**Simultaneous nitrification-denitrification using baffled osmotic membrane bioreactor-
microfiltration hybrid system at different oxic-anoxic conditions for wastewater
treatment**

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Abstract

The efficacy of a baffled osmotic membrane bioreactor-microfiltration (OMBR-MF) hybrid system equipped with thin film forward osmosis membrane for wastewater treatment was evaluated at laboratory scale. The novel OMBR-MF hybrid system involved baffles, that separate oxic and anoxic zones in the aerobic reactor for simultaneous nitrification and denitrification (SND), and a bioreactor comprised of thin film composite-forward osmosis (TFC-FO) and polyether sulfone-microfiltration (PES-MF) membranes. The evaluation was conducted under four different oxic-anoxic cycle patterns. Changes in flux, salinity build-up, and microbial activity (e.g., extracellular polymeric substances (EPS) were assessed. Over the course of a 34 d test, the OMBR-MF hybrid system achieved high removal of total organic carbon (TOC) (86–92%), total nitrogen (TN) (63–76%), and $\text{PO}_4\text{-P}$ (57–63%). The oxic-anoxic cycle time of 0.5-1.5 h was identified to be the best operating condition. Incorporation of MF membrane effectively alleviated salinity build-up in the reactor, allowing stable system operation.

Keywords: Microfiltration (MF); Osmotic membrane bioreactor (OMBR); Simultaneous nitrification-denitrification (SND); Salinity build-up.

1. Introduction

Membrane bioreactors (MBRs) have been extensively used for municipal and industrial wastewater treatment. MBRs typically produce high quality of water, smaller foot print, and less sludge yield, as compared with conventional activated sludge (CAS) process (Cornelissen et al., 2008; Wang et al., 2011). MBR is generally based on microfiltration (MF) or ultrafiltration (UF) membranes for the retention of suspended solids and bacteria. Despite the successful applications with MBR, membrane fouling remains as a challenge. Fouling reduces permeate flux. This is due to the deposition of soluble and particulate materials onto and into the membrane, attributed to the interactions between activated sludge components and the membrane and increases the frequency of membrane cleaning and replacement (Achilli et al., 2009). The particulate, colloidal and soluble organic matters all can be the potential membrane foulants (Defrance et al., 2000) and significant relative contribution of soluble microbial products (SMP) to membrane fouling has been reported (Wang et al., 2011).

An innovative osmotic membrane bioreactor (OMBR) has a strong potential for minimising membrane fouling. Natural osmotic pressure (27 bar) is a driving force in OMBRs than hydraulic pressure in conventional MBRs. OMBR offers low fouling propensity and less cleaning frequency due to a lack of hydraulic pressure across the membrane. Also, membrane fouling in OMBR is largely reversible (Chekli et al., 2017; Zhao et al., 2019). The FO membrane pore size is in the range of 0.25–0.37 nm which is 2-3 orders of magnitude smaller than the MF or UF membrane used in MBR (Zhang et al., 2019). In OMBRs, hydrophilic FO membrane can achieve inorganic and organic compounds rejection similar to reverse osmosis (RO). However in contrast, hydrophilic MF or UF membranes in MBRs achieve comparatively less rejection of organic and inorganics than FO, and induce severe membrane fouling caused by high hydraulic pressure (Kim et al., 2019).

1 In OMBRs, water flows through a FO membrane from the activated sludge (or bioreactor) side
2 (also called as feed side, FS) to the concentrated draw solution (DS) side with osmotic pressure
3 difference as a driving force. Despite the advantages of OMBRs, accumulation of salts on the
4 membrane in the feed side remains a challenge (Wang et al., 2014b). The accumulation of salts
5 in the bioreactor, which is caused by reverse salt transport of draw solutes (from draw to feed
6 sides) and high rejection of feed solutes by the membrane in the feed side, induces an elevated
7 salinity condition (concentration polarization) in the feed side. This subsequently reduces the
8 osmotic pressure difference between the feed and permeate sides (i.e. driving force), reducing
9 permeate flux. Moreover, the salt accumulation can inhibit bacterial viability and microbial
10 consortia in the bioreactor (Wang et al., 2014a), which can deteriorate the biological treatment
11 process. Various approaches including minimisation of sludge retention time (SRT) have been
12 tested to mitigate the salinity build-up (Wang et al., 2014a). A short SRT resulted in a low
13 sludge concentration, e.g., the mixed liquor suspended solid (MLSS) was in the range of 1.02
14 $\pm$ 0.10 g/L in the OMBR at SRT of 10 d, which could reduce the performance of an OMBR
15 (Aftab et al., 2017; Bell et al., 2016; Zhu et al., 2018). Moreover, at lower SRTs biological
16 nitrification could not be achieved, since the nitrifying bacterial community requires a
17 relatively long SRT (Wiesmann U. et al., 2006). Moreover, accumulation of ammonium ions
18 likely enhance the diffusion of ammonium ions through FO membrane to DS, which
19 deteriorates permeate water quality (Wang et al., 2014a; Yap et al., 2012). Therefore,
20 incorporation of a MF/UF filtration unit has been proposed to mitigate salinity build-up by
21 discharging the saline water as permeate but maintaining sludge volume (Holloway et al., 2015;
22 Wang et al., 2014b). Further, the RO concentrate disposal from seawater reverse osmosis
23 (SWRO) plant was not permitted due to high levels of total dissolved solids (TDS) and other
24 contaminants. In this scenario, OMBR could be a viable alternative to overcome RO

concentrate disposal problem. When SWRO brine is used as a draw solution, diluted RO water can be discharged to the ocean without significant issues.

Nitrification in a CAS systems performed by two different nitrifying bacteria communities occurs in two-step process. Firstly, ammonia oxidizing bacteria (AOB) oxidize ammonium to nitrite (nitrification, Equation 1) and secondly, nitrite oxidizing bacteria (NOB) convert nitrite to nitrate (nitrification, Equation 2). The nitrification process has the following stoichiometric equations (Wiesmann 1994).

During denitrification process the reduction of nitrate occurs through the conversion of nitrate to nitrogen gas (N₂) as stated in Equation 3, which is removed from the system achieving nitrogen removal from the wastewater.

Kimura and Watanabe (2005) proposed a baffled membrane bioreactor by inserting baffles into a conventional submerged bioreactor and creating alternating aerobic and anoxic conditions. This novel arrangement achieved efficient nitrification-denitrification without mixed liquor recirculation. Due to the continuous aeration, baffles inner zone was successfully sustained at oxic conditions, whereas the outer zone was remained at between oxic and anoxic conditions (Kimura and Watanabe, 2005). The baffled MBR could provide the advantage of a smaller footprint (e.g. no additional anoxic tank) because the baffle can eliminate mixing of the anoxic biomass and sludge recycle between the oxic and anoxic tanks (Kimura et al., 2007). Previous studies have examined OMBRs along with cellulose triacetate (CTA)-based FO membranes, whereas no previous studies have evaluated baffled OMBRs. This study evaluates the baffled OMBR performance using a thin film composite (TFC) polyamide (PA)-based FO membrane,

which has higher water flux, lower reverse salt transport and better resistance to hydrolysis and biological degradation than CTA-FO membrane (Hou et al., 2016). This study aimed to assess the efficacy of baffles in a hybrid OMBR-MF system for improved simultaneous nitrification-denitrification in single-stage reactor design. A TFC-FO membrane and MF membrane were used in a bioreactor to provide high SRT (i.e., high TN removal) and to minimise salinity build up, respectively. A laboratory-scale hybrid MF-OMBR system was operated under four different oxic-anoxic conditions. Changes in flux and salinity build-ups, TOC and TN removal, protein, and biopolymer quantification and MLSS in a bioreactor were recorded for analysing the treatment efficiency.

2. Materials and methods

2.1. FO and MF membrane characteristics

The flat-sheet TFC-PA membrane (Toray Chemical Korea Inc., South Korea) was employed in the present study. Table 1 shows the characteristics of the membranes. The membranes were oriented as AL-FS (active layer facing the feed solution) configuration. Before module preparation, membranes were stored in distilled water at 4 °C. The feed solution in all experiments was the OMBR mixed liquor. The submerged FO membrane module was custom designed and fabricated in stainless steel. The membrane module has a channel for a flow having a width of 11 cm, a length of 12 cm, and a depth of 0.5 cm. Mesh spacers were used on the DS side to provide additional support to the membrane and promote mixing of DS. The total effective FO membrane area was 264 cm². The PES-MF membrane, supplied by Uniqflux Membranes LLP (UNHF2012, Pune, India), had a nominal pore size of 0.33 µm and an effective surface area of 1000 cm².

2.2. Feed and draw solutions characteristics

The chemicals used in this study were reagent grade procured from Sigma Aldrich (Sydney, Australia). The feed to the OMBR-MF hybrid system was simulated sewage consisting of 300 mg/L glucose, 50 mg/L yeast, 15 mg/L KH_2PO_4 , 10 mg/L FeSO_4 , 60 mg/L $(\text{NH}_4)_2\text{SO}_4$, and 30 mg/L urea. The simulated sewage was prepared from stock solution providing 350 mg/L chemical oxygen demand (COD), 16 mg/L ammonium nitrogen ($\text{NH}_4\text{-N}$), 28 mg/L total nitrogen (TN) and 3.5 mg/L phosphate ($\text{PO}_4\text{-P}$). Thus, this composition best describes the secondary effluent concentration.

[Insert Table.1]

1.1 M NaCl (64 g/L) was dissolved into the deionized (DI) water to prepare DS. Osmotic pressure and diffusivity were obtained by OLI Stream Analyser 3.2 (OLI System Inc., Morris Plains, NJ, USA). Electrical conductivity and osmotic pressure were 91.26 mS/cm and 51.78 atm, respectively for 1.1 M NaCl. The use of simulated seawater brine as a draw solution (DS) is another novel aspect of the system.

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

This study was conducted using a laboratory-scale OMBR-MF hybrid system (Fig. 1). This lab-scale hybrid system is comprised of a synthetic feed water tank, a plexiglass bioreactor, a hollow fiber MF membranes, a flat sheet plate and frame forward osmosis membrane module, and a separate concentrated draw solution and a diluted draw solution tanks sitting on a digital weighing balance. The effective dimensions for plexiglass tank were 24.5 cm (L), 15.5 cm (W), 40 cm (H), which is equivalent to the working volume of 11.5 L. Plexiglass partitions (25 cm length) were installed on three inside walls to be located at a depth of 5 cm (above the bottom of the tank), so that 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 inward towards aerobic zone at approximately 30° angle in the bottom 3.5 cm length to avoid dead zone formation and to attain thorough

1 mixing of biomass (Fig. S1, Supplementary Information (SI)). When water level into the
2 bioreactor was assumed to be at the top of the baffles the estimated volume ratio of the outer
3 to the inner tank was about 2. The oxic and anoxic cycle time was adjusted by changing the
4 position of the level controller. The theory and working details of the baffled reactor are
5 discussed elsewhere (Kimura et al., 2007).

6 A plate-and-frame membrane module was designed and fabricated for the lab scale
7 experiments. Flat sheet commercial TFC FO membranes (Toray, Gwangju, Korea) were fixed
8 in-between two plates. FO membrane samples were suspended vertically and parallel to the
9 MF membrane module. This flat sheet membrane module was placed into the plexiglass tank
10 for osmotic dilution in a continuous mode of operation. The air diffuser was installed inside
11 the oxic chamber of the bioreactor for oxygen supply at 3 litres per minute (LPM) air-flow rate
12 to air-scouring both membranes.

13 [Insert Fig.1]

14 The seed sludge was collected from aerobic tank of the water recycling facility at Central Park,
15 Sydney, Australia. The sludge was acclimatized with simulated wastewater as a feed for 45
16 days at infinite SRT (except sample collection) prior to starting the experiments to achieve
17 steady performance as security measure. Initial biomass concentration was adjusted at 3.5 g/L
18 in the baffled OMBR-MF hybrid system operation. During acclimatization steady state reached
19 when more than 90% TOC removal efficiency was obtained consistently for week. The
20 OMBR-MF system was located in a temperature-controlled laboratory environment (22 ± 1
21 °C). During the bioreactor operation, the hydraulic retention time (HRT) and SRT were
22 adjusted to 32 h and 65 d, respectively. 1.1 M NaCl solution (DS) was used as simulated RO
23 brine.

The concentrated draw solution and diluted draw solution containers were each placed on a weighing balance (Adam PGL 15001, Perth, Australia) and connected to a data logger. The permeate from the bioreactor increased diluted draw solution weight with time. Thus, pure water flux was calculated based on the difference in weight between the diluted and concentrated DS. Diluted DS tank was emptied, and concentrated DS tank was top up with the fresh DS two times a daily. By changing the height of the level controller, the oxic-anoxic cycle time was adjusted. At the same time, the level controller was also connected in parallel to regulate a feed pump (peristaltic pump Longer WT600 2J, Hebei, China).

In this hybrid system operation, additional MF membrane was introduced and operated continuously, driven by a peristaltic pump (Longer BT100 2J, Hebei, China). To accomplish constant HRT of OMBR-MF hybrid system, MF permeates flux was changed manually by the change of FO water during the entire operation (i.e. 34 days). The salt accumulation regarding TDS and conductivity of the mixed liquor, permeate and DS were measured regularly using conductivity meter (HACH, Düsseldorf, Germany) and solution pH was measured with pH meter (HACH, Düsseldorf, Germany). The operating conditions are listed in Table 2. The operation protocol of OMBR-MF study is shown in (Fig. S3, Supplementary Information).

[Insert Table.2]

2.4 Analytical methods

2.4.1 Water quality parameters

TOC concentration was measured with a TOC analyser (Analytik Jena Multi N/C 3100, Jena, Germany). MLSS and mixed liquor volatile suspended solids (MLVSS) in the bioreactor were measured using APHA (2005) standard method. The concentration of dissolved oxygen (DO) was measured by using a DO meter (Vernier, USA). COD of wastewater was analysed according to standard methods (APHA, 2005). $\text{NH}_4\text{-N}$, TN, $\text{NO}_2\text{-N}$, $\text{NO}_3\text{-N}$, and $\text{PO}_4\text{-P}$ were

measured using Hach TNTplus™ reagent vials and by photometric method (Spectroquant Cell Test, NOVA 60, Merck, Bayswater, Australia).

2.4.2 SMP and EPS extraction and quantification

The measured quantity (50 mL) of the mixed liquor was sampled from the bioreactor for SMP and EPS analysis (Raunkjær et al., 1994). Then sludge sample was transferred into the centrifuge tube, and centrifuged at 3,000 rpm speed for half an hour. The clear fluid from the top was filtered through a 0.45 µm cellulose acetate filter which is referred to as SMP. The SMP in the filtrate was quantified on terms of protein and biopolymer concentration. The dense sludge settled in the bottom of the centrifuge tube were gently mixed with pH 7 phosphate buffer. Measured quantity of the Dowex (7.5 g/g VSS) a cation exchange resin was mixed followed by centrifugation for 2 h at 900 rpm to obtain EPS. EPS were then filtered through 1.2 µm syringe filter, and remaining cation resin and residues were discarded (Deng et al., 2014). The extracted SMP and EPS samples were analysed for protein and polysaccharide concentrations. For protein analysis, the modified Lowry method (Sigma, Australia) and for polysaccharide Anthrone-sulfuric acid method (Raunkjær et al., 1994) were used respectively.

2.4.3 Statistical analysis

Data from anoxic cycle time and TN removal were analysed using a one-way ANOVA. The statistical analyses were performed using SPSS software version 25.0 (SPSS Inc., Armonk, NY, USA).

3. Results and discussion

3.1 Flux performance and salt accumulation at different oxic-anoxic conditions

Fig. 2 shows variations in permeate flux with time for both FO and MF membranes and salinity increase rate in the OMBR-MF hybrid system. The MF membrane was started to operate at day 1 to mitigate salinity build up in the reactor. In the OMBR–MF during Run 1, initial FO water flux was 10.70 LMH but decreased to 6.80 LMH after 7 days of continuous operation.

At the same time salinity increased from 0.55 to 1.81 g/L in the reactor. In another OMBR run with TFC FO membrane (Wang et al., 2016), water flux decreased from 15.30 LMH to 5.0 LMH while the salinity increased up to 5 g/L in the bioreactor during the first week of operation, since the OMBR was operated without MF membrane. In the beginning of the Run 2, on day 8 both flux and salinity increased. This was due to the physical cleaning of the membrane that would have removed foulant on the membrane surface. In OMBR operation there is always trade-off between flux and salinity build up. So, as flux increased after physical cleaning salinity also increased thereby reducing the effective driving force for permeation and causing eventual flux decline. Run 2 operations showed a more stable flux varying between 8.75 - 6.80 LMH and somewhat stable salinity around 2.5 g/L in the bioreactor. Before commencing the subsequent runs under different oxic-anoxic cycle times, the FO membrane was physically cleaned with DI water followed by gentle cleaning with a sponge ball.

[Insert Fig.2]

The OMBR-MF operation was then resumed to examine the flux behaviour in the Runs 3 and 4 respectively. On day 16 after completion of Run 2 having performed physical cleaning, MF membrane was solely operated for 24 h to bring down salinity to initial TDS levels and then to resume operation at initial salinity. At the beginning of the Run 3, 8.71 LMH initial FO flux was observed which about 18.3% was lower than initial FO flux during Run 1. The FO flux varied between 8.71 – 6.06 LMH during Run 3 and a steady salinity build-up was observed from 0.32 - 1.60 g/L. Likewise in Run 4 flux varied between 7.62 - 6.19 LMH with salinity increasing to 2.4 g/L. MF membrane flux was manually adjusted in the range 1.05-2.10 LMH during Run 1 to 4 to maintain constant HRT which also helped in overall salinity build up alleviation in the reactor to a specific value (2.5 g/L).

As can be seen in Fig. 2, salinity increased in Run 1 and 3 with time as compared to Run 2 and 4 respectively. This is due to relatively higher FO flux and simultaneous reverse salt flux which

induced higher salinity to build up during Run 1 and 3 respectively. Again, this can be correlated to very fresh FO membrane in Run 1 and bleeding off excess salt from the reactor when commencing with Run 3 (Table 3). The reason for stable salinity level during Run 2 and 4 could be initial low FO flux as compared to Run 1 and 3, which can be attributed to relatively less reverse salt flux keeping salinity level stable. However, due to high salt accumulation from Run 1 and Run 3 actual salinity during Run 2 and Run 4 were higher than Run 1 and 3 respectively. As stated above, overall salinity during OMBR-MF operation was up to 2.5 g/L. Thus, the salinity build up during the operation of the OMBR-MF system was relatively low in comparison of earlier OMBR studies and hence did not adversely affected biological activity in the reactor. Other studies reported 5.5 g/L to 25 g/L salinity build up during the 30 d OMBR operation employing NaCl DS (Luo et al., 2016; Wang et al., 2014a; Zhang et al., 2012). Further, the results obtained here are comparable with a stable mixed liquor conductivity of approximately 5 mS/cm, which was observed during OMBR operation with continuous MF extraction in a previous study (Luo et al., 2015).

From Fig 2, it is evident that the flux decline was relatively high in for Runs 1 and 3. The FO flux decline could be related to the salt accumulation (as indicated by rapid TDS increase) and membrane fouling due to the accumulation of MLSS on the membrane surface when submerged in the bioreactor. Wang et al. (2014a) also reported the FO flux decline during OMBR operation. However, incorporation of MF membrane in the MF-OMBR system helped to maintain higher FO flux as compared to other OMBR studies. Wang et al. (2014b) had also reported that by incorporation of MF membrane could mitigate OMBR flux decline achieving 2.75 times higher permeate water flux than that in normal OMBR operation. Compared to Runs 1 and 3, the flux decline was not significantly high for Runs 2 and 4, which is likely due to low initial flux.

3.2 Total organic carbon removal

The OMBR-MF system was operated over 33 d applying four different conditions (Runs 1- 4). Biological process performance of the baffled OMBR-MF hybrid system was evaluated concerning the removal of carbonaceous organics and nutrients, biomass production, and biological activity. TOC removal variation during Run 1 to Run 4 in reactor supernatant, MF permeate and FO permeate were 86.1-92.3%, 89.0-93.9% and 98.3-98.6%, respectively (Fig. S4). The OMBR FO channel was characterized by a very high TOC removal efficiency (98%) during all four experimental runs. TOC removal obtained in this study agrees with other TFC-FO membrane bioreactor studies due to high rejection by the membrane in combination with bacterial biodegradation (Hu et al., 2017; Wang et al., 2014a). In this study, TOC removal in the bioreactor is shown to be affected by the oxic-anoxic cycle times; 86% TOC removal was observed in Run 1, 88% in Run 2, on average 92% Run 3 and 4, respectively. TOC concentrations in the MF permeate were observed to steadily decrease from 10.3 to 5.9 mg/L throughout all runs. It is believed that the increase in MF membrane rejection can be attributed to the better sludge acclimatization (heterotrophic bacteria proliferation) under continuous operation of bioreactor as well as pore blocking and fouling of MF membrane (Chang et al., 2002).

3.3 Nitrogen removal

In this baffled OMBR-MF system, nitrification is carried out in the aerobic chamber when the MLSS level is above the top of the inserted baffles (Fig. S2 supplementary data) the autotrophic nitrifying bacteria oxidize ammonia to nitrate (Equation 1 and 2). On the other hand, denitrification occurs in the outer anoxic chamber when the liquid level is low, and the heterotrophic denitrifiers release nitrogen gas by nitrate ion reduction (Equation 3) (Belli et al., 2015; Xu et al., 2014). The duration of the oxic-anoxic condition would have a great impact on the treatment performance especially regarding nitrogen removal.

Fig. 3 shows the TN concentrations in the influent, bioreactor supernatant, MF permeates and FO permeate as well as the removal efficiencies at different operating periods. As shown in Fig. 3, significant differences between TN concentrations in the influent (27.50 mg/L) and treated water in the reactor (6.50-10.10 mg/L) was found during the different oxic-anoxic cycle times tested. In this baffled OMBR study higher overall TN removal (62.80-71.20) can be related to denitrification occurring in the outer zone of the hybrid baffled MF-OMBR. The TN removal observed in the baffled MF-OMBR was due to the co-existence of heterotrophic and autotrophic micro-organisms in the bioreactor (Kimura and Watanabe, 2005). The average effluent TN concentration decreased with the decrease of the anoxic time (Run 1-3). It is likely that at constant HRT, as the anoxic cycle time reduced, more nitrates circulated to the anoxic zone, where the denitrifying bacteria could have removed nitrate by denitrification and denitrification efficiency was increased, thus leading to an improved nitrogen removal performance.

[Insert Fig.3]

The nitrogen removal efficiency of the OMBR operated with short anoxic cycle times (e.g. 1.5 h, Run 3, 1.0 h Run 4) was higher 76.1 and 71.2% than that with the longer anoxic cycle times (e.g. 2.5 h Run 1, 2.0 h Run 2). Further, in the effluents of all the four different operational Runs, on an average 0.1 mg/L of $\text{NO}_2\text{-N}$ was observed. The very low $\text{NO}_2\text{-N}$ concentration in the effluent and mostly $\text{NO}_3\text{-N}$ presence in the treated effluent suggested that nitrification was almost perfectly completed (data not shown). This can be attributed to the 65 d longer SRT in the bioreactor was favourable for the enrichment of nitrifying bacteria and nitrification could not have been the rate-determining step in this process (Wu et al., 2013). Consequently, in OMBR-MF hybrid system, the degree of the denitrification in the anoxic environment decides TN removal efficiency (Ahn et al., 2003). In the present study, the treatment performance of the baffled MF-OMBR was influenced by the duration of oxic-anoxic cycle time. The TN

removal varied from 63 to 76% at different oxic-anoxic cycle time without addition of external carbon source.

However, specifically in Run 4 (0.5-1.0 h oxic-anoxic cycle time), TN removal was low as compared to Run 3 (0.5-1.5 h oxic-anoxic cycle time). Due to short anoxic time in Run 4, it was believed that mixed liquor must have circulated more frequently between interior oxic zone and exterior anoxic zone in such a manner that dissolved oxygen could not be depressed enough to support sufficient denitrification as compared to Run 3 (Kimura et al., 2007). Nah et al. (2000) previously reported that when the oxic-anoxic cycle time was shortened low, TN removal efficiency was observed indicating that length of the anoxic period may not be enough for complete denitrification. It has been reported that high DO concentration in the anoxic chamber would stoichiometrically compete for the available substrate in the feed as well as kinetically influence the denitrifying bacteria (Tan and Ng, 2008). Plósz et al. (2003) also demonstrated that oxygen entering a bioreactor affects denitrification metabolically. In Run 3, best TN removal efficiency was achieved. So, 0.5-1.5 h oxic-anoxic cycle time may be considered as optimum. This cycle time led to sufficient diffusional limitation of DO. This favourable condition might have obtained satisfactory removal of available nitrate that transferred from the aerobic zone. In addition, TN removal values of the four test runs at different anoxic cycle time studied using ANOVA. We observed that the significance value which is below 0.05 and, therefore, there is a statistically significant difference in the TN removal and different anoxic cycle time studied.

The efficiency creating an anoxic environment in the baffled OMBR-MF system is demonstrated by DO measurements in the anoxic zone. Fig. 4 shows the DO profile in the outer zone of the baffles, roughly in the middle of the anoxic zone about 12 cm down from the tip of the baffle. The measured values are shown in Fig. 4 were obtained on day 4, 16, 22 and 33 when good removal of nitrogen was observed, representing each Run 1 to 4 respectively.

The average DO concentrations during anoxic cycle for Run 1 to 4 varied between 0.01 to 0.31 mg-O₂/L, while measured DO during the oxic cycle varied from 2.10 to 6.02 mg-O₂/L during Run 1 to 4, respectively. Outer zone of the baffled reactor functioned as an anoxic reactor for a long time (corresponding to different oxic-anoxic cycle time), consequently better denitrification accomplished. The obtained experimental results also confirmed efficient TN removal for such SND process in a single reactor. In a baffled OMBR-MF the feeding process during the anoxic phase in an outer zone of the baffled reactor accelerated denitrified activity, expediting easy nitrate removal and subsequently improved total nitrogen removal (Belli et al., 2015).

[Insert Fig.4]

Further, influent carbon to total nitrogen ratio plays a vital role in the nitrogen removal efficiency of the SND process. In this study, COD/TN ratio was maintained around 12 which is very much in line with the other findings that higher COD/TN ratio is favourable to obtain improved nitrogen removal efficiency (Fu et al., 2009; Meng et al., 2008). Besides, in the SND process almost neutral pH has been reported. Usually, the pH variations depend on the extent of the SND phenomena. In this study, pH variation was always observed within neutral pH range from 6.9 to 7.42. Therefore, daily pH adjustment was not essential thus saved chemical cost, due to the characteristics of the baffled reactor process configurations. Other reports also suggested that denitrification process beneficially raises pH in the anoxic zone by giving back half of the alkalinity to fulfil nitrification requirement in the oxic zone (Ge et al., 2010).

In conventional biological nutrient removal (BNR) process, the removal of nitrogen occurs by using anoxic and oxic reactors in tandem. On the other hand, in baffled OMBR simultaneous nitrification and denitrification take place in a single reactor. Although the removal of nitrogen in OMBR is not higher than the conventional BNR but the performance of baffled OMBR is

comparable to the performances of conventional BNR system reported in the literature (Ahn et al., 2003; Patel et al., 2005; Ueda and Hata, 1999). As reported by Ahn et al. (2003) when evaluating the performance of sequencing anoxic/anaerobic membrane bioreactor (SAM) and the modified Luzack-Ettinger (MLE) MBR processes, overall TN removal efficiency was around 60% and 67.4%, respectively. Another MBR pilot-scale study by Ueda and Hata (1999) reported 79% total nitrogen removal. In above-mentioned MBR studies, an additional anoxic tank and sludge recirculation configuration were deployed. Patel et al. (2005) reported 77% TN removal in a modified membrane bioreactor. However, the authors further reported that single-stage MBRs are close to the modified MBR process in terms of nitrification but compromised denitrification with high nitrate concentration in the effluent. Thus, based on above discussion it could be said that the performance of the baffled OMBR employed in the present study is comparable with the conventional nitrogen removal technologies.

3.4 Phosphorous removal

PO₄-P removal in the various stages of the baffled OMBR-MF system show removal of 57.1-63.1% in the reactor, 61.1-66.1% in the MF permeate and 97.9-98.3% in the FO permeate for all four runs (Fig. 5). A stable and effective removal of PO₄-P (> 98%) in FO processes is seen throughout the operating period. Under interchanging anoxic and oxic cycles, the PAOs release the orthophosphate in the anoxic/anaerobic part of the cycle and then accumulate a greater amount of orthophosphate in the oxic cycle time. Kundu et al. (2014) also examined phosphorous removal from sewage in lab-scale SBR employing two different configurations, viz. anoxic-oxic (A/O) and anoxic-oxic-anoxic (A/O/A). Wang et al. (2014a) reported that due to the very high forward osmosis membrane rejection very low phosphorous was observed in the permeate in OMBRs. Holloway et al. (2007) examined FO membrane performance and reported very high TP removal efficiency of 99%.

During Run 1 highest $\text{PO}_4\text{-P}$ removal was achieved in the bioreactor. This can be attributed to the prolonged anoxic cycle time and subsequently, very low DO concentration (0.17-0.02 mg/L) attained during Run 1. This might have released phosphorous during an anoxic cycle, followed by phosphorous uptake during an oxic cycle. Besides, regular sludge wasting might have achieved $\text{PO}_4\text{-P}$ removal in the bioreactor.

Further, previous MBR report suggests that longer SRT adversely affects the biological phosphorus removal capability of the bioreactor and it might be responsible for low sludge yield, limiting phosphate uptake by new cellular material (Cosenza et al., 2012). Also, in the baffled OMBR-MF system, possibly under longer anoxic cycle time in Run 1 and 2, denitrifying-polyphosphate-accumulating organisms (DPAO) might have contributed to denitrification of $\text{NO}_3\text{-N}$ using available substrate. Several studies have suggested that PAOs are more efficient in enhanced phosphorus uptake in oxic environments than anoxic, and phosphorus removal can be reduced by excessive anoxic phosphorus uptake (Brown et al., 2011). In addition, previous studies reported that when phosphorus is assimilated under anoxic conditions, P assimilation in oxic conditions decreases by up to 60% (Ekama and Wentzel, 1999).

[Insert Fig.5]

3.5 Biomass activity in baffled OMBR-MF system

Soluble and bound extracellular polymeric substances variation in the OMBR is presented in Fig. S5 when the baffled OMBR operated under different oxic-anoxic cycle times (Run 1-4). SMP analyses from the bioreactor show not much variation in protein content but carbohydrate concentrations being higher corresponding to longer anoxic cycles (e.g. Run 1 vs 2). SMP concentrations were generally found to be higher for the longer anoxic cycle times (e.g. 14.1 mg/L at 2.5 h, 11.2 mg/L at 2.0 h, 8.2 mg/L at 1.5 h, 9.4 mg/L at 1.9 h). Tan and Ng (2008)

1 have reported that membrane biofouling propensity significantly increases with high SMP
2 concentrations in MBR. The higher SMP and EPS concentration measured in Run 1 may
3 explain the higher membrane-fouling tendency observed.

4 MLSS and MLVSS were increased from 3.8 to 5.3 g/L and 3.3 to 4.6 g/L respectively during
5 the entire OMBR-MF study. Further, superior retention capacity of FO membrane can be
6 attributed to the increased sludge retention by membrane, thereby increasing SRT and less
7 sludge removal. Zhang et al. (2015) reported that longer SRT is favourable for less sludge
8 production. A similar trend during OMBR-MF operation was also observed in our study at 65
9 d SRT. The change in the MLVSS/MLSS ratio in the range 0.84-0.88 was observed in this
10 study. The sludge concentration and so its activity decrease has also been observed in another
11 OMBR study (Alturki et al., 2010).

12 However, the literature on the membrane fouling during denitrification is rather sparse. Kim
13 and Nakhla (2010) found that denitrifying phosphorous accumulating organisms (DPAO)
14 could consume the storage intracellular carbon during the endogenous denitrification, and thus
15 result in formations of SMP and EPS. In present study, bound EPS was found varying from
16 29.9 to 38.1 mg/gVSS in increasing order from Run 1 to 4. This can be ascribed to increase in
17 biomass concentration with time. A 32.3 mg/g VSS bEPS was measured at 90 d SRT in aerobic
18 MBR study (Zhang et al., 2015) which was less than present SND study. However, in the
19 present study, bEPS were higher in Run 2 and 4 as compared to Run 1 and 3. This can also be
20 attributed to relatively high TDS concentration (salinity) in the mixed liquor. Wang et al.
21 (2014a) found higher bEPS in regular OMBR due to the excessive salinity build-up which was
22 found approximately ten times higher than hybrid MF-OMBR. In this study, MF membrane
23 incorporation helped to alleviate the salinity build up thereby reduced EPS release.

24 4. Future research perspectives for baffled OMBR

In this study, the OMBR-MF hybrid process is proposed for municipal wastewater treatment, with the aim of higher removal of total nitrogen. Conventional nitrogen removal technologies are less cost effective as these processes require larger space and chemical and energy consumption is also high. Therefore, MBR technologies with total nitrogen removal and wastewater reclamation in a single reactor are more attractive as an alternative due to smaller footprint. The OMBR can successfully be employed for water reclamation from various resources, including municipal and industrial wastewaters. However, the FO operation has some limitation such as reverse salt diffusion of draw solute and draw solution regeneration requirement. Also, reverse salt diffusion of draw solute often has a negative effect on membrane fouling. Seawater reverse osmosis brine or fertilizer DS can be employed as a DS and after osmosis dilution diluted SWRO brine can be safely discharged to the sea or fertilizer DS can be utilised for fertigation after suitable dilution. Therefore, SND can be achieved for total nitrogen removal in single OMBR employing baffles.

Nevertheless, there are still some challenges and scopes for future work that exists for this novel baffled OMBR-MF hybrid process, and therefore, the following recommendations are suggested:

1. There is also scope to assess draw solutes incorporating surfactants in baffled OMBR-MF hybrid system. The performance can be evaluated in terms of water flux, salinity, and nutrient removal as compared to simple FO process (Chekli et al., 2018).
2. Since microbial consortia play a vital role in biodegradation of organics, it would be interesting to address microbial community dynamics in an oxic and anoxic zone in the baffled bioreactor to elucidate its impact on nutrient and possibly on most recently studied organic micropollutants (OMPs) removal (Pathak et al., 2018).
3. Nanotechnology may also offer unique opportunities to combat biofouling so, new membranes with less fouling potential need to be explored, such as fabricated from

nanomaterials or carbon nanotubes (CNT). Further, it is essential to characterise fouling on the FO membrane in the long run without disturbing continuous operation and to devise robust automatic antifouling and cleaning strategy. Moreover, ongoing research efforts to make outer selective hollow fiber FO membrane modules capable of producing around 14 LMH initial flux showing more perspectives in wastewater treatment. Compared to inner selective hollow fiber FO membranes outer selective hollow fiber membrane is more promising in terms of higher sustained flux and less fouling propensity. However, both of those types of FO hollow fiber membrane reports are scarce and scope exists to scale –up and commercialise FO membranes (Tran et al., 2019).

5. Conclusions

This study investigated TOC and nutrient removal in a baffled OMBR-MF system under four different oxic-anoxic conditions. In a reactor 0.5-1.5 h oxic-anoxic conditions (Run 3) has achieved 92% TOC, 76% TN and 63% phosphate removal. Combining MF membrane effectively maintained salinity build-up (<2.5 g/L) in the reactor. Further, an innovative OMBR achieved FO water flux in the range 10.70 LMH to 6.80 LMH during the first two test runs. After physical cleaning 8.71 LMH initial FO flux was observed which about 18.3% lower than initial flux. In the end (completion of Run 4) 6.19 LMH flux was attained and 2.4 g/L salinity build up was noticed. High SRT (65 d) favoured nitrifying bacteria to proliferate, and denitrification was the rate-limiting step for SND process. SMP data showed that anoxic cycle time had pronounced effect on membrane biofouling so it is important to set operating conditions in order to prevent fouling. In the SND process almost neutral pH has been reported and daily pH adjustment was not essential thus saved chemical cost, due to the characteristics of the baffled reactor process configurations. MLSS increased to less than 6 g/L that can be attributed to weak fouling of FO membrane, and merely physical cleaning had contributed to

1 higher flux recovery after two weeks of operation. Thus, baffled OMBR-MF system can
2 become a viable option for complete treatment of domestic wastewater.

3 **Acknowledgements**

4 This study is funded by the Bhutan Trust Fund for Environmental Conservation (BT FEC)
5 [Project Grant No. MB0167Y16]. PhD candidate Nirenkumar Pathak would like to
6 acknowledge scholarship support from Commonwealth of Australia under Research Training
7 Program (RTP).

8 **Competing interests:** The authors have declared that no competing interests exist.

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Fig. 1. Schematic diagram of the laboratory-scale baffled OMBR-MF hybrid system. 1. Feed tank; 2. Concentrated draw solution; 3. Diluted draw solution; 4. Oxic zone; 5. Anoxic zone; 6. FO membrane cell; 7. MF membrane; 8. Baffles; 9. Air diffuser; 10. Water level controller; 11. Pump; 12. Pressure gauge; 13. Weighing balance with data logger; 14. MF permeate.

Fig. 2. Flux and salinity variation at different oxidic-anoxic conditions.

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Fig. 3. Total Nitrogen (TN) removal in OMBR-MF hybrid system.

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Fig. 4. Dissolved oxygen variation in OMBR-MF system during the oxic-anoxic cycle time.

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Fig. 5. Phosphate removal in OMBR-MF hybrid system.

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Table 1. Characteristics of FO and MF membranes.

Characteristics	FO	MF
Membrane area (m ²)	0.0264	0.1
Material	TFC PA*	PES*
Geometry	Flat sheet	Hollow fibre
A	1.29 *	NA
B	0.57	NA
S (μm)	358	NA
Outer diameter (mm)	NA	1.0
Module working length (cm)	NA	15.5
Pore size (μm)	NA	0.33
No. of fiber in one membrane module	NA	210

2 *TFC PA: Thin film composite polyamide, A water permeability coefficient, (LMH/bar), B is
3 salt permeability coefficient, (kg/m²/h); S is Structural parameter, PES: Polyether sulphone,
4 LMH: Litres per square meter per hour, NA: Not applicable.

5

Table 2. Operating conditions in the OMBR-MF hybrid system.

Parameters	Description
FO Membrane orientation	FO mode (AL-FS)*
Reactor volume	11.5 L
Draw solution (DS) concentration	1.1 M NaCl
DS Flowrate	10 ml/min @5.8 rpm*
MLSS*	3 -5.5 g/L
Aerobic cycle time	0.5 h
Anaerobic cycle time	2.5, 2, 1.5 and 1 h
HRT*	32 h
FO-MF hybrid system	
SRT*	65 d
F/M* Ratio	0.1 d ⁻¹
Air Flow rate	3 LPM*

*AL-FS: active layer feed side, rpm: revolution per minute, MLSS: mixed liquor suspended solids, HRT: hydraulic retention time, SRT: sludge retention time, F/M: food to microorganism ratio, LPM: litres per minute.