transport properties of OSHF TFC FO membranes such as water permeability coefficient (A), solute permeability coefficient (B) and structural parameter (***S***) values were estimated using a FO model algorithm developed by Tiraferri et al. (2013). The FO performance (water flux and specific reverse solute flux (SRSF)) of the home-made OSHF TFC membranes used in this study was investigated using a lab-scale FO membrane experiment unit. The performance tests were conducted using DI water as FS and 30 g/L NaCl as DS under AL-FS orientation. 2L of DS was circulated with a flowrate of 25 mL/min

(13.9 cm/s) in the lumen side of OSHF TFC FO membrane. 2 liters of FS was circulated with a flowrate of 1.0 L/min (5.4 cm/s) at the outer side of the membrane.

2.2 *The lab-scale OMBR system*

[Insert Figure 1]

A laboratory-scale side-stream OMBR system as shown in Fig. **1** was used in this study. This system consists of a feed solution tank, a transparent acrylic plastic bioreactor tank with an adequate volume of 5 L, a newly developed innovative OSHF TFC FO membrane module as described above was installed inside stream mode, a draw solution tank, two variable speed gear pumps (Cole Palmer model 75211-15, 50-5000 rpm and 0.07 HP, Thermo Fisher Scientific, USA), a conductivity and pH meter (HACH, Germany), a balance (Nimbus Precision Balances: NBL 4602e, ADAM Equipment, USA) connected to a computer for online data logging.

2.3 *Synthetic wastewater and draw solution*

Otherwise stated, in this study chemicals used were of the reagent grade (Merck, Australia). Artificial wastewater was used as influent for OMBR system. This synthetic wastewater influent was prepared daily and comprised of 300 mg/L glucose, 30 mg/L urea, 15 mg/L KH_2PO_4, 50 mg/L yeast, 10 mg/L $FeSO_4$, and 60 mg/L $(NH_4)_2SO_4$. This OMBR influent provides a total organic carbon (TOC) of 109.24 ± 3.25mg/L; 306 ± 12 mg/L chemical oxygen demand (COD), 17.68 ± 0.58 mg/L ammonium nitrogen (NH_4^+-N), 25 ± 3 mg/L total nitrogen (TN) and 3.58 ± 0.12 mg/L phosphate (PO_4-P). Sodium chloride (NaCl) at analytical grade (Sigma Aldrich, Merck, Australia) with the concentration of 30 g/L was used as the draw solution in this study. The 30 g/L of NaCl was used as DS to simulate the RO brine at CPRWF. Draw solution was changed daily to avoid the accumulation of contaminant.

2.4 *Operation of OMBR system*

Seed activated sludge was initially collected for acclimatization from the recycled water facility at Central Park, Sydney, Australia. It was acclimatized for two months until the removal performance of the system has achieved relatively stable and effective removal of total organic carbon (TOC) (> 90%). The mixed liquor suspended solid (MLSS) was then adjusted to 5.5 g/L which is in the bioreactor’s MLSS range at CPRWF before being added into the OMBR system. During the acclimatizing process, the total dissolved solid (TDS) of mixed liquors was also maintained in a range of 550 mg/L - 650 mg/L which is similar to mixed liquor’s TDS value at CPRWF.

The influent was continuously fed into the OMBR using a floating ball valve to control the water level in the reactor. Air flowrate of 3 L/min was provided using an air stone (Aqua One, Australia) to supply oxygen at a concentration of above 3 mg/L for microorganisms. Since this is a side-stream OMBR system, mixed liquor was used as feed solution and was circulated at the flow rate of 1.0 L/min (equivalent to a crossflow velocity of 5.4 cm/s) from the reactor through OSHF membrane module and back to the reactor. Draw solution was circulated at the flow rate of 25 mL/min (equivalent to a crossflow velocity of 13.9 cm/s) from a 2 L acrylic tank through the OSHF membrane module and back to the tank. The OMBR system was continuously operated for 6 days under regime 1 using a pristine membrane module OSHF-1. A new batch of mixed liquor having similar characteristics to the one used in regime 1 and a pristine membrane module OSHF-2 were used for another on-going 8 day operation under regime 2 using. Operation was remained stable for both regimes at a constant temperature of 22 ± 1˚C. As for operation regime 1, after every 12 hours of operation, OMBR system was temporarily paused for OSHF membrane cleaning, changing draw solution and sampling. As for operation regime 2, one cycle of operation was 24 hours prior to membrane cleaning. Salinity accumulation and pH of the mixed liquor in the bioreactor were measured by regularly monitoring total dissolved solids (TDS) and pH of the mixed liquor with a multi-parameter meter (HACH, Germany).

2.5 *Membrane fouling mitigation methods*

In this study, three cleaning methods were utilized at different stages of both operating regimes depending on the rate of water flux decline. These cleaning strategies are described below:

2.5.1 Physical cleaning

Physical cleaning was first applied at initial cycles until water flux dropped by 50% of the initial water flux of the very first cycle (cycle 1). Deionized (DI) water was used in this cleaning strategy. Physical cleaning was implemented for 15 minutes by increasing flow rate of DI water at the feed side to 1.5 L/min (equivalent to a crossflow velocity of 8.07 cm/s) while maintaining the crossflow velocity in the lumen side of OSHF membrane.

2.5.2 Osmotic backwashing

Osmotic backwashing was carried out when the water flux recovery of physical cleaning achieved below 70% and water flux declined by 50 % of the initial water flux at cycle 1. Osmotic backwashing was conducted as per the following procedure:

Step 1: OSHF membrane was washed for 5 minutes using DI water to remove mixed liquor inside the membrane module. To accomplish this, DI water was circulated through inside and outside of the fiber with the same crossflow velocities of physical cleaning (8.07 cm/s at outside, 13.9 cm/s at the lumen side).

Step 2: A 30g /L NaCl solution was used as an osmotic backwash agent and was circulated at a flow rate of 1.0 L/min outside the fiber, while DI water was circulated in the lumen side of the fiber at a flow rate of 25 mL/min. This main step of osmotic backwashing was operated for 1 hour.

Step 3: the first step was repeated with the replacement of NaCl solution by DI water, to remove residual NaCl in the membrane module before the next cycle.

2.5.3 Chemical cleaning

A two-step sequential chemical cleaning was conducted using two types of cleaning agents including: 0.1% sodium hydroxide (NaOH) + 0.1% sodium dodecyl sulfate (SDS) (w/v) providing an alkaline solution (pH = 12); and an acidic solution comprising of 2% citric acid (w/v) (pH = 3). Chemicals used in this section were at reagent grade and used as received from Sigma Aldrich (Australia). A protocol of chemical cleaning strategy is detailed herein below:

Step 1: OSHF membrane was washed for 5 minutes using DI water to remove mixed liquor inside the membrane module. At this step, DI water was circulated on both sides of the fiber with the same crossflow velocities of physical cleaning (8.07 cm/s at outside, 13.9 cm/s at the lumen side).

Step 2: The alkaline solution was introduced outside of OSHF TFC FO membrane with an identical operating condition as working condition (5.4 cm/s) while DI water was kept circulating with 13.9 cm/s in the lumen side. This cleaning agent was circulated at the outer side of fiber for 1 hour.

Step 3: After 1 hour circulation, alkaline solution was drained and replaced by DI water. DI water was then circulated at a crossflow velocity of 8.07 cm/s for 5 minutes to remove residual of cleaning agent before introducing the second cleaning agent.

Step 4: This step was carried out by circulating citric acid solution at a crossflow velocity of 5.4 cm/s for 1 hour. This process aims to clean inorganic foulants which were not removed by the alkaline agent. DI water was kept circulating in the lumen side of fiber with a crossflow velocity of 13.9 cm/s.

Step 5: A similar method as highlighted in step 3 was implemented to remove residual of citric acid in the membrane module before the commencement of the next cycle.

2.6 *Analytical methods*

2.6.1 *Water flux measurement*

The experimental water flux J_w (L/m^2 h - LMH) was determined using following equation:

$$J_w = \frac{\Delta V}{A_m \times \Delta t} \quad (1)$$

Where: ΔV is the net increase in the diluted DS volume (L) recorded by a balance over a time interval Δt (h) using an effective area of FO membrane A_m (m^2).

2.6.2 *Biological and necessary water quality parameters*

All characteristic parameters of mixed liquor in the OMBR system such as mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS) and chemical oxygen demand (COD) were analyzed following standard methods for the examination of water and wastewater (APHA, 2005). A dissolved oxygen (DO) meter (Vernier, USA) was used for monitoring the DO concentration in the mixed liquor during experiments. TOC concentrations of the water samples collected from influent, effluent and diluted draw solution were determined using the Analytikjena Multi N/C 2000. NH_4^+-N concentration was measured using Ammonium Test and photometer (Spectroquant, NOVA 60, Merck) supplied by Merck. In order to attain accurate analytical values, water samples from diluted draw solution were diluted several times to minimize the interference of chloride and ensure the proper range of analytes.

2.6.3 *Membrane autopsy*

Samples of pristine membrane and fouled membrane were collected for analysis of surface and cross-sectional morphologies of membrane surface and fouling cake layer. Before being coated with gold using a high vacuum sputter coater (EM ACE600, Leica), these membrane samples were dried at room temperature, for cross-sectional analysis, nitrogen liquid was used for freezing and fracturing membrane samples. Images of morphological structures and elemental composition of membrane samples were then captured using a field emission scanning microscope and energy diffusive X-ray (EDX) analyzer (FE-SEM, Zeiss Supra 55VP, Carl Zeiss AG) operated in a voltage range of 5 – 10 kV with different image magnification.

3 Results and discussion

3.1 *Characteristics of outer-selective hollow fiber membrane module*

[Insert Table 1]

Intrinsic transport properties of two membrane modules such as water permeability coefficient (A); salt permeability coefficient (B); and structural parameter (S) are shown in Table **1**. In general, these three A, B and S values of two OSHF TFC FO membrane modules used in this study were relatively similar in which OSHF-1 showed faintly better properties with higher A value, lower B value, and lower S value. The A values for OSHF-1 and OSHF-2 membranes were 2.26 LMH bar^{-1} and 2.24 LMH bar^{-1} while their B values were 0.28 LMH for OSHF-1 and 0.30 LMH for OSHF-2. The intrinsic selectivity (B/A) of OSHF-1 was 0.12 bar which was slightly lower than that of OSHF-2 with 0.13 bar. Furthermore, the *S* values of OSHF-1 was 190 µm which was lower than 195 µm for OSHF-2. The marginal difference of these transport properties of two OSHF TFC FO membrane modules indicates the stability in the fabrication of OSHF TFC FO membrane modules, which is very important for the consistency of two operation regimes.

The FO performances of the two OSHF TFC membrane modules used in this study were assessed**.** OSHF-1 and OSHF-2 achieved excellent performances in terms of water fluxes and

SRSF, exhibiting high and similar water fluxes of 16.5 LMH and 16.3 LMH, respectively. These membranes also showed a low SRSF value of 0.14 g/ L for OSHF-1 and of 0.15 g/L for OSHF-2. OSHF-1 showed slightly higher water flux and relatively lower SRSF compared to OSHF-2.

3.2 *Water flux and mixed liquor salinity.*

[Insert Figure 2]

Fig. **2(a)** shows the changes in water flux of OMBR system using two long-term operating regimes. In the first regime, OMBR system was run continuously over a period of 6 days using membrane module OSHF-1 and operating cycle of 12 hours while a 24-hour cycle was applied for 8 consecutive days of OMBR operation in regime 2using membrane module OSHF-2. As can be seen, water fluxes of both regimes continuously dropped following a similar pattern. The initial water fluxes for the first two cycles of both were relatively similar for these two regimes with 14.4 LMH and 14.1 LMH for regime 1 and regime 2, respectively. These water fluxes then gradually declined before the commencement of three cleaning methods. This result is in good agreement with studies reported previously and could be due to salinity accumulation in the bioreactor and membrane fouling (Zhang et al., 2017). Another reason for the flux decline in this study was the continuous dilution of the draw solution as the operation was conducted in a batch mode in which both the DS and FS were recycled back to their respective reactors. Salinity build-up in the bioreactor increases the osmotic pressure in the mixed liquor side while dilution of draw solution reduces the osmotic pressure of draw solution side, and hence; the reduction of net driving force for water permeation.

There are three distinct periods of water flux decline can be observed in Figure **2(a)** for both regimes. In the first period, 6 cycles of regime 1 and 3 cycles of regime 2 were run, physical cleaning method was applied at the end of each cycle. Water fluxes gradually dropped to approximately 8.0 LMH at the end of these cycles. During this first period, fouling was not severe therefore the primary reason of flux decline was ascribed to the dilution of draw solution and salinity build-up in the reactor. The second period of flux drop occurred at cycle 7, 8, and 10 of regime 1 and cycle 4, 5, and 6 of regime 2 when fouling became more critical. This phenomenon was one additional reason caused flux significantly decreased to 6.9 LMH and 7.5 LMH at the end of cycle 8 (regime 1) and cycle 6 (regime 2), respectively in the second period.

In the third period, osmotic backwashing and chemical cleaning were carried out which showed high recovery of water flux. Water flux recoveries of two fouling mitigation strategies indicated that chemical cleaning was much more effective than that of osmotic backwash. As for regime 1, initial water flux of cycle 9 after osmotic backwash was at 10.0 LMH while it reached 12.9 LMH when the membrane was chemically washed. Regime 2 experienced a similar pattern with lower water flux recovered at 10.3 LMH after fouled membrane was cleaned using the backwashing method, while it achieved 13.4 LMH after chemical cleaning method was used to mitigate fouling issues. Interestingly, flux decline in regime 1 was more severe even though the salinity build-up in the reactor of regime 1 was less compared to its regime 2. This could be attributed to higher initial flux and greater permeate that regime 1 obtained compared to that of regime 2.

One of the inherent issue of OMBR system is the accumulation of salinity in the mixed liquor caused by reverse diffusion of draw solute and the high rejection of membrane (Luo et al., 2017). Fig. **2(b)** shows the salinity increase in terms of total dissolved solids (TDS) of mixed liquor in the reactor over two operation regimes of OMBR system. Salinity build-up during these two regimes followed similar pattern with a slightly slower increase of the regime 2. After 6 days operation with regime 1, TDS in the reactor rose from 528 mg/L to 916 mg/L, while it raised from 590 mg/L to 1,012 mg/L when OMBR system was operated with regime 2 over 8 days. Results proved that OSHF TFC FO membrane had consistent rejection and low reverse solute flux during the operation of OMBR. The increase of mixed liquor TDS contributed to the flux drop since it reduced the net osmotic pressure. The increments of mixed liquor TDS before and after cleaning were relatively consistent indicating that fouling control strategies did no damage to the membranes.

3.3 *Efficiency of fouling mitigation methods*

Despite FO being generally considered a low-fouling membrane process due to lower flux conditions and the absence of hydraulic pressure , it can still subject to the risk of particle deposition, biofilm formation, and scaling under unfavorable conditions (Liu and Mi, 2012; Zhang et al., 2012a). Membrane fouling not only deteriorates membrane separation performance but also reduces membrane lifespan and operating costs (Holloway et al., 2015; Liu and Mi, 2012; Zhang et al., 2014). In this study, for each operation regime, three fouling mitigation strategies (physical cleaning; osmotic backwashing; and chemical cleaning) were sequentially carried out aiming to remove fouling cake layer and to recover water flux.

[Insert Figure 3]

Fig. **3** shows an initial water flux and flux recovery rate of OSHF TFC FO membrane after being cleaned by different fouling mitigation methods. In general, the membrane cleaning efficiency and flux recovery trend were comparatively similar for two operation regimes. Enhanced physical cleaning was used for the first 8 cycles of regime 1 (12-hour cycle time) and the first 6 cycles of regime 2 (24-hour cycle time). This method demonstrated an outstanding cleaning efficiency, where the membrane after 15 minutes of hydraulic cleaning using DI water at enhanced CFV restored water flux to 96.0%. For the operation regime 2, flux recovery rate was slightly higher at 97.1% compared to regime 1. This can be ascribed to the lower initial flux of 13.8 LMH at the very first cycle of regime 2 compared to 14.0 LMH of regime 1 and hence fouling deposition on the membrane is expected to be slightly lower under regime 2. The effectiveness of this physical cleaning method however declined during the subsequent operation cycles where the flux recovery rates were 61.8% at cycle 8 of regime 1 and 66.2% at cycle 6 of regime 2. The higher efficiencies of physical cleaning during the initial cycles could be attributed to the ease for cleaning of OSHF TFC FO membrane module and less severity of membrane fouling at initial cycles in both operation regimes.

When the membrane was signficantly fouled, a different membrane fouling mitigation strategy was adopted instead of physical cleaning. This included using osmotic back washing of the membrane using NaCl solution on the feed side and DI water on the DS side of the FO membrane. As can be seen from the Fig. **3**, water flux recoveries were 70.2% and 74.7% for regime 1 (Fig. 3(a)) and regime 2 (Fig. 3(b)), respectively which is only a marginal improvement over the physical cleaning approach. This relatively lower cleaning efficiency could be attributed to the complex cake layer formed on the membrane by different activated sludge components including microorganisms, extracellular polymeric substances, soluble macro/micro organics, and inorganics (Zhang et al., 2012b). Moreover, backwashing solution had a concentration of only 30 g/L of NaCl which might now have been enough to generate adequate water flux to effectively clean and restore the membrane flux. Internal pore blocking of the membrane are likely responsible for the poor flux recovery by osmotic backwashing technique. This lower backwashing efficiency for water flux recovery results obtained in this study are in a good agreement with some other reported studies (She et al., 2016; Valladares Linares et al., 2013a; Valladares Linares et al., 2013b).

Two-step alkaline-acidic cleaning is well-known as effective fouling mitigation strategy for fouled membranes (Wang et al., 2014c; Wang et al., 2015). This cleaning strategy, therefore was utilized for cleaning the fouled OSHF TFC FO membranes. Fig. **3** shows the cleaning efficiency using two-step chemical cleaning for activated sludge-fouled OSHF TFC FO membrane over two operation regimes (cycle 11 for regime 1 and cycle 8 for regime 2). These fouling mitigation methods exhibited excellent ability with over 90% of water flux recovery after cleaning process for both operating regimes. This result shows that alkaline-acidic sequential cleaning strategy is a very effective approach for mitigating membrane fouling with the OSHF TFC FO membranes during long-term operation (Wang et al., 2015). SDS is a surfactant which has unique structures including both hydrophobic tails (dodecyl) and hydrophilic heads (sulfonic groups). This characteristic enables it to solubilize macromolecules by forming micelles with proteins, oil and fat in water (Wang et al., 2014c; Wang et al., 2015). SDS not only can break up the bindings of foulants and metal ions through hydrophilic interaction between water molecules and hydrophilic heads but also can absorb organic foulants by hydrophobic tails. Sodium hydroxide (NaOH) in SDS solution can break down big foulants into soluble organic substances and/or fine particles. NaOH also provides alkaline condition which helps to solubilize and hydrolyze those fine particles and soluble organic substance into smaller molecules (Wang et al., 2015). Ang et al. (2011) reported that foulants become negatively charged due to the deprotonation of their functional groups at a higher pH value (e.g., pH 11). This can facilitate the efficiency of cleaning process due to the repulsive interaction among negatively charged foulants. (Ang et al., 2011). This explains why the cleaning efficiency using mixed reagent of NaOH and SDS was significantly enhanced.

The high efficiencies of membrane fouling mitigation strategies shows that using OSHF TFC FO membrane, fouling can be reasonably controlled with physical cleaning methods during initial cleaning cycles and chemical cleaning was excellent when the membrane was completely fouled. However, osmotic backwashing alone exhibited less effectiveness compared to alkaline and acid cleaning. This is understandable since chemical cleaning has been proved to be a highly effective method in removing fouling cake layer when activated sludge was used as feed solution (Wang et al., 2014c; Wang et al., 2015).

3.4 *Removal of contaminants*

Performance of OMBR system was also assessed in terms of removal of primary contaminants including TOC and NH_4^+-N. Samples were collected during the experiment operation time and after osmotic backwashing and chemical cleaning in both two operation regimes. This helped to investigate the contaminant removals of OMBR system and OSHF TFC FO membrane during operation time and after each fouling mitigation method. TOC and NH_4^+-N removals are presented in **Fig. 4.** Generally, OMBR system exhibited consistently excellent TOC and NH_4^+-N removals with over 98% over 6 days and 8 days of operation regime 1 and regime 2, respectively under all the different fouling mitigation/cleaning strategies. Other recent studies also reported high overall TOC removals. Wang et al. (2016b) reported 96% of total TOC removal by the OMBR system (Wang et al., 2016b). Achilli et al. (2009) also reported a 98% rejection of FO membrane in their work with overall TOC removal of OMBR system was 99% (Achilli et al., 2009). Qiu and Ting (2013) noted that a stable overall TOC removal of up to 98% in their study. High removal of TOC was also achieved by Zhu et al. (2018) with 98 to 100% (Zhu et al., 2018). Similar to TOC removal, NH_4^+-N removals during experiment operation time in both regimes were stable at a rate of more than 98% which was much higher removal rate compared to the overall removal rate of 80 – 90% in a study conducted by Qiu and Ting (2014). This consistently high removal of NH_4^+-N removal was due to the combined effect of biological nitrification and excellent rejection property of FO membrane. The high removals of NH_4^+-N in the OMBR system obtained in this study were in good agreement with previous OMBR studies (Achilli et al., 2009; Luo et al., 2017; Pathak et al., 2018; Qiu and Ting, 2013; Wang et al., 2014b; Wang et al., 2017; Wang et al., 2016b).

[Insert Figure 4]

3.5 *Membrane autopsy using SEM and EDX analysis*

[Insert Table 2]

Surface morphologies of the pristine membrane and fouled OSHF TFC FO membrane after OMBR operation were investigated. The SEM image shows that the pristine membrane has a typical surface morphology of the selective polyamide layer of the TFC membrane while the surface SEM images of fouled membranes shows a cake layer formed on the active layer surface of the membrane in both operation regimes. In addition, the cross-sectional images of the fouled membranes also clearly show that the fouling cake layer on the membrane surface of regime 1 (12-hour cycle) was thicker than that of regime 2 (24-hour cycle) with 23 µm and 20 µm, respectively. This could be attributed to the slightly higher water flux for regime 1 compared to regime 2 because of which a higher external concentration polarization of foulants and subsequently thicker foulant cake layer formation is expected on the membrane surface. This was also the likely reasons for attributing faster water flux decline in regime 1 compared to that of regime 2 as discussed earlier under Section 3.2.

Cake layers were analyzed using EDX for determination of elemental compositions which are tabulated in Table **2**. Compared to pristine membrane containing carbon, oxygen and sulfur, fouling layers of the two fouled membranes comprise of some other elements such as sodium, magnesium, silica, phosphorous, sulfur, chloride, potassium, calcium and iron, which are typical chemical constituents found in the mixed liquor (Bell et al., 2016; Sun et al., 2016). Generally, the composition of these elements was comparatively similar in the fouling layers for both operation regimes. This result indicates that biofouling and inorganic scaling simultaneously occurs on the surface of membrane due to the presence of above elements in the fouling layer. This is consistent with the results obtained in other studies with OMBRs (Wang et al., 2017; Zhu et al., 2018). The active layer of OSHF TFC membrane can be prone to easy surface damages such as due to sharp and pointed particulates present in the mixed liquor. However, a consistently low rate of salinity build-up in the OMBR system indicates that no membrane damages had occurred during the operation of the OMBR system under two regimes showing the robustness of the OSHF membrane.

4 Conclusions

Performance of OMBR system for treating municipal wastewater using novel OSHF TFC FO membrane was investigated based on water flux; mixed liquor's salinity build-up, contaminants removal; and fouling cleaning efficiency. The newly developed membrane possesses excellent intrinsic properties which was proved by high performance in OMBR system. High cleaning efficiencies were attained using this membrane with different fouling mitigation methods. OMBR system achieved high removals of TOC and NH_4^+-N (98%). This study shows that the OSHF membrane has a great potential for OMBR system towards sustainable FO – RO hybrid system and fertilizer drawn forward osmosis system for wastewater reclamation.

Supplementary data of this work can be found in online version of the paper

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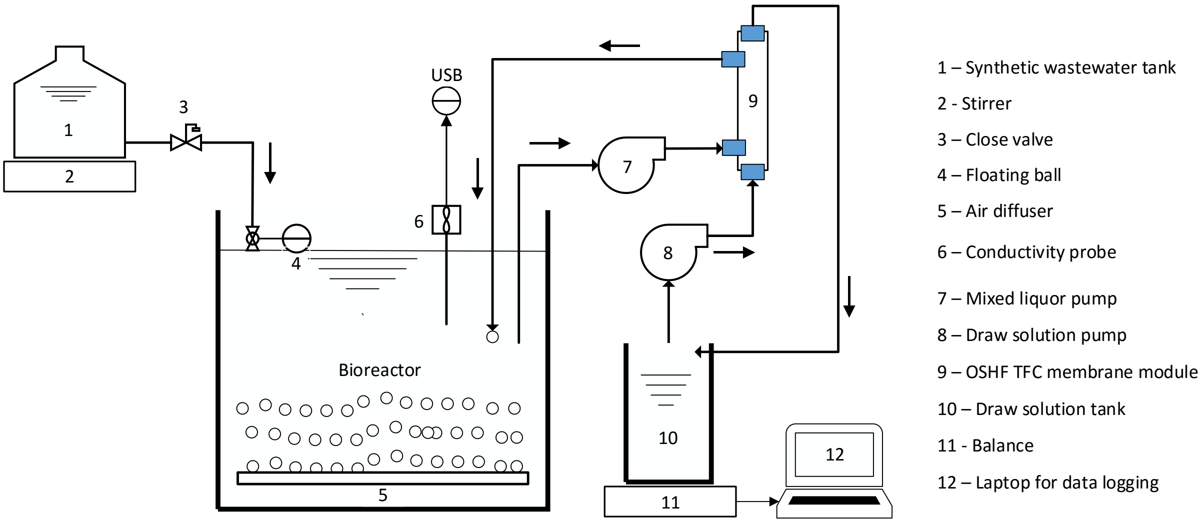
Figure 1. Diagram of the lab-scale osmotic membrane bioreactor system.

Figure 2. Water flux profile (**a**) and Mixed liquor total dissolved solid (TDS) (**b**) during two operation regimes under three different fouling control strategies. Two pristine membrane modules OSHF-1 and OSHF-2 were used in regime 1 and regime 2, respectively.

Figure 3. Initial water flux and flux recovery after three cleaning strategies over (a) operation regime 1 and (b) operation regime 2 (initial water flux shown here is the averaged value of water flux in the first hour of every cycle).

Figure 4. Removal of pollutants in the OMBR system during two operation regimes. (a) TOC removal; (b) NH_4^+-N removal in regime 1; (c) TOC removal; (d) NH_4^+-N removal in regime 2.



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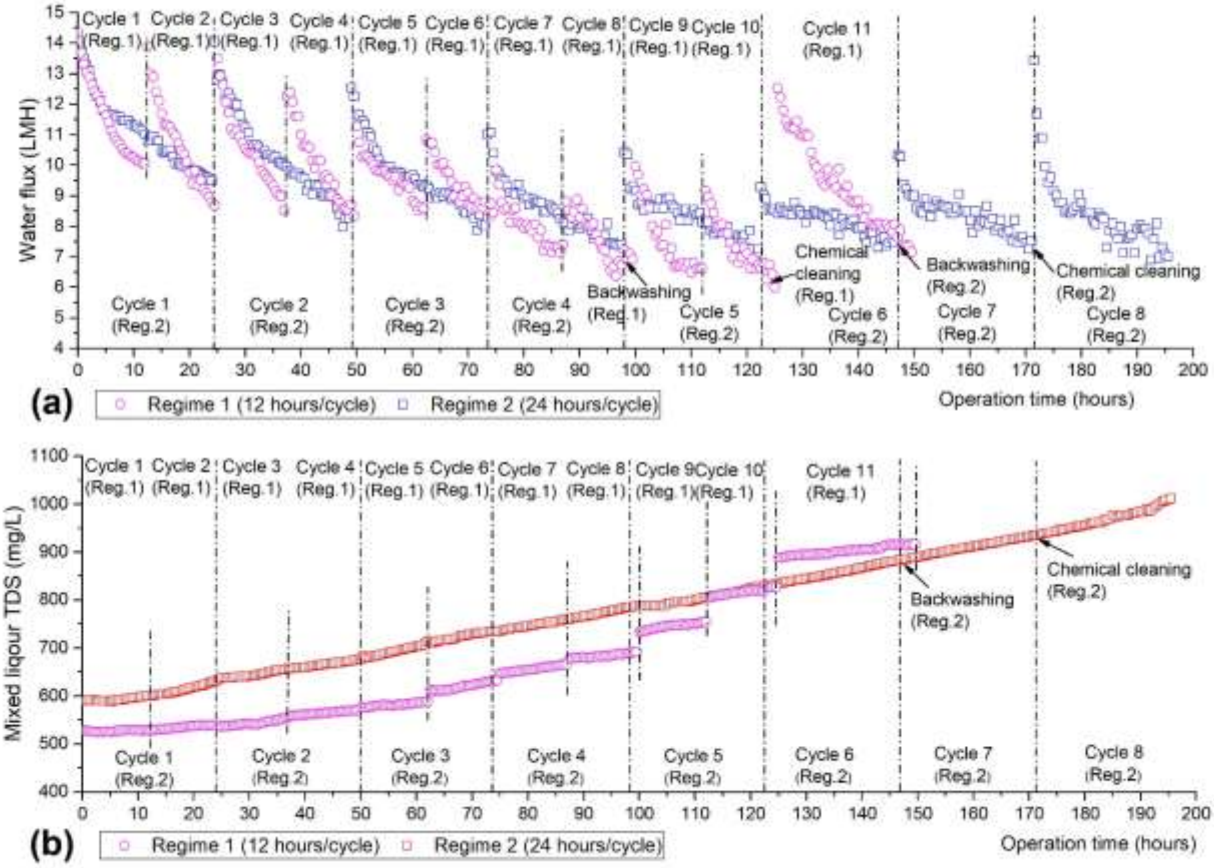

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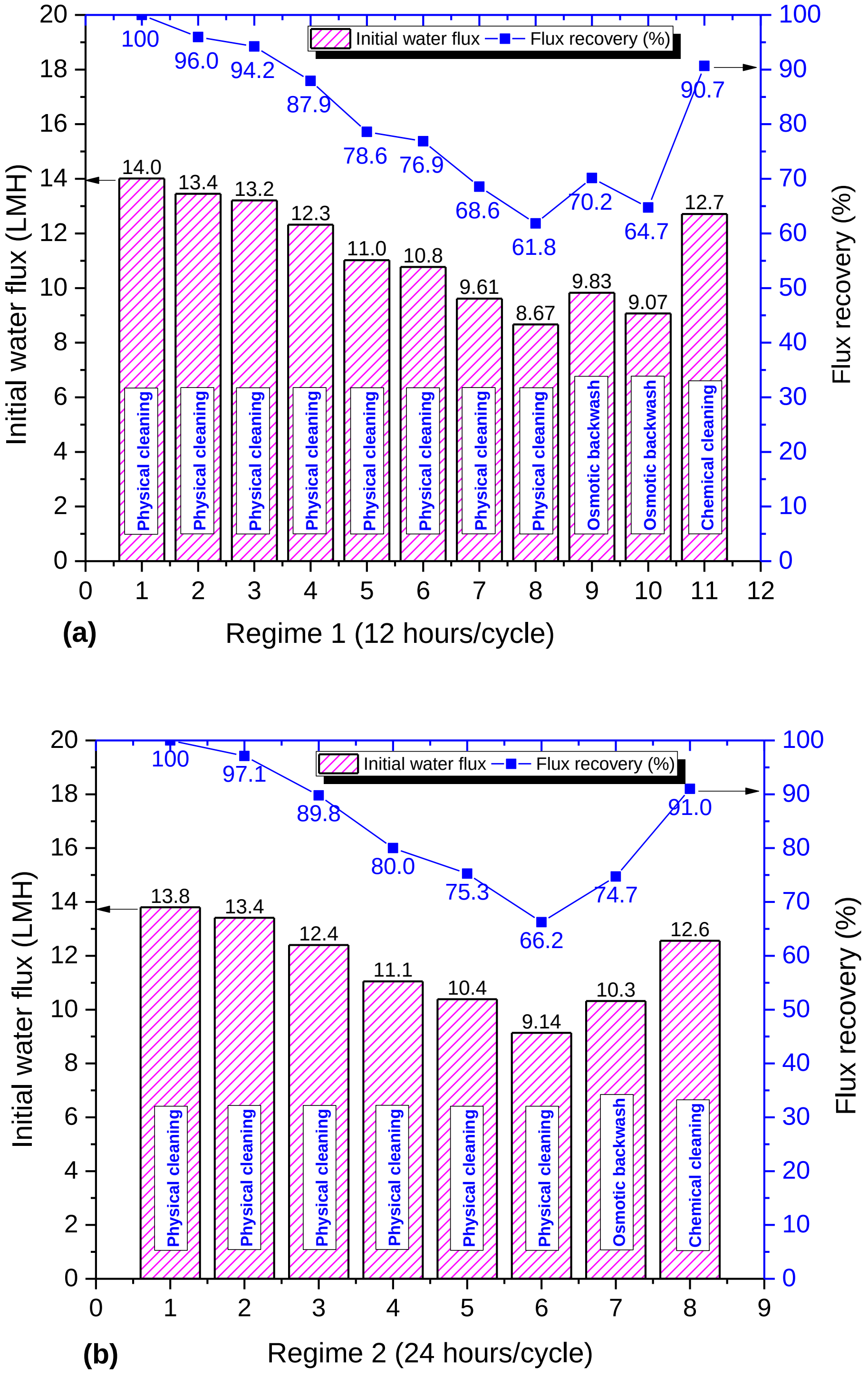

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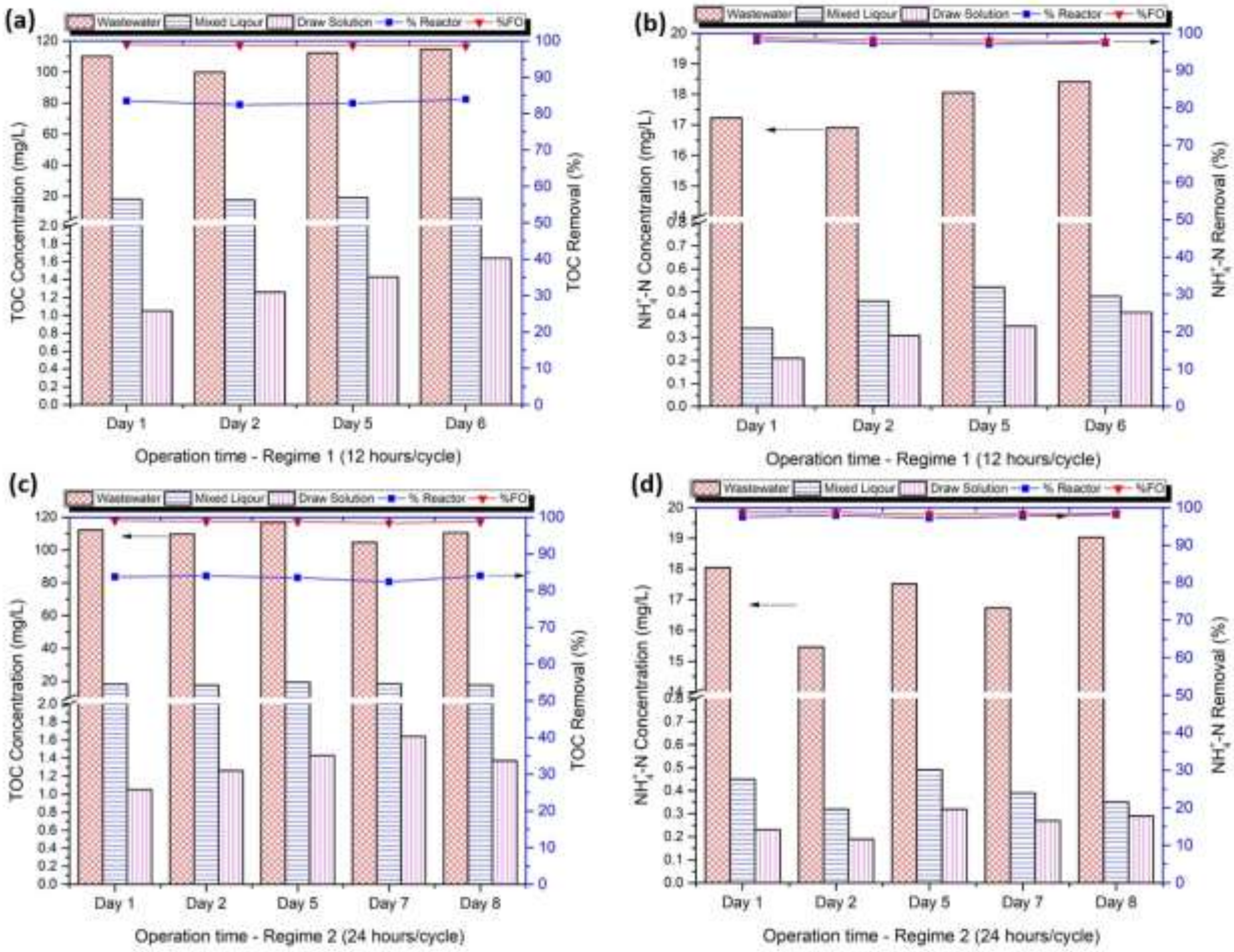

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Table 1 Intrinsic characteristics of two OSHF TFC FO membrane modules used in this study.

OSHF TFC FO membranes	A ($Lm^{-2}h^{-1}bar^{-1}$)	B ($Lm^{-2}h^{-1}$)	B/A (bar)	S (μm)
OSHF-1	2.26	0.28	0.12	190
OSHF-2	2.24	0.30	0.13	195

Table 2 The elemental compositions of pristine and fouled membranes in two operation regimes by energy-dispersive X-ray (EDX) analysis.

Weight %	C	O	Na	Mg	Al	Si	P	S	Cl	K	Ca	Fe
Pristine	69.29	21.06						9.65				
Regime 1	59.58	34.71	0.32	0.3	0.41	0.18	2.02	0.89	0.2	0.15	0.78	0.44
Regime 2	56.58	37.06	0.36	0.29	0.39	0.35	2.27	0.86	0.26	0.18	0.84	0.58