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Integration of micro-filtration into osmotic membrane bioreactors to prevent salinity build-up

Xinhua Wang, Bo Yuan, Yao Chen, Xiufen Li *, Yueping Ren

Laboratory of Environmental Biotechnology, School of Environmental and Civil Engineering, Jiangnan University, Wuxi 214122, PR China

HIGHLIGHTS

- MF effectively alleviated the salinity build-up in the OMBR by solute discharge.
- MF membrane was helpful for increasing the water flux of FO membrane in the OMBR.
- MF increased the removals of activated sludge for TOC and NH_3–N in the OMBR.
- Reversible fouling of FO membrane in the MF-OMBR was severer than that in the OMBR.
- FO flux decline was severer than the expectation mainly due to the membrane fouling.

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ABSTRACT

The high salinity remains as one of major obstacles of the osmotic membrane bioreactor (OMBR). In this study, a new pathway was explored to prevent the salinity build-up by integrating the micro-filtration (MF) membrane to the OMBR (MF-OMBR). The results indicated that the salinity characterized by conductivity in the MF-OMBR was effectively alleviated and controlled at a lower value of about 5 mS/cm, and the stable flux of forward osmosis (FO) membrane correspondingly increased to approximately 5.5 L/(m^2 h). Besides, the addition of MF membrane in the OMBR could increase the total organic carbon (TOC) and ammonium nitrogen (NH_3–N) removals due to the activated sludge by improving the microbial activity. The membrane fouling especially the reversible fouling in the MF-OMBR was severer compared to that in the conventional OMBR, which resulted in a lower water flux than the expectation due to the increase of filtration resistance and external concentration polarization.

1. Introduction

Recently, the osmotic membrane bioreactor (OMBR) combining forward osmosis (FO) with activated sludge process has attracted growing interests in the field of wastewater treatment and reclamation (Cornelissen et al., 2008, 2011; Achilli et al., 2009; Yap et al., 2012). In the OMBR, the wastewater flows across a selectively permeable membrane from the activated sludge to the draw solution by an osmotic driving force. Compared to the conventional membrane bioreactor (MBR), the OMBR has a lower fouling propensity due to using the osmotic pressure instead of hydraulic pressure as the driving force (Cornelissen et al., 2008, 2011; Achilli et al., 2009; Yap et al., 2012). Besides that, higher quality pure water is obtained in the OMBR because a variety of contaminants can be effectively rejected by the FO process (Cornelissen et al., 2008, 2011; Achilli et al., 2009; Yap et al., 2012).

Although the OMBR has many advantages over the conventional MBR, there are still some drawbacks such as internal concentration polarization (ICP) and reverse salt transport. ICP induced by the accumulation of feed solutes or the dilution of draw solution in porous support layer has been recognized as a major obstacle of FO membrane because severe ICP eventually results in the water flux decline (Cath et al., 2006; McCutcheon and Elimelech, 2006; Tang et al., 2010). In addition, the reverse salt transport, i.e., the transport of salt from draw solution to the bioreactor passing through the FO membrane, is expected to occur due to the difference in solute concentration between the draw solution and the bioreactor solution (Hancock and Cath, 2009; Alturki et al., 2012). The high retention property of the FO membrane inevitably leads to the build-up of feed solutes and the salt due to reverse transport in the OMBR. Subsequently, the salt accumulation results in the elevated salinity condition in the OMBR, which

* Corresponding author. Tel.: +86 510 85326516.
E-mail addresses: xhwang@jiangnan.edu.cn (X. Wang), xfli@jiangnan.edu.cn (X. Li).

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not only causes a reduction of driving force but also has adverse impacts on the microbial activity and population structure in the bioreactor (Lay et al., 2011; Yap et al., 2012).

In order to mitigate the salt accumulation in the OMBR, many efforts have been put into the selection of draw solutes and development of an ideal FO membrane (Yap et al., 2012). Besides that, sludge retention time (SRT) has been recognized as a feasible method to alleviate the salt accumulation in the OMBR through the daily sludge discharge. Some researchers have stated that the salinity in the OMBR would reach a constant value depending on SRT (Achilli et al., 2009). Additionally, a theoretical model on the salt accumulation behavior was further developed, which indicates the critical importance of hydraulic retention time (HRT) and SRT for optimizing the OMBR operation (Xiao et al., 2011). However, the mitigation of salt accumulation in the OMBR at a lower SRT is limited. Wang et al. (2014) found that even if the OMBR was operated at SRT of 10 d, the stable salinity was still high and had adverse impacts on the performance of OMBR. Thus, it is necessary to search for other methods to further control the high salinity induced by the salt accumulation in the OMBR.

Based on the fact that the soluble salt in the bioreactor could pass through the micro-filtration (MF) membrane with the pore size in the range of 0.1-1 μm, the MF process might be a feasible method to prevent the salinity build-up in the OMBR by discharging the soluble salt through the MF membrane. To the best knowledge of the authors, this is the first study actively applying the MF membrane to alleviating the salt accumulation in the OMBR. Thus, the objectives of this study are to evaluate the feasibility of applying the MF membrane for reducing the salt accumulation in the OMBR and to investigate the corresponding impacts of the addition of MF membrane on the performance of OMBR such as total organic carbon (TOC) and ammonium nitrogen (NH_3–N) removals, water flux and membrane fouling of FO membrane, microbial activity and extracellular polymeric substances (EPS).

2. Methods

2.1. Experimental set-up

As shown in Fig. 1, a bench-scale submerged OMBR with a MF membrane for solute discharge (called as MF-OMBR) was used in this study. The MF-OMBR had an effective volume of 7.56 L, and two membrane modules including a MF module and a FO module were immersed in the bioreactor with the activated sludge. The applied flat-sheet FO membrane with an embedded polyester screen mesh was provided by Hydration Technology Inc. (HTI, Albany, OR), which has been widely used in many studies due to its relatively smooth and hydrophilic properties (Tang et al., 2010; Yap et al., 2012). It was made of cellulose triacetate (CTA) with an effective membrane area of 0.056 m^2. According to the method reported by Cath et al. (2013), its water permeability coefficients (A) and salt permeability coefficients (B) were determined, and their values were approximately 0.8 L/(m^2 h bar) and 2×10^{-7} m/s, respectively. Considering the fact that the membrane orientation plays a significant role in FO applications, the membrane orientation of the active layer facing feed solution was adopted in order to avoid the aggravated fouling especially pore-clogging in the support layer (Lay et al., 2011). With regard to the MF membrane (supplied by Zizheng Environment Inc., China) applied in the MF-OMBR, it was made of polyvinylidene fluoride (PVDF) with a mean pore size of 0.20 μm and an effective membrane area of 0.059 m^2. The extensively applied PVDF membrane in the MBRs was used in this study due to its high mechanical strength, thermal stability and chemical resistance (Liu et al., 2011). The municipal wastewater was continuously pumped into the MF-OMBR, and the aeration was provided through an axial perforated tube below the two membrane modules. The influent pump was controlled by a water level sensor to maintain a constant water level. In the MF-OMBR, analytical grade sodium chloride (NaCl) with the concentration of 1 M was used as the draw solution, which was circulated at the flow rate of 0.5 L/min from a 2 L glass reservoir through the FO membrane module and back to the reservoir. A conductivity control system including a conductivity probe, a control device and a concentrated salt adding device with 5 M NaCl was used to control the draw solution at constant concentration of 1 M. As for the MF membrane module, its effluent flow rate was controlled by a peristaltic pump, and its trans-membrane pressure (TMP) was monitored by a pressure sensor.

2.2. Operating conditions

The synthetic municipal wastewater with the concentrations of chemical oxygen demand (COD), TOC, total suspended solids (TSS), NH_3–N, total nitrogen (TN) and total phosphorus (TP) of 373.3 ± 18.5, 151.1 ± 7.8, 40 ± 3.8, 33.1 ± 1.5, 46.8 ± 3.8 and 3.18 ± 0.25 mg/L, respectively, was used as the influent water of

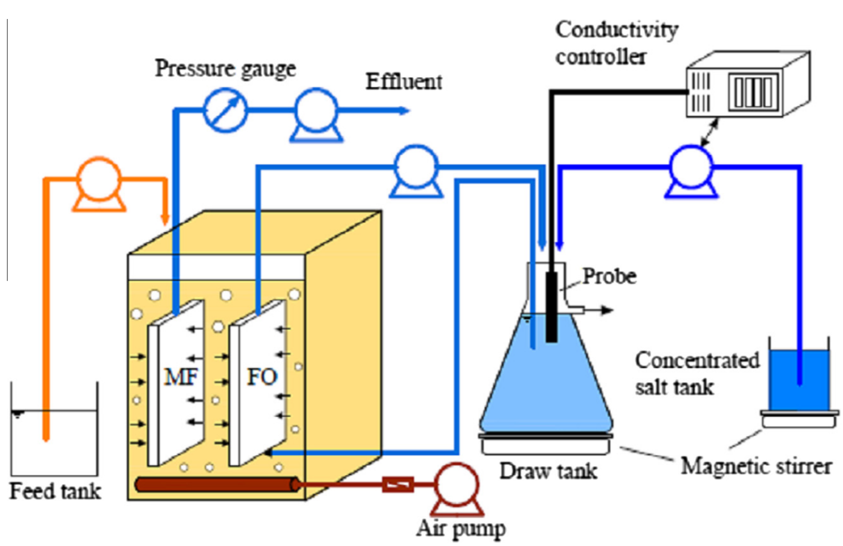

Fig. 1. Flow diagram of the MF-OMBR set-up.

the MF-OMBR. Activated sludge applied in the MF-OMBR was obtained from a bench-scale submerged membrane bioreactor (SMBR) continuously treating the same synthetic wastewater for about 3 months. The information on the SMBR and the composition of synthetic wastewater could be found in previous studies (Chen et al., 2011; Wang et al., 2012). The MF-OMBR was put in a temperature-controlled room to maintain the temperature in the range of 23 ± 0.5 °C. Continuous aeration with the intensity of 0.5 m^3/h was used in order to induce a cross-flow velocity for membrane fouling reduction and supply oxygen for microorganisms. The gas sparging velocity on the membrane surface and the dissolved oxygen (DO) were approximately 0.15 m/s and 3.5 mg/L, respectively. During the entire operation of the MF-OMBR, the SRT was fixed at 10 d by discharging a certain volume of excess sludge from the bioreactor once a day. It should be noted that the hydraulic retention time (HRT) of the MF-OMBR varied in a range of 8.5–13.5 h during the entire operation due to the flux decline of FO membrane. The operating condition of FO membranes was given in Section 2.1, and the MF membrane was continuously operated under constant flux mode with the flux of 5 L/(m^2 h) (LMH).

2.3. Analytical methods

Water flux through the FO membrane was calculated based on the weight change of the draw solution, and the conductivity of the mixed liquor (C_{ml}) was monitored and recorded by a conductivity device (OKD-650, Shenzhen OK Instrument Technology Co., Ltd., China) in order to characterize the salinity. NH_3–N and TOC in the influent, sludge supernatant and membrane effluents of FO and MF were conducted according to the Chinese NEPA standard methods (2002) and by a TOC analyzer (TOC-Vcsh, Shimadzu, Japan), respectively. The total dissolved solids (TDS) of the MF effluent were determined according to the Chinese NEPA standard methods (2002).

The specific extraction of EPS including soluble microbial products (SMP) and bound extracellular polymeric substances (BEPS) from activated sludge could be found in previous literatures (Chen et al., 2011; Wang et al., 2012). Both the SMP and BEPS extractions were normalized as the sum of protein and polysaccharide. Polysaccharide was measured by the phenol sulfuric acid method with glucose as a standard (Dubois et al., 1956), while protein was determined by a modified Lowry method using Bovine serum albumin (BSA, Sigma fraction V, 96%) as protein standard (Lee et al., 2001). Dehydrogenase activity (DHA) was analyzed by the TTC-dehydrogenase activity determination methods described by Zhang et al. (2011). All the above analyses were conducted at least 3 times, and their mean values ± standard deviations were reported.

The fouled FO membrane module was taken out at the end of the operation, and then the FO membrane samples were obtained by cutting from the middle of the fouled FO membrane. A field-emission scanning electron microscopy (FESEM) (S-4800, Hitachi, Japan) was used to capture the surface images of the fresh and fouled FO membrane samples. Prior to SEM observation, membrane samples were prepared by freezing the membrane at −80 °C in a chiller for 2 h followed by freeze drying at −48 °C for 6 h using a freeze dryer (FreeZone 25, Labconco, Czech Republic), and coated with a uniform layer of gold. The inorganic composition of the fouling layer on the FO membrane surface was analyzed by an energy diffusive X-ray (EDX) analyzer (Falcon, EDAX Inc., America), while the major functional groups of the organic foulants on the FO membrane surface were characterized by a Fourier transform infrared (FT-IR) spectrophotometer (Nicolet iS10, Thermo Fisher Scientific, America).

2.4. Water flux test

In order to evaluate the contribution of membrane fouling to the flux decline of the FO membrane, the water flux of FO membrane before and after the operation of the MF-OMBR was measured by a bench-scale filtration cell (Supplementary Fig. S1). The effective FO membrane area needed in this filtration cell was 27 cm^2. Two peristaltic pumps were used to circulate the feed solution (deionized water) and the draw solution (NaCl) at a flow rate of 0.5 L/min, respectively. The concentration of draw solution was controlled at a constant concentration of 1 M by the same conductivity control system as the MF-OMBR mentioned above. The filtration cell was put into a temperature-controlled room to maintain the temperature in the range of 23 ± 0.5 °C, and the duration of the test was approximately 8 h in order to obtain the constant flux.

The virgin FO membrane with effective area of 27 cm^2 was firstly placed in the filtration cell to obtain the initial water flux (F_i). After the continuous operation, the fouled FO membrane was cut into the size of 4.5 cm × 6 cm and then was used to quantify the final water flux (F_f) by the filtration cell. Moreover, the water flux (F_w) of the fouled FO membrane after washed by the deionized water was also tested. The difference between the F_i and F_f could be used to characterize the flux decline due to the membrane fouling only, while the flux decline calculated by the difference between F_i and F_w could be attributed to the irreversible membrane fouling. Based on the water flux tests, the flux decline due to different reasons could be calculated using the following equations:

$$L_t = (F_0 - F)/F_0 \times 100\% \quad (1)$$

$$L_f = (F_i - F_f)/F_i \times 100\% \quad (2)$$

$$L_{in} = (F_i - F_w)/F_i \times 100\% \quad (3)$$

where L_t is the total flux decline in the OMBR (%), L_f is the flux decline due to membrane fouling (%), L_{in} is the flux decline due to irreversible membrane fouling (%), F is the flux in the end of the MF-OMBR (LMH), F_0 is the flux in the initial operation of the MF-OMBR (LMH), F_i is the flux of the fresh FO membrane in the filtration cell (LMH), F_f is the flux of the fouled FO membrane in the filtration cell (LMH) and F_w is the flux of the fouled FO membrane after washing by the deionized water in the filtration cell (LMH).

3. Results and discussion

3.1. Salt accumulation and water production

Results for the conductivity of the mixed liquor (C_{ml}) and water production (J_w) in the MF-OMBR are presented in Fig. 2. From Fig. 2, it could be observed that the water flux of FO membrane had a reduction in the initial stage of the MF-OMBR and then was stable at about 5.5 LMH after 25 d. The water flux decline of FO membranes in the MF-OMBR was in accordance with the results reported in previous studies on the OMBR (Lay et al., 2011; Yap et al., 2012). Wang et al. (2014) has reported that the stable water flux of FO membrane was about 2 LMH in a submerged OMBR, which had the same FO membrane and operating conditions including the synthetic municipal wastewater and SRT as the MF-OMBR in this study. Thus, the stable water flux in the MF-OMBR was 2.75 times more than that in the above publication, indicating that it was feasible to mitigate the flux decline of FO membrane in the OMBR by adding the MF membrane. Furthermore, it should be noted that the flux of the MF membrane was stable at 5 LMH without any physical and chemical cleaning during the whole operation of MF-OMBR.

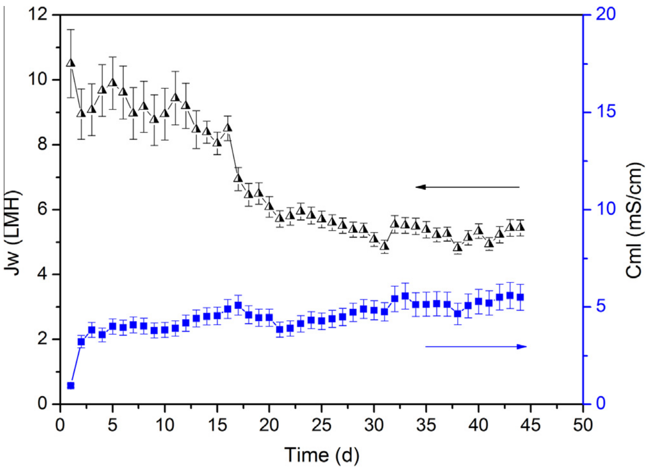

Fig. 2. Variations of water flux (J_w) and conductivity of the mixed liquor (C_{ml}) in the MF-OMBR.

The flux decline of FO membrane could be attributed to several reasons such as ICP, salt accumulation and membrane fouling (McCutcheon and Elimelech, 2006; Achilli et al., 2009). Thus, salinity in the OMBR was a significant factor affecting the flux decline. As shown in Fig. 2, the salinity of the mixed liquor characterized by conductivity had a slight increase and was stable at about 5 mS/cm in the MF-OMBR, which was 90% lower in comparison with the stable salinity of mixed liquors in a submerged OMBR treating synthetic municipal wastewater (Wang et al., 2014). Furthermore, the variation of TDS in MF effluent was also determined (Supplementary Fig. S2). It could be seen that the variation of the soluble salinity was similar as the change of C_{ml}, indicating that the application of MF membrane to discharging the soluble salt could effectively decrease the salinity and further alleviate the salt accumulation in the OMBR. Combining the variations of water flux shown in Fig. 2, it could be concluded that the higher stable flux in the MF-OMBR was mainly attributed to the lower salinity. Moreover, it should be pointed out that the water flux of FO membrane in the MF-OMBR was also much lower than the theoretical value calculated by the salinity difference between the draw solution and the mixed liquor, suggesting that ICP and membrane fouling also played significant roles in the flux decline of the MF-OMBR.

3.2. Removals of TOC and NH_3–N

Compared with the conventional OMBR, the mode of effluent is different in the MF-OMBR, which has the effluent from MF membrane except for the effluent from FO membrane. Thus, the effluent quality of the MF-OMBR should be represented by the mixed stream of MF and FO effluents. The concentrations of TOC and NH_3–N in the mixed effluent of the MF-OMBR could be obtained according to the mass balance of TOC and NH_3–N in FO and MF effluents:

$$C_{mix(t)} = \frac{C_{MF(t)}V_{MF(t)} + C_{FO(t)}V_{FO(t)}}{V_{MF(t)} + V_{FO(t)}} \quad (4)$$

where $C_{mix(t)}$ is the TOC or NH_3–N concentration in the mixed effluent at time t, $C_{MF(t)}$ is the TOC or NH_3–N concentration in the MF membrane effluent at time t, $C_{FO(t)}$ is the TOC or NH_3–N concentration in the FO membrane effluent at time t, $V_{MF(t)}$ is the volume of the MF membrane effluent at time t and $V_{FO(t)}$ is the volume of the FO membrane effluent at time t. Based on the real operation of the MF-OMBR, the $V_{MF(t)}$ could be obtained by the water flux and the effective membrane area because the flux of MF was constant at 5 LMH during the entire operation, while $V_{FO(t)}$ was calculated based on the weight change of the draw solution due to the flux decline of FO membrane.

The concentrations of TOC and NH_3–N in the influent, sludge supernatant, MF effluent, FO effluent and mixed effluent are listed in Fig. 3. It could be observed from Fig. 3(a) that the TOC removal efficiency due to the activated sludge in the MF-OMBR, i.e. the difference between the influent and the supernatant, was in the range of 80.2–90.2%, while the TOC removal efficiency of the FO membrane was more than 99% in the entire operation of the MF-OMBR due to the high retention of FO membrane. High TOC removal of FO membrane was consistent with the results in previous literatures (Lay et al., 2011; Wang et al., 2014). However, the TOC removal efficiency of the MF membrane was much lower than that of the FO membrane. Apparently, it could be seen from Fig. 3(a) that the TOC concentration in the MF effluent was slight lower than that in the supernatant but much higher than that in the FO effluent, indicating that the MF membrane could retain only a small part of TOC in the supernatant. Due to the lower retention of MF membrane for the TOC in the supernatant, the TOC removal efficiency between the mixed effluent and the influent was in the range of 90.4–96.8%, which was lower than that only due to the FO membrane. With respect to NH_3–N removal in the MF-OMBR, it could be observed from Fig. 3(b) that there was no significant difference in the NH_3–N concentration between the supernatant and the effluents of MF and FO membranes, indicating that the NH_3–N

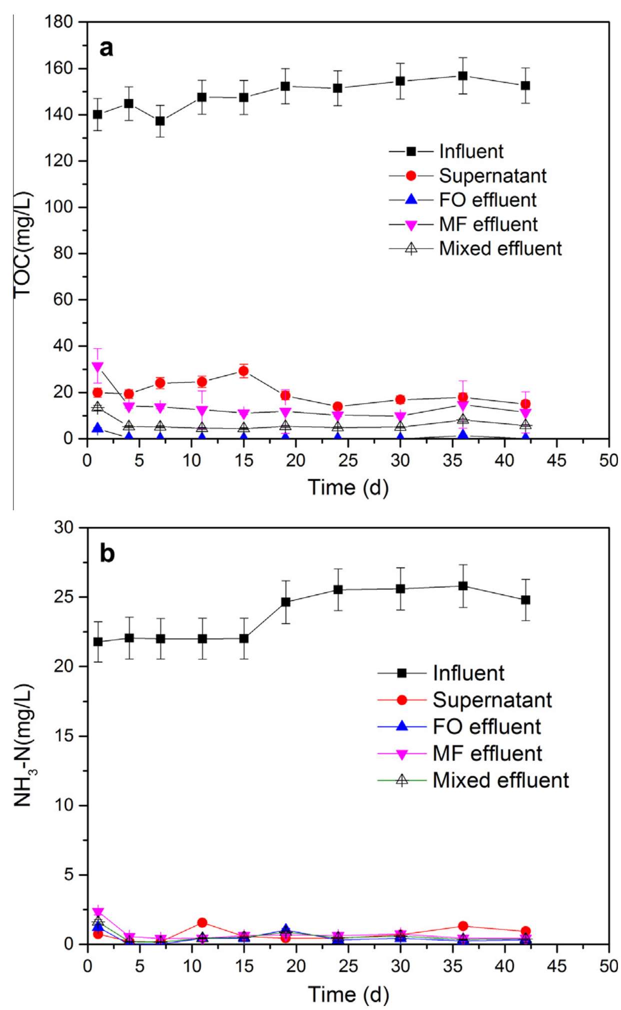

Fig. 3. Variations of TOC (a) and NH_3–N (b) concentrations in the influent, sludge supernatant, FO effluent, MF effluent and mixed effluent.

removal efficiency of activated sludge was very high. Furthermore, similar NH_3–N concentration in FO and MF effluents suggested that the retention of FO membrane for NH_3–N in the supernatant was not better than that of MF membrane in this study. In fact, it has been reported that the retention of FO membrane for NH_3–N was limited (Yap et al., 2012; Wang et al., 2014). Due to the effective removal of activated sludge for NH_3–N, the removal efficiency of the mixed effluent for NH_3–N was more than 99% in the MF-OMBR. Thus, the addition of MF membrane did not deteriorate the NH_3–N concentration in the mixed effluent.

Compared to the TOC and NH_3–N concentrations of the supernatant in the conventional OMBR with the same operating conditions and influent wastewater (Wang et al., 2014), they were much lower in the MF-OMBR. It might be attributed to the higher microbial activity in the MF-OMBR due to its lower salinity based on the fact that the high salinity could result in the loss of metabolic activity, the release of soluble microbial products and the reduction of the capacity to sustain shock loads (Rene et al., 2008; Yogalakshmi and Joseph, 2010). In order to demonstrate this hypothesis, the DHA that has been widely applied for determining the biological activity was determined to characterize the microbial activities of bulk sludge in the MF-OMBR (Molina-Munoz et al., 2010). The variations of DHA with the operation time in the MF-OMBR are shown in Fig. 4. From Fig. 4, it could be observed that the DHA decreased with the extension of the operation time and was finally stable at about 33.24 mg TF/(L h), which was much bigger than that in the conventional OMBR of approximately 6.88 mg TF/(L h) (Wang et al., 2014). The above results strongly supported the hypothesis that higher salinity affected the TOC and NH_3–N removal of activated sludge in the conventional OMBR. Thus, it could be concluded that the application of MF membrane in the OMBR could not only increase the water flux but also enhance the TOC and NH_3–N removals of activated sludge due to the increase of microbial activity.

3.3. EPS

EPS consisting of SMP and BEPS based on the distribution feature of EPS on cell has been recognized as an important index with significant impacts on the sludge properties and membrane fouling (Wang et al., 2009). Variations of SMP and BEPS in the MF-OMBR are presented in Fig. 5. As shown in Fig. 5, both SMP and BEPS increased with the operation of the MF-OMBR. It is well known that the high salinity in the conventional OMBR could cause the release of EPS due to the increase of osmotic pressure during

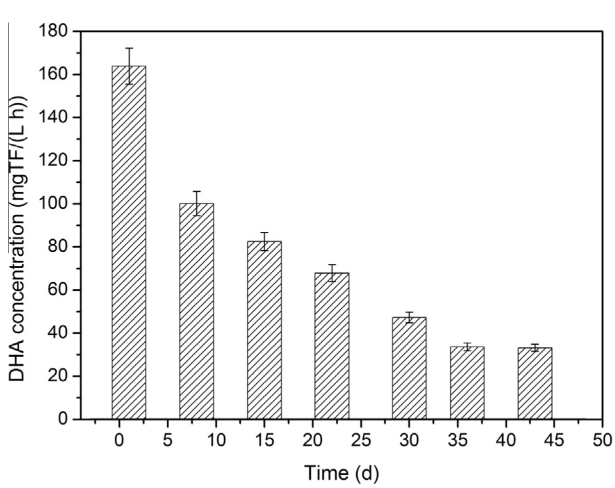

Fig. 4. Variations of DHA in the MF-OMBR.

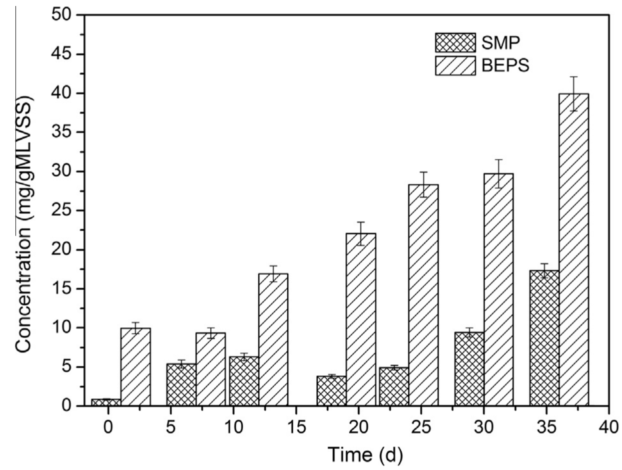

Fig. 5. Variations of SMP and BEPS in the MF-OMBR.

metabolism, enhancement of cell lysis or stimulation of efflux mechanism (Chen et al., 2014). However, the salinity in the MF-OMBR was effectively controlled at a low value (as shown in Fig. 2). Thus, the salinity was not the reason for the increase of EPS in the MF-OMBR. In this study, the MLSS decreased from about 6.86 g/L to the stable value of about 1.10 g/L with the operation of the MF-OMBR due to the applied lower SRT of 10 d, which resulted in the increase of the sludge loading. In this case, the microorganisms would utilize the excess substrates to generate EPS (Chen et al., 2014). Some generated EPS could be degraded, while others that were difficult to degrade could be retained by the MF and FO membranes, thus leading to the increase of the EPS concentration.

The production of EPS in the MF-OMBR system was comparable to the values associated with the conventional OMBR. Wang et al. (2014) investigated the EPS production in a conventional OMBR with the same operating conditions and influent wastewater as this study, and reported that the stable concentrations of BEPS and SMP were about 45 and 22 mg/g VSS, respectively, which were more than that in the MF-OMBR. It might be attributed to the fact that the salinity in the conventional OMBR was 10 times more than that in the MF-OMBR. As discussed above, the high salinity would inevitably lead to the increase of EPS.

3.4. Assessment of flux test

In order to compare the flux decline in the MF-OMBR with that in the conventional OMBR, the data reported in Wang et al. (2014) was collected and re-calculated according to Eqs. of 1–3. The flux decline due to different reasons for the conventional OMBR and MF-OMBR is summarized in Fig. 6. Apparently, the total flux decline (L_t) in the conventional OMBR was severer than that in the MF-OMBR, which was consistent with the variations of water flux in both OMBRs. However, the flux decline due to membrane fouling (L_f) in the MF-OMBR was about 45%, while it was only approximately 29% in the conventional OMBR, suggesting that the FO membrane had a severer membrane fouling in the MF-OMBR. Based on these facts, it could be indicated that the lower L_t in the MF-OMBR was not due to membrane fouling but because of the lower salinity (as shown in Fig. 2) achieved by the addition of the MF membrane. If further analyzing the data shown in Fig. 6, it could be found that the flux decline due to irreversible membrane fouling (L_{in}) in the MF-OMBR was less than that in the conventional OMBR, implying that the severe membrane fouling in the MF-OMBR was mainly due to the reversible membrane fouling. In fact, no obvious foulants could be found on the FO membrane surface in the conventional OMBR after continuous operation

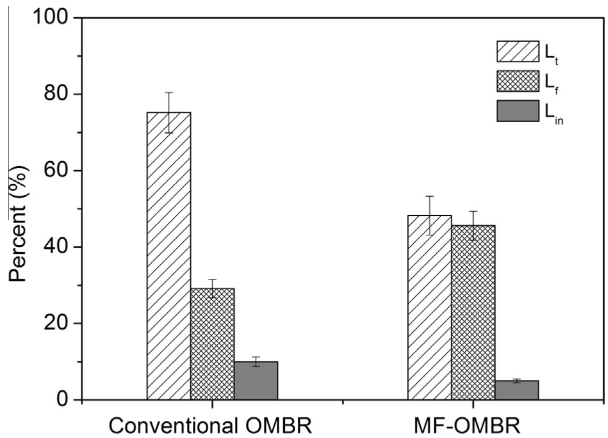

Fig. 6. Flux decline due to different reasons in the conventional OMBR and MF-OMBR. The data of the conventional OMBR was collected from Wang et al. (2014).

(Wang et al., 2014), while the FO membrane was covered with a thick cake layer in the MF-OMBR. The results on serious FO membrane fouling in the MF-OMBR obtained in this study were significantly different from most previous studies with results of slight FO membrane fouling (Achilli et al., 2009; Cornelissen et al., 2011).

3.5. Evaluation of membrane fouling

In order to further understand the severe FO membrane fouling in the MF-OMBR, some in-situ analytical instruments such as FESEM, EDX and FT-IR were used to investigate the virgin and fouled FO membranes. It could be observed from the FT-IR spectra of virgin and fouled FO membranes in the MF-OMBR (Supplementary Fig. S3) that there were significant differences in spectrum shape between virgin and fouled FO membranes. The spectrum of virgin membrane showed three sharp peaks at 1734, 1212 and 1033 cm^{-1}, respectively, which are attributed to the structure of CTA (Parida and Ng, 2013). However, the peaks at 1734 and 1212 cm^{-1} could not be observed in the spectrum of fouled membrane in the MF-OMBR. It might be due to the fouling layer covering on the fouled FO membrane surface, which affected the infrared absorption of the function bond of CTA. In comparison, the spectra of the fouled membrane presented some absorption peaks that were absent in virgin FO membrane. Two sharp peaks at 2920 and 2848 cm^{-1} due to stretching of C-H bonds were observed in the spectrum of the fouled FO membrane (Meng et al., 2007; Wang et al., 2008), and the spectrum of the fouled FO membrane also showed two sharp peaks (1629 and 1541 cm^{-1}), which are unique to the protein secondary structure, namely amides I and II (Meng et al., 2007; Wang et al., 2008). It indicated that protein was one of the foulants on the fouled FO membrane surface in the MF-OMBR. A Recent literature also reported that protein is a major foulant compound in the bio-separation process using filtration membranes (Li et al., 2013). In addition, a sharp peak at 1033 cm^{-1} exhibited the character of polysaccharides or polysaccharides-like substances (Croué et al., 2003), suggesting that polysaccharides were also presented in the fouled FO membrane surface. The FT-IR results demonstrated that the polysaccharides and proteins were the major foulants of the FO membrane, which was in good agreement with the previous study on the FO membrane biofouling in the conventional OMBR (Zhang et al., 2012).

SEM and EDX images of virgin and fouled FO membranes in the MF-OMBR (Supplementary Fig. S4) revealed that the fouled FO membrane was covered with a cake layer on the active layer compared with the virgin FO membrane. Zhang et al. (2012) has reported that the fouling mainly occurred not in the internal pore but on the FO membrane surface. As discussed in previous publications, the cake layer severely reduced the mass transfer coefficient and thus enhanced the external concentration polarization (ECP) except for directly decreasing the water permeability induced by the increase of membrane resistance (Zhang et al., 2012; She et al., 2012). Consequently, the formation of cake layer would result in the severe flux decline. Furthermore, it could be seen that the elements of C, N, O and P were observed on the virgin FO membrane surface, while more elements were detected on the fouled FO membrane surfaces such as C, N, O, Na, Mg, Al, Si, P, S, Cl, K, Ca and Fe. High rejection property of the selective layer of FO membrane might be responsible for the formation of inorganic scaling. The weight fraction distribution of all the elements (Supplementary Table S1) on the fouled FO membrane indicated that Na, Si, P, S, Cl, Ca and Fe were the main inorganic elements in addition to C. The biopolymers contain anion groups such as SO_4^{2-}, CO_3^{2-}, PO_4^{3-} and OH^-, and the cations such as Mg^{2+}, Al^{3+}, Fe^{3+} and Ca^{2+} could be easily precipitated by these negative ions (Wang et al., 2008; Mi and Elimelech, 2010). The organic foulants as mentioned in above sections coupled the inorganic precipitation would enhance the formation of cake layer and thus causing the severe membrane fouling in the MF-OMBR.

3.6. Economic analyses

Based on the fact that the economic feasibility is very important in order to decide any method for treatment of municipal wastewater, the economic analyses of the MF-OMBR and the conventional OMBR were conducted at a large-scale application of 240 m^3/d. With regard to the membrane-based bioreactor, the membrane costs are the main component of capital costs, and the aeration comprises almost 50–80% of the total energy costs (Drews, 2010; Wang et al., 2013). Thus, economic analyses of the MF-OMBR and the conventional OMBR were focused on the membrane costs and the energy costs due to aeration. Membrane costs of the MF-OMBR and the conventional OMBR were calculated based on the results obtained in this study and the related report (Wang et al., 2014), respectively. As shown in Table 1, it could be found that the membrane costs including the MF and FO membranes in the MF-OMBR were only 28.4% of that in the conventional OMBR. However, energy costs due to aeration in the MF-OMBR might be about 10% more than that in the conventional OMBR because of the bigger gas sparging velocity needed for the more serious membrane fouling in the MF-OMBR. It should be pointed out that the effluents of the MF-OMBR and the conventional OMBR should be further treated by the RO process in order to obtain the pure water. In this study, it was assumed that the mixed effluent of the MF-OMBR had the same water flux, operating pressure and water recovery as the effluent of the conventional OMBR during the RO process. Thus, the membrane and energy costs of the RO process for the MF-OMBR and the conventional OMBR were same. However, the membrane fouling during the RO process for treating the mixed effluent of the MF-OMBR should be severer due to its worse effluent quality.

3.7. Challenges and perspectives

Compared to the conventional OMBR system, the MF-OMBR system could effectively alleviate the salt accumulation and increase the water flux of FO membrane by the addition of the MF membrane. Additionally, the microbial activity in the MF-OMBR was much larger than that in the conventional OMBR due to the lower salinity inhibition, thus leading to a higher removal efficiencies of the activated sludge for TOC and NH_3–N.

Table 1
MF and FO membrane costs in the MF-OMBR and the conventional OMBR.

	MF-OMBR		Conventional OMBR
Membrane type	MF membrane	FO membrane	FO membrane
Membrane materials	PVDF	CTA	CTA
Company	Zizheng Environment Inc. (Shanghai, China)	Hydration Technology Inc. (HTI, Albany, OR)	Hydration Technology Inc. (HTI, Albany, OR)
Flux (LMH)	5	5.5	2
Membrane area (m^2)	980	930	5000
Price (US$/$m^2$)	120	240	240
Membrane costs (US$)	117,600	223,200	1,200,000
Total costs (US$)	340,800		1,200,000

Based on the above facts, it could be concluded that it is feasible of applying the MF membrane for reducing the salt accumulation in the OMBR. However, as a new OMBR system, there would be challenges associated with the application of MF-OMBR systems, especially in the aspects of the effluent quality and membrane fouling. As stated before, the effluent quality of MF membrane especially the TOC concentration was worse than that of FO membrane due to its lower retention, hence the total effluent quality of the MF-OMBR would deteriorate. In addition, the severer FO membrane fouling especially reversible membrane fouling in the MF-OMBR absolutely retarded the water passing through the FO membrane mainly due to the decrease of the effective driving force caused by the concentration polarization of solute on the FO membrane surface called ECP. In this case, the stable flux of FO membrane in the MF-OMBR was not satisfied and much lower than the expectation according to the lower salinity environment.

Therefore, future work should focus on the following aspects: (1) Testing different MF membranes and reducing the effluent production of MF membrane such as application of the intermittent operation mode in order to alleviate the MF membrane fouling and the reduction of water quality in the MF-OMBR, respectively; (2) Selection and development of an optimized FO membrane with lower fouling propensity; (3) Searching for the effective method for reducing the FO membrane fouling especially the reversible membrane fouling, for example, increasing the cross-flow velocity along the FO membrane surface.

4. Conclusions

The findings of this study demonstrated that the application of MF membrane could effectively control the salinity at a lower concentration and enhance the stable FO flux in the OMBR. The addition of MF membrane in the OMBR could increase TOC and NH_3–N removals due to the activated sludge by improving the microbial activity. The membrane fouling, especially reversible membrane fouling, was severer in the MF-OMBR than that in the conventional OMBR, which resulted in the severe FO flux decline mainly by increasing the filtration resistance and ECP.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.biortech.2014.05.121.

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