Performance of a novel baffled osmotic membrane bioreactor-microfiltration hybrid system under continuous operation for simultaneous nutrient removal and mitigation of brine discharge

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**Highlights**

- An OMBR-MF hybrid system performance was examined employing TFC-PA FO membrane.
- Simultaneous nitrification-denitrification achieved in a reactor inserting baffles.
- Reasonably high flux and low salinity build up were observed due to MF membrane.
- No physical or chemical membrane was performed during 38 days of operation.
- High organic matter and nutrient removal efficiency was obtained.

**Abstract**

The present study investigated the performance of an integrated osmotic and microfiltration membrane bioreactor system for wastewater treatment employing baffles in the reactor. Thus, this reactor design enables both aerobic and anoxic processes in an attempt to reduce the process footprint and energy costs associated with continuous aeration. The process performance was evaluated in terms of water flux, salinity build up in the bioreactor, organic and nutrient removal and microbial activity using synthetic reverse osmosis (RO) brine as draw solution (DS). The incorporation of MF membrane was effective in maintaining a reasonable salinity level (612–1434 mg/L) in the reactor which resulted in a much lower flux decline (i.e. 11.48–6.98 LMH) as compared to previous studies. The stable operation of the osmotic membrane bioreactor–forward osmosis (OMBR-FO) process resulted in an effective removal of both organic matter (97.84%) and nutrient (phosphate 87.36% and total nitrogen 94.28%), respectively.

**1. Introduction**

Diminishing fresh water supplies due to the impacts of global warming, rapid industrialization and urbanization have prompted...
increased interest in indirect and direct reuse of impaired water (Trussell, 2012; Wang et al., 2017). However, there are still many challenges faced in wastewater treatment processes, especially in relation to nutrient and trace organic removal (Nguyen et al., 2016). In particular, nutrient removal is very important for water reuse, especially to prevent water quality deterioration via eutrophication. (Devia et al., 2015; Mun et al., 2011). Fan et al. (1996) reported that perfect nitrification could be achieved in the membrane bioreactor (MBR) system (Ahn et al., 2003). However, the energy required for sludge recirculation and mixing in an anoxic tank accounts for 10–20% of the total energy consumption in a common MBR (Kurita et al., 2015). To overcome these shortcomings, researchers introduced the alternating anoxic and oxic conditions in a submerged MBR by intermittent aeration for total nitrogen removal. However, in the intermittently aerated MBR, filtration operation is limited to the aeration periods, mainly to prevent membrane fouling (Song et al., 2010).

In recent years, more studies have shown that nitrification and denitrification could occur concurrently in one single reactor under aerobic conditions with low dissolved oxygen, through the so-called simultaneous nitrification and denitrification (SND) process (Fu et al., 2009). Kimura and Watanabe (2005) have proposed a baffled membrane bioreactor, in which baffles are inserted in a submerged MBR, and the level of water in the reactor is controlled to facilitate simultaneous nitrification/denitrification without sludge recirculation. The inner zone of the baffles maintains an aerobic condition because of aeration, whereas the outer zone alternates between aerobic and anoxic conditions (Kimura and Watanabe, 2005). Thus, a baffled MBR offers advantages such as small footprint (no additional anoxic tank) and baffle design substitutes stirring of anoxic biomass and sludge recycle between oxic and anoxic tank (Kimura et al., 2007).

More recently, osmotic membrane bioreactors (OMBR) have attracted growing interests in the field of low strength domestic wastewater treatment (Wang et al., 2016b). OMBR have many advantages such as low and reversible fouling, minimum cleaning and energy efficient process (Luo et al., 2017; Wang et al., 2016a). However, OMBR also have some limitations such as salinity build-up (i.e. accumulation of dissolved salts inside the bioreactor) (Nguyen et al., 2016). In order to mitigate the salinity build up, various approaches have been tested including operating at short sludge retention time (SRT) (Wang et al., 2014b). However, ammonia removal via biological treatment in the OMBR cannot be completed at low SRT since the nitrifying bacteria population would decrease due to their relative long generation time. Moreover, diffusion of accumulated high concentration ammonia across the FO membrane eventually leads to the deterioration of permeate quality (Wang et al., 2014b; Yap et al., 2012). Therefore, for long-term operation of the OMBR, incorporation of microfiltration (MF)/ultrafiltration (UF) has been suggested to mitigate salinity build up. The MF/UF membranes could let the salts pass through but retain the activated sludge (Holloway et al., 2015a; Wang et al., 2014a).

Further, literature review shows that the concentrate produced from seawater reverse osmosis (SWRO) plants have up to twice more salt concentration than the receiving water (Tularam and Ilahee, 2007). As reported by (Abualtayef et al., 2016), the potential harm of brine to the environment yields from either its higher than normal salinity compared to point of discharge, or due to pollutants that otherwise would not be present in the receiving waterbody. These pollutants include chloride and other biocides, heavy metals, anti-scalants, coagulants, and cleaning chemicals (Abualtayef et al., 2016). Besides, in the case of thermal desalination, the hotter brine leads to environmental damage, especially to fragile ecosystems such as corals. Due to these negative effects, direct disposal to seawater of RO concentrates is doomed to disappear (Perez-Gonzalez et al., 2012). Jenkins et al. (2012) reviewed RO concentrate discharge regulations and standards which have been applied around the world particularly in the countries like the US, Australia and Israel. These range from salinity increments within 1 parts per thousand (ppt), 5%, or absolute levels such as 40 ppt. These limits typically apply at the boundary of a mixing zone whose dimensions are of order 50–300 m around the discharge (Jenkins et al., 2012). In contrast, although the TDS values of RO concentrates from brackish water desalting (from inland plants) are significantly less than seawater TDS, they are typically greater than 10,000 mg/L, which makes them more compatible with ocean water than fresh waters (Perez-Gonzalez et al., 2012). In this context, OMBR are presented as an innovative and viable technique to mitigate RO brine discharge.

The present study investigates for the first time the performance of an integrated osmotic and microfiltration membrane bioreactor system for municipal wastewater treatment employing baffles in the reactor. Thus, the single-stage reactor design employed here combines aerobic and anoxic processes to reduce the footprint and decrease energy costs of continuous aeration and sludge recycling in order to achieve simultaneous nitrification-denitrification. The process performance was investigated in terms of water flux and salinity build up, organic and nutrient removal and microbial activity using simulated RO brine as a DS.

2. Materials and methods

2.1. FO and MF membrane characteristics

The FO membrane used in this study was a flat-sheet TFC polyamide (PA) membrane (Toray, Korea). The characteristics of this membrane are detailed in Table S1 (SI). Membranes were stored in distilled water at 4 °C prior to use, and were oriented AL-FS (active layer facing feed solution) during the experiments with the feed solution being the OMBR mixed liquor. The membrane chemistry is proprietary, though it is believed that the TFC membrane has embedded polyester screen support and a negatively charge surface (Luo et al., 2016b). The submerged FO membrane module was custom designed and fabricated. Two stainless steel plates were attached to each side of the stainless steel block. The two FO membrane coupons were secured in place on each side of the stainless steel block with the stainless steel plates and then fixed using bolts and nuts. The channel in the membrane module ran through the stainless steel block having a width of 11 cm, a length of 12 cm, and a depth of 0.5 cm. Mesh spacers were used on DS side to provide additional support to the membrane and promote mixing of DS. Two ¼” nozzles were provided on each side of the stainless steel plates allowing DS to flow through the channel (Fig. S1 Supplementary information [SI]). The total effective FO membrane area was 264 cm². The MF membrane was supplied by Uniqflux Membranes LLP, India and was made of polyethersulfone (PES) with a nominal pore size of 0.33 µm and an effective surface area of 1000 cm². The characteristics of this membrane are shown in Table S1.

2.2. Feed and draw solutions characteristics

All the chemicals used in this research were of reagent grade (Sigma Aldrich, Australia). The influent water of the OMBR system was a synthetic municipal wastewater that consisted of 300 mg/L glucose, 50 mg/L yeast, 15 mg/L KH₂PO₄, 10 mg/L FeSO₄, 60 mg/L (NH₄)₂SO₄, and 30 mg/L urea. The synthetic wastewater prepared daily and had concentrations of total organic carbon (TOC), ammonium nitrogen (NH₄-N), total nitrogen and phosphate (PO₄-P) of...
100, 16, 28 and 3.5 mg/L, respectively. Sodium bicarbonate (NaHCO₃) was used for alkalinity to maintain a neutral pH. DS were prepared by dissolving 64 g/L sodium chloride in deionized (DI) water. Osmotic pressure and diffusivity were obtained by OLI Stream Analyzer 3.2 (OLI System Inc., Morris Plains, NJ, USA). For 1.1 M NaCl, electrical conductivity and osmotic pressure are 91.26 mS/cm and 51.78 atm, respectively.

2.3. Baffled osmotic membrane bioreactor-microfiltration (OMBR-MF) system and operation

A lab-scale baffled OMBR-MF system was used in this study and a schematic of the system is shown in Fig. S2 (SI). This hybrid system consisted of a feed solution reservoir, a plexiglass bioreactor with a submerged plate and-frame FO membrane cell and a hollow fiber MF membrane module, a concentrated DS reservoir and a diluted DS reservoir. The bioreactor tank (i.e., 24.5 cm length * 15.5 cm width * 40 cm height) had an effective volume of 11.5 L. On the three inside walls of the tank, plexiglass partition of 25 cm length was running from top to 5 cm above the bottom of the tank thus making hollow baffle box inside the tank with a size of 18.5 cm length, 12.5 cm width and 25 cm height. The baffles were bent at approximately 30° angle in the end (3.5 cm length) to avoid dead zone formation and to attain thorough mixing of biomass. The volume ratio of the outer tank to the inner tank was approximately 1.9 when the water level was at the top of the inserted baffles (Fig. S3 SI). By changing the position of the level controller, theoxic and anoxic cycle times can be adjusted accordingly. The concept and operating details of the baffled reactor is discussed elsewhere by Kimura et al. (2007).

A plate-and-frame membrane module was prepared using commercial TFC FO membranes (Toray, Korea) and the module was immersed in the bioreactor tank for osmotic filtration. FO membrane samples were suspended vertically and parallel to the MF membrane module. The air diffuser was installed inside theoxic chamber of the bioreactor for oxygen supply at 2 litres per minute (LPM) air-flow rate thus subjected both membranes to air scouring. The MF membrane, operated continuously, was driven by a peristaltic pump (Longer BT100 2 J). The MF permeate flux was changed manually in accordance with the change of FO water flux in order to maintain stable oxic and anoxic cycle times during the entire operation (i.e. 38 days). A high resolution (±0.1 kPa) pressure sensor (Keller, Reinachterstrasse, Basel, Switzerland) was installed to record the trans-membrane pressure (TMP). Both of the concentrated and diluted DS reservoirs were placed on the weighing balance (Adam PGL 15001) and connected to a computer. The weight difference between the diluted and concentrated DS was used to calculate the FO water flux. A water level controller was used to adjust theoxic-anoxic cycle time as well as to regulate the feed pump (peristaltic pump Longer WT600 2 J) to feed synthetic wastewater to the bioreactor.

The seed sludge was collected from the recycled water facility at Central Park, Sydney, Australia. The sludge was acclimatized for a month prior to adding into the baffled OMBR-MF system. The OMBR-MF hybrid system was continuously operated for 38 days under similar conditions at a constant temperature of 22 ± 1 °C. The mixed liquid suspend sludge (MLSS) was adjusted to 4700 mg/L initially. Throughout baffled OMBR-MF operation, the sludge retention time (SRT) was controlled at 115 days by daily wasting 100 mL of mixed liquor from the bioreactor. The hydraulic retention time (HRT) for baffled OMBR + MF combined system was set at 30.25 h. A 1.1 M NaCl DS was used as simulated reverse osmosis (RO) brine. The concentrated DS was refilled twice a day and the diluted DS tank was emptied. The salt accumulation in the bioreactor was determined by monitoring the conductivity of the mixed liquor with a conductivity meter. The pH, total dissolved solids (TDS) and conductivity of the mixed liquor, permeate and DS were measured regularly (HACH, Germany). The operating conditions are listed in Table S2 (SI). No membrane cleaning was conducted for both FO and MF membrane during the entire operation (i.e. 38 days).

A detailed mass balance of the OMBR-MF hybrid system is also presented in Fig. S4 and Equations S1 to S11 (SI) to provide a better understanding of the salt accumulation phenomena occurring in the bioreactor.

2.4. Analytical methods

2.4.1. Measurement of water flux

The experimental water flux $J_w$ (L/m² h) was calculated by measuring the net increase in diluted DS volume with time as follows:

$$J_w = \frac{\Delta V}{A \Delta t}$$

where $\Delta V$ is the total increase in the volume of the permeate water (L) collected over a predetermined period, $\Delta t$ (h) and $A$ is the effective FO membrane area (m²).

2.4.2. Biological parameters and basic water quality parameters

The mixed liquid suspended solids (MLSS), mixed liquid volatile solids (MLVSS) and the specific oxygen uptake rate (SOUR) of the mixed liquor in the OMBR were determined according to the APHA, AWWA, WEF (1998). The concentration of dissolved oxygen was measured by using a DO meter (Vernier, USA), TOC of the influent and effluent was measured using the Analytikjena Multi N/C 2000. Chemical oxygen demand (COD) was analysed according to standard methods (APHA, 1998). NO₃-N, NO₂-N, NH₄-N, TN and PO₄-P were measured using Hach TNTplus™ reagent vials by photometric method (Spectroquant Cell Test, NOVA 60, Merck). Samples were diluted as necessary to minimize chloride interferences and ensure that analytes were within the desired range.

2.4.3. SEM-EDX analysis

The surface and cross-sectional morphologies of pristine and fouled membrane samples were observed by scanning electron microscopy and an energy diffusive X-ray (EDX) analyzer (SEM, Zeiss Supra 55VP, Carl Zeiss AG). Samples taken from each membrane were coated with gold. The SEM images were carried out at an accelerating voltage of 10 kV, and different image magnifications at various areas were obtained for each sample.

3. Results and discussion

3.1. Water flux and salinity build up in the baffled OMBR-MF hybrid system

The OMBR-MF hybrid system was operated at constant DS concentration of approximately 64 g/L (i.e., 1.1 M NaCl). Water flux of both FO and MF membranes as well as TDS concentration in terms of mg/L NaCl (Equation S10 (SI)) as a function of time over the course of the OMBR-MF experiments are shown in Fig. 1. From the beginning of baffled OMBR-MF operation, MF was operated to mitigate the salinity build up in the reactor (Equation S11 (SI)). Initial FO flux was 11.9 LMH and this flux varied in the range of 11.54–6.98 LMH during the 38 days of continuous operation. During the first five days of operation, more than 9.5 LMH FO flux was observed and then decreased by 1 LMH during the first three weeks of operation. In between 18 and 30 days, the FO flux fluctuated around 8 LMH. In the final phase of the OMBR-MF operation, FO flux gradually decreased to around 7 LMH. Overall, an average
of 8.56 LMH FO flux was achieved during the 38 days of continuous operation. The decrease in the FO water flux could be related to the internal concentration polarization (ICP) effect, salt accumulation and biofoulants accumulation on the membrane surface due to MLSS in the reactor. The FO flux decline during OMBR operation has also been reported by other researchers (Wang et al., 2014a). Recently, Wang et al. (2016a) observed that the water flux in the OMBR with TFC FO membrane quickly reduced from about 15.3 LMH to approximately 8.0 LMH during the first 8 days of operation. The FO flux slightly decreased to the final value of about 3.0 LMH while 22 mS/cm mixed liquor conductivity was reached at the end of the operation. In another continuous OMBR study an average 9 LMH flux was achieved over 30-day examination. However, in order to maintain 9 LMH flux, backwashing was performed at 14, 21 and 28 days respectively (Achilli et al., 2009). In the present study, no membrane cleaning was performed during the 38 days of continuous operation and almost similar average flux (8.56 LMH) was obtained. Moreover, (Holloway et al., 2015b) operated OMBR with UF membrane and reported that during the first three weeks, when OMBR was operated without UF membrane, flux declined considerably (down to 4.2 LMH) and thereafter, by incorporation of UF membrane, FO flux was found to increase and remain stable (4.8 LMH) for the rest of the study. In another MF-OMB study by Wang et al. (2014b), the MF membrane was continuously operated under constant flux at 5 LMH but significant FO flux decline was reported during the first 30 days of operation after which almost stable FO flux (5.5 LMH) was obtained. In the present study, even though MF membrane was operated in parallel to FO, the FO flux decline was gradual and relatively slow. Nevertheless, MF was operated at very low flux (1.1–2.4 LMH) while high FO flux was continuously achieved (average of 8.56 LMH). This also could be attributed to the high permeability of TFC PA membrane in comparison to the above cited studies that employed CTA membrane. As reported by Luo et al. (2016a), salinity build-up in the bioreactor is an intrinsic phenomenon associated with OMBR operation and the rate of solute diffusion through the membrane depends on membrane selectivity, diffusion coefficient of the solute and on the concentration difference across the membrane (Holloway et al., 2015a; Nguyen et al., 2016). In practice, to prevent the inhibition of the microbial community activities due to reverse salt flux, the maximum bioreactor tank salinity must not exceed 2 g/L (Holloway et al., 2014; Nguyen et al., 2015). In this study, during the course of operation, the conductivity, TDS and pH (data not shown for pH) in the reactor, FO and MF permeates were measured regularly. Results showed that, due to simultaneous operation of MF membrane with FO, the TDS value did not increase significantly and remained almost stable in the mixed liquor (612–1434 mg/L). The salt permeating through the MF membrane was also helpful to maintain a high driving force between concentrated DS and mixed liquor. It could be seen that the trend of salinity (in terms of EC) variation of the reactor mixed liquor and MF permeate was similar (Fig. S5, SI) indicating that the incorporation of MF membrane to discharge the soluble salt could effectively decrease the salinity and further alleviate the salt accumulation in the OMBR.

The measured TDS concentration in the bioreactor changed from 612 to 1434 mg/L corresponding to 1.24–2.92 mS/cm during the first week of study and then after, an average TDS of less than 1200 mg/L was obtained during the rest of investigation time. This salinity range is well below the value reported in previous studies where a stable mixed liquor conductivity of approximately 5 mS/cm was observed during OMBR operation with continuous MF extraction (Luo et al., 2016a; Qiu et al., 2015; Wang et al., 2014b). Further, the result obtained here compares favourably with Luo et al. (2015) work as they reported that after a small increase in the first week, the mixed liquor conductivity stabilised at approximately 400 mg/L after incorporation of MF membrane. Similarly, Holloway et al. (2015b) observed that TDS reached a peak concentration (approximately 8000 mg/L) during OMBR testing without UF membrane. This could be attributed to the small hydrated radius of monovalent ions (Na with a hydrated radius of 0.18 nm and Cl with a hydrated radius of 0.19 nm) which could easily pass through the FO membrane (membrane pore size: 0.37 nm) (Nguyen et al., 2016). In the previous study, as soon as the UF subsystem was operated in parallel to OMBR, the TDS concentration rapidly declined and remained constant at approximately 1000 mg/L until the end of the UF-MBR investigation. After incorporation of UF membrane, a stable FO flux of 4.8 LMH was achieved over the duration of the investigation without a single membrane cleaning (Holloway et al., 2015b). Finally, in their work on OMBR, Alturki et al. (2012) observed a rapid increase in the mixed liquor conductivity from 0.27 to 8.27 mS/cm within seven days without housing the submerged MF/UF membrane in the bioreactor (Alturki et al., 2012). It should be emphasized that during the 38 days of baffled OMBR-MF test runs, both the FO and MF membranes were not offered any physical cleaning nor backwashing. During long-term operation, FO biofouling could significantly affect the OMBR-MF hybrid system performance. In order to mitigate biofouling of FO membrane different cleaning techniques can be adopted. The first one could be the physical cleaning of the FO membrane. For this cleaning strategy, OMBR operation should be stopped and FO membrane module should be taken out from the system followed by cleaning with deionised water (DI) and then gentle cleaning with sponge ball. Since fouling layer in FO process is not compact with no applied pressure (She et al., 2016), one step cleaning can recover the desired initial flux. When the flux recovery is not satisfactory then chemical cleaning can be performed. As reported by Holloway et al. (2015), chemically enhanced osmotic backwashing is conducted by replacing the draw solution with a very low salinity base (NaOH) and/or acid (HCl) cleaning solutions, which are continuously recirculated on the draw solution side of the membrane.

3.2. TOC and PO₄-P removal

Biological process performance of the baffled OMBR-MF hybrid system was assessed with regards to the removal of basic contaminants (i.e. TOC, NH₄-N, TN, and PO₄-P), sludge production, and biological activity. TOC removal in reactor mixed liquor, MF permeate and diluted draw solution (FO) was 91.31%, 93.51% and 97.84% respectively. The average TOC concentration in the feed, reactor, MF permeate and FO was 98.55 mg/L, 8.55 mg/L, 6.25 mg/L and
2 mg/L respectively (Fig. 2(a)). The removal of TOC from the OMBR FO channel was over 97% during the entire experimental period. As in the previous study Qiu and Ting (2013) noted that the overall removal rate of organic matter constantly reached up to 98% and the TOC in the DS was less than 5.0 mg/L during OMBR operation. Recently, Wang et al. (2016a) also achieved 96% TOC removal with TFC membrane when treating 140 mg/L fed TOC, with an average of 5.2 mg/L TOC reached in FO permeate.

In the current study, higher TOC removal has been achieved in both reactor and FO permeate as compared to (Wang et al., 2016a). Thus, biological degradation contributed greatly to reduce the concentration of TOC, overcoming the concentration process caused the FO membrane rejection. In the diluted DS and in the MF permeate, TOC concentration further decreased due to the FO and MF membrane rejection, respectively. Nevertheless, the reverse draw solute flux undesirably impacted the biological treatment of OMBR. In fact, TOC concentration in the bioreactor increased slightly at the beginning of OMBR operation (day 7 and 9 respectively). This observation is consistent with that reported by (Luo et al., 2016a,b) and could be attributed to the high rejection of almost all the organic matter by the FO membrane, which caused a significant accumulation of non-degradable or/and refractory dissolved organic matter (DOM) within the bioreactor (Qiu and Ting, 2013).

It has been reported that, during OMBR operation, increase salinity in the bioreactor could inhibit the metabolic activity of biomass and plasmolysis causing the release of intracellular constituents and soluble microbial products (Wang et al., 2014a). This study also showed that the presence of high TDS can interfere with the oxygen transfer and affect the biological metabolism thereby reducing the capacity of the reactor to sustain shock loads. The incorporation of MF membrane in the present study successfully kept a salinity level well within the control (Fig. 1) leading to high TOC removal efficiency (Fig. 2(a)).

Phosphate can be eliminated by two different mechanisms: assimilation and luxury uptake (Rosenberger et al., 2002). The baffled OMBR-MF system showed very stable and effective performance in achieving high removal of PO4–P with 81.22%, 87.36% and 93.46% in reactor, MF and FO processes (Fig. 2(b)). A relatively high and stable phosphorous removal was observed corresponding to the average 0.23 mg/L obtained in FO permeate. In the baffled OMBR, mixed liquor suspension is cyclically exposed to aerobic and anoxic conditions, and enhanced biological phosphorus removal might have occurred to some extent. In the present study, the observed low DO concentration in the anoxic zone can create a pseudo-anoxic condition which would have favoured phosphorous release. During aerobic condition phosphorus uptake by bacteria could have happened. Thus, phosphorus removal can be achieved similarly to what was observed by (Kimura and Watanabe, 2005). Another possible explanation for the good removal of phosphorus is precipitation with inorganic substances. Aggregates of phosphorus and inorganic substances would settle in dead zones of the bottom of OMBRs (Rosenberger et al., 2002). In present study, salting out might have occurred and some potassium phosphate in the reactor from feed wastewater may have precipitated out with the salt transported in the reactor from draw solution. Thus, regular sludge withdrawal might have achieved phosphorus removal through biomass and settled phosphorus discharge from dead zones. Guo et al. (2008) achieved more than 98% of PO4–P removal in sponge-submerged membrane bioreactor. The explanation given was due to the sponge providing a good anaerobic condition around the surface of the sponge and the anaerobic condition inside the sponge which makes the aerobic submerged membrane bioreactor able to achieve a higher removal efficiency of PO4–P (Guo et al., 2008). In our study, during the OMBR operation it was observed that biomass clung to the outer baffle wall as well to the inside wall of the reactor in anoxic zone. Specifically, when switching over to anoxic cycle, the low DO concentration in the anoxic zone might have created anaerobic condition inside of the biomass that attached to the wall and phosphorous release would possibly have occurred. Qiu et al. (2015) reported 97.9% of phosphate phosphorus (PO4–P) rejection by the FO membrane in a hybrid microfiltration-forward osmosis membrane bioreactor (MF-FOMBR). In two studies by Nguyen et al. (2015, 2016), their OMBR systems achieved more than 99% and more than 98% phosphate (PO4–P) removal respectively. Moreover, Holloway et al. (2007) also showed that very high rejection of phosphate (99.6–99.9%) by the FO membrane could be attained during concentration of anaerobically digested sludge centrate. Indeed, the FO membrane can almost completely reject PO4–P due to its negative charge and the relatively large radius diameter (0.49 nm) of the orthophosphate ion, which is the dominant phosphate species under the conditions tested (Aftab et al., 2015; Praveen and Loh, 2016).

The polyphosphate accumulating organisms are susceptible to saline conditions, and the increased osmotic pressure within their cells due to salt accumulation could diminish their phosphate accumulating capacity (Lay et al., 2010). Kinetics studies have suggested that nitrogen and phosphorus removal efficiency dropped to 20% and 62%, respectively, when salt concentration was 5% NaCl in the bioreactor (Nguyen et al., 2016). However, in this study, PO4–P build-up in the bioreactor was not observed which can be.
attributed to reasonably low salinity as compared to other OMBR reports.

3.3. Nitrogen removal

In the proposed baffled OMBR-MF hybrid system, nitrification is carried out in the whole chamber when the liquid level was above the top of the inserted baffles (Fig. S2) while denitrification proceeds in the outer (anoxic) zone when the liquid level was low. Fig. 3(a) shows the removal of NH$_4$–N during baffled OMBR-MF continuous operation. Initially, 97% of NH$_4$–N was removed by the reactor, which then decreased down to 80% after 3 days of operation as shown by the NH$_4$–N concentration increase observed in the reactor. The stability of nitrification was thus affected since the increase in supernatant NH$_4$–N concentrations on day 7 (12.5 mg/L) and day 9 (8.9 mg/L) significantly affected effluent concentrations. The drop in the NH$_4$–N removal could be attributed to the effect of salinity on biomass. Additionally, the ammonia-oxidizing bacteria (AOB) are generally slow growing and more sensitive to changes in environmental conditions such as temperature and salinity (Qiu and Ting, 2013). Therefore, the activity of the AOB and nitrite-oxidizing bacteria (NOB) could easily be inhibited by elevation of salinity (non-halophilic bacteria) which hampered the biological conversion of NH$_4$–N (Ye et al., 2009). Ye et al. (2009) also showed that a high salinity of 1.02 (W/V%) can be detrimental to the survival of many AOB and other bacteria. Besides, the survival bacteria were also shown to be strongly inhibited. Consequently, the NH$_4$–N removal efficiency decreased greatly. When the salinity level in the bioreactor was then stabilised after 11 days, the nitrifiers regained their potential to remove NH$_4$–N and as a result the nitrifying activity was restored. In fact, after 19 days of operation, the supernatant NH$_4$–N concentration decreased significantly and the conversion of NH$_4$–N finally recovered to more than 97% till the end of the study.

Furthermore, although nitrifiers are slow growing bacteria, the proposed baffled OMBR-MF with a prolonged HRT of 30.25 h and 115 days SRT was favourable for the relative long generation time of the nitrifying bacteria allowing better removal efficiencies. Aftab et al. (2015) also observed similar behaviour in their study when targeting NH$_4$–N removal. In current study, the average NH$_4$–N concentration of 0.84 and 0.46 mg/L in the MF permeate and diluted DS was obtained respectively. Wang et al. (2016a) achieved about 97% NH$_4$–N removal in mixed liquor and 99% NH$_4$–N removal in FO permeate respectively. Most of the OMBR studies reported almost perfect nitrification. However, in this study complete nitrification has not been observed. It is worthwhile to note that in order to achieve simultaneous denitrification, air flow rate was kept quite low during baffled OMBR-MF operation. Therefore, in addition to salinity effect, low air flow rate might have created limited oxygen supply and non-homogeneous aeration; hence complete nitrification might have been compromised. In fact, Kurita et al. (2015) studied a baffled MBR and observed that, due to reduced aeration rate, the supply of oxygen to the biomass was apparently not sufficient; resulting in limited nitrification and an increased concentration of NH$_4$–N in the treated water. This result can also be correlated to the aerobic-anoxic cycle time (30 min–90 min, respectively). In fact, due to prolonged anoxic cycle time, denitrification performance possibly would have been improved though offset by the increase of NH$_4$–N. However, during the second and third week of baffled OMBR-MF operation, reasonably high but unstable removal was observed and then from 20 day onwards, more than 95% NH$_4$–N removal in mixed liquor was achieved.

Total nitrogen concentration in the treated water was apparently lower than that in the feed water (Fig. 3(b)). This reduction in TN was accomplished by denitrification due to the creation of anoxic conditions associated with the insertion of the baffles. Hence, the TN removal was well achieved under elevated salinity conditions and overall TN removal rate reached 94.28% in the diluted DS (FO permeate). Considering the all system, 70.38% of average TN removal could be achieved by the reactor (biological process) without recirculation of mixed liquor for 38 days.
continuous operation. Indeed, removal of nitrogen was significant without addition of external carbon, indicating the effectiveness of the proposed baffled OMBR-MF hybrid system. Fig. 3(c) shows the changes in concentration of NH$_4^+$-N, NO$_2^-$-N, and NO$_3^-$-N in the mixed liquor during the operation. Denitrification allows maintaining a relatively low NO$_3^-$-N concentration with an average concentration of 5.12 mg/L in mixed liquor supernatant within the 38 days of operation. TN removal efficiency is also plotted in Fig. 3(c). In the aerobic bioreactor, TN consumption occurs mainly through microbial assimilation. At the same time, nitritification converts NH$_4^+$-N to nitrite (NO$_2^-$-N) and then nitrate (NO$_3^-$-N) under aerobic conditions (Luo et al., 2016b). Incomplete nitritification is usually manifested by the detection of both NH$_4^+$-N and NO$_2^-$-N in the bioreactor. From Fig. 3(c), it is clearly seen that the removal of nitrogen in the baffled OMBR-MF was limited by nitritification especially during the first week of operation. However, considerably good denitrification has been achieved throughout the operation. Overall, TN removal was considerably high in this study.

Fig 3(c) shows that, when NO$_3^-$-N concentration in the mixed liquor increases, the T-N in the treated water also increases and TN removal efficiency decreases. Also, in the baffled OMBR-MF system, the high rejection of the FO membrane prolonged the retention time of NO$_2^-$-N and NO$_3^-$-N within the bioreactor, which also facilitated the removal of NO$_2^-$-N and NO$_3^-$-N to nitrogen during the anoxic cycle. One explanation for the better TN removal performance was the improvement in the creation of an anoxic environment. This was confirmed by dissolved oxygen (DO) measurements in the anoxic zone. Fig. 4 shows the DO profile in outer zone of the baffles at two different depths in the aerobic and anoxic zone, respectively. The measured values shown in Fig. 4 were obtained on day 31, when good removal of nitrogen was observed. As can be seen in Fig. 4, at time (t = 0 min) a wastewater pump has started and thus wastewater was fed to the reactor. When water level in the reactor reached on top of the inserted baffles at desired height, the level controller sensor activated and then feed pump stopped automatically. After 30 min, the water level had dropped back to the top of the inserted baffles (end of aerobic cycle time). In order to achieve efficient denitrification in the system, creation of a good anoxic condition in the exterior zone of the reactor is essential. Further, the denitrification is the limiting step in the removal of nitrogen in the operation. In present work, a good anoxic condition was achieved in the outer zone (DO < 0.5 mg/L as shown in Fig. 4). The experimental data also confirmed that the outer zone worked as an anoxic reactor for a long period in the total operation time which probably explained the superior denitrification performance of the proposed baffled OMBR-MF system. Furthermore, the experimental results also proved that the creation of an anoxic environment could be achieved even in the bottom part of the reactor at 18 cm depth (Fig. 4). This was probably due to the high concentration of MLSS (4.7–6.1 g/L) which eventually promoted better denitrification. Several studies have indicated the negative effect of salinity on MLSS (as low as 1 g/L MLSS) which adversely led to poor MLVSS/MLSS ratio as low as 0.45 (Wang et al., 2014a). Finally, in the current study, a reasonably high C/N ratio of about 14 has been maintained in baffled OMBR-MF system which is favourable enough to prevail better denitrification and reduction of total nitrogen for baffled OMBR-MF hybrid system.

### 3.4. Biomass activity

Water extraction by the MF membrane from OMBR mixed liquor did not significantly impact biomass characteristics (Fig. 5). Since very little excess sludge was discharged everyday (i.e. 115 days SRT) from the bioreactor, the MLSS concentration improved with time during the operation. In the later stage (21–38 days), the growth rate of MLSS was steady about 6 g/L due to low sludge organic loading. The MLSS concentration in the reactor varied from 4.7 to 6.1 g/L over the 38 days of continuous operation. Some studies (Li et al., 2016) demonstrated that high salinity in the mixed liquor adversely impacts on the MLSS. For example, (Wang et al., 2016a) operated OMBR, employing both CTA and TFC membranes, at 0.76 g/L stable MLSS and the MLVSS/MLSS ratio dropped down to 41% within 33 days operation. This finding was correlated with the increase salinity level in the reactor; (reaching up to 20 mS/cm), clearly showing the adverse effect of salinity on microbial activity. In another study, (Luo et al., 2016b) noted that a small but noticeable decrease in MLSS concentration was observed during OMBR operation with 0.5 M NaCl as a DS and yet at infinite SRT conditions.

Alturki et al. (2012) reported that build-up of salinity level up to 4.13 g/L of NaCl led to a gradual decrease in the ratio of MLVSS over MLSS from 0.87 to 0.66 after seven days of operation. The decrease in the MLVSS/MLSS ratio indicates that biological activity of the reactor may have deteriorated over time (Alturki et al., 2012). Further, an increase in the osmotic stress could result in the dehydration and plasmolysis of bacterial cells and thus reduce their viability (Luo et al., 2016a). However, in the present work, the relatively low salt concentration (<1.5 g/L over the entire operating time) in the bioreactor enabled the normal growth of the microbial community due to continuous salt bleeding by MF membrane and daily withdrawn mixed liquor (100 mL) from the bioreactor. Thus, high concentration of active biomass as well as a stable and high

![Fig. 4. Dissolved oxygen (DO) profile with aerobic-anoxic cycle time in baffled OMBR-MF hybrid system.](image)

![Fig. 5. Variation of MLSS, MLVSS and MLVSS/MLSS ratio and SOUR in baffled OMBR-MF hybrid system with time.](image)
The specific oxygen uptake rate (SOUR) was widely used to understand the effect of a higher concentration of materials including salt on the activity of biomass in various aerobic processes (Choi et al., 2007). In this study, respiration test of the activated sludge showed a significant decrease in the SOUR from an initial 4.51–3.03 mg O₂/g MLVSS/h during the first week of baffled OMBR-MF hybrid system, suggesting a deterioration of biological activity (Fig. 5). This decrease in SOUR could be well correlated to the increase in salinity during the first week of operation. These observations are consistent with previous studies especially within the first two weeks of operation and could be attributed to the inhibition of elevated bioreactor salinity on biomass growth and activity (Luo et al., 2016a). Indeed, as can be seen in Fig. 5, in the following weeks, measured SOUR was well within the optimum range prescribed for MBR operation (i.e. 3–5 mg O₂/g MLVSS/h) while the salinity level in the bioreactor was stabilised (Fig. 1).

3.5 Fouling behaviour

Fig. S6(a) (SI) shows the image of FO membrane before and after (fouled) 38 days of continuous operation. No significant foulant deposition has been observed on the membrane surface. Since the baffled OMBR-MF system was operated with the membrane active layer facing the feed side (AL-FS mode), foulant build up occurs on the active layer, where it could easily be removed by hydraulic shear force due to continuous aeration (Mi and Elimelech, 2008). This is different from what occurs with microporous membranes in traditional MBR, where the initial foulant deposition takes place within the porous structure of the membranes, and hydraulic shear forces cannot effectively remove the foulant (Qiu and Ting, 2013). Fig. S6(b) (SI) shows SEM images of both pristine and fouled TFC membranes, respectively. It could be observed from SEM images that the surface of pristine TFC FO membrane is rough and more rugged (valley like structure-Magnification × 5000). As shown in SEM observation for fouled membrane, compared with the original membrane, the active layer of fouled membrane surface was almost fully covered with a rather thin and compact gel-like fouling layer. The foulant layer was 2.38 μm thick as deduced from the SEM image. This is smaller than the typical thickness of 20–50 μm fouling layer found in MBRs (Lay et al., 2011). It is worthwhile to note that the fouling layer on the FO membrane surface was very thin, and it had only a small effect on the water flux during the entire operation time. The EDX analysis for both pristine and fouled membrane (Fig. S7) was also performed. From EDX analysis of pristine membrane, it can be seen that the TFC FO membrane surface mainly includes C, O and S. On the other hand, many inorganic elements such as Na, Ca, Fe, Al, Cl, P and Si were observed on the fouled membrane surface. The source for many of those inorganic elements was most probably the synthetic wastewater (e.g. Fe, P). The presence of Na and Cl could be correlated to the reverse salt diffusion from the DS.

4. Conclusions

Primary findings drawn from this study can be summarized as follow:

- The performance of a novel baffled OMBR-MF hybrid system was examined for water flux, salinity build up and nutrient removal specifically SND was successfully achieved in a single reactor.
- An average of 8.56 LMH FO flux was achieved during 38 days of continuous operation.
- MF membrane incorporation alleviated salinity build up and hence better performance was achieved.
- The dissolved oxygen profile during aerobic-anoxic cycle confirmed < 0.5 mg/L oxygen favourable for denitrification.
- More than 97% TOC, 87% PO₄-P and 94% TN removal was confirmed. From EDX analysis of pristine membrane, it can be seen that Al, Cl, P and Si were observed on the fouled membrane surface. On the other hand, many inorganic elements such as Na, Ca, Fe, Al, Cl, P and Si were observed on the fouled membrane surface.

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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.2017.03.069.

References


