High strength domestic wastewater treatment with submerged forward osmosis membrane bioreactor

Bilal Aftab, Sher Jamal Khan, Tahir Maqbool and Nicholas P. Hankins

ABSTRACT

Forward osmosis membranes are less prone to fouling with high rejection of salts, and the osmotic membrane bioreactor (OMBR) can be considered as an innovative membrane technology for wastewater treatment. In this study, a submerged OMBR having a cellulose triacetate membrane, with the active layer facing the feed solution configuration, was operated at different organic loading rates (OLRs), i.e., 0.4, 1.2 and 2.0 kg-COD/(m³·d) with chemical oxygen demand (COD) concentrations of 200 mg/L, 600 mg/L and 1,000 mg/L, respectively, to evaluate the performance on varying wastewater strengths. High organic content with sufficient amount of nutrients enhanced the biomass growth. High OLR caused more extrapolymeric substances production and less dewaterability. However, no significant differences in fouling trends and flux rates were observed among different OLR operational conditions.

Key words | draw solute, forward osmosis, membrane fouling, organic loading rate, osmotic membrane bioreactor

INTRODUCTION

The membrane bioreactor (MBR), a combination of biological degradation followed by membrane separation, is gaining attention in the wastewater treatment market. Some distinct advantages of the MBR over the activated sludge process include high-quality reclaimed water, smaller bioprocess footprint and less sludge production. However, membrane fouling is creating a hurdle in the widespread application of this technology (Cornelissen et al. 2008; Zhang et al. 2012). An innovative membrane technology, osmotic membrane bioreactor (OMBR), is an alternate solution to this problem and works on natural osmosis with less energy requirement. As compared to conventional submerged membranes, the forward osmosis (FO) membranes have less membrane fouling tendency because the suction pressure is replaced by osmotic pressure (Lay et al. 2010a; Wang et al. 2014a). OMBR with high hydraulic retention time (HRT) also produces high-quality treated water (Chen et al. 2014; Coday et al. 2014; Holloway et al. 2015; Wang et al. 2014a). In FO, the draw solution (DS) induces osmotic pressure difference between the feed solution (FS) and DS (Cath et al. 2006; Qin et al. 2010; Yen et al. 2010; Tang & Ng 2014). A suitable DS-aided OMBR can replace the conventional MBR coupled with nanofiltration or reverse osmosis process. Despite these advantages of OMBR over conventional MBR, the major drawbacks of this technology are lower membrane flux and reverse DS transport into the bioreactor, which can inhibit the biomass growth and affects the reactor performance (Qiu & Ting 2014; Tan et al. 2015). In this context, higher internal concentration polarization plays a major role in reducing water flux (McCutcheon et al. 2006; Zhao & Zou 2011). FO membranes also have a high-retention property that leads to solute in feed water accumulating in the bioreactor, which can have a negative effect on the microbial diversity (Qiu & Ting 2013). Researchers are trying to maintain less salinity in bioreactors by optimizing sludge retention time (SRT) (Wang et al. 2014a). Chen et al. (2014) tried to resolve this problem of reversed salt by periodically removing supernatant from the settled biomass. Wang et al. (2014b) installed a flat sheet micro-filtration membrane to extract saline water from biomass and to reduce reversed salt solute concentration in bioreactor with a submerged FO membrane. Suitability of OMBR for different wastewater concentrations with varying HRT and organic loading rate (OLR) is yet to
be evaluated. Owing to high HRT, OMBR can be efficient in treating high-strength wastewaters.

In this study, a laboratory-scale OMBR was operated at three different OLRs, i.e., OLR1 at 0.4 kg-COD/(m³·d), OLR2 at 1.2 kg-COD/(m³·d) and OLR3 at 2.0 kg-COD/(m³·d) (COD: chemical oxygen demand). Flux decline, organic and nutrients removal, extrapolymeric substances (EPS) production and variation in mixed liquor suspended solids (MLSS) were measured at constant SRT of 30 days.

MATERIALS AND METHODS

Experimental set-up and operation

Flat sheet cellulose triacetate FO membrane having effective surface area of 0.12 m² from Hydration Technology Innovations (USA) was submerged in a laboratory-scale OMBR with working volume of 4.5 L. The OMBR was operated under the active layer facing feed solution (AL-FS) as AL-FS causes less fouling compared with active layer facing draw solution (Wang et al. 2010). Operating conditions are shown in Table 1 and the recipe used for the preparation of synthetic wastewater for three different OLRs is reported in Table 2. Glucose was used as substrate for the biomass to evaluate the effects of variable strength wastewaters on FO-MBR performance. Synthetic wastewaters as FS with COD concentrations of 200 mg/L, 600 mg/L and 1,000 mg/L corresponding to OLR of 0.4 kg-COD/(m³·d) (OLR1), 1.2 kg-COD/(m³·d) (OLR2) and 2.0 kg-COD/(m³·d) (OLR3), respectively, were treated using OMBR. NaCl solution (0.5 M) was used as DS. DS was circulated inside the membrane module with optimized cross-velocity of 500 mL/min using a peristaltic pump (Masterflex, 77200-62, Cole Parmer, USA). To keep the conductivity constant, 5 M NaCl as concentrated DS was incrementally added to the diluted DS using a timer and peristaltic pump. Flux of the membrane was measured by the DS volume difference per minute using a data logger weighing balance (Shimadzu, UX6200H, Japan). The membrane module used was made of acrylic plastic (6 mm thickness) with nine baffles as shown in Figure 1. By using an air pump, dissolved oxygen level of 3–4 mg/L was maintained in the reactor for effective biomass growth and membrane scouring. After each filtration run, the membrane was backwashed for 2 hours using 0.5 M NaCl solution as FS and circulating deionized water as DS inside the module with the speed of 500 mL/min.

SRT was maintained at 30 days to evaluate the difference in the biomass growth in terms of MLSS and mixed liquor volatile suspended solids (MLVSS) at different OLRs. SRT maintenance was also helpful in reducing conductivity in the bioreactor as salinity affects the biomass growth and biodegradation performance.

Analytical methods

Acclimatized activated sludge from a semi-pilot-scale MBR treating synthetic wastewater was used as seed sludge for continuous OMBR operation.

EPS extraction and quantification

EPS play a major role in membrane fouling since they act like a glue and cause biofilm formation and floc agglomeration. Polysaccharides (PS) and protein (PN) are two main components of EPS. EPS can be found in three forms, i.e., (i) soluble EPS, (ii) loosely bound (LB) EPS and (iii) tightly bound (TB) EPS. Fifty millilitres of activated sludge sample was centrifuged at 4,000 rpm at 4 °C using a refrigerated centrifuge (Centurion K241R, UK). The supernatant was collected for the soluble EPS analyses and remaining
sludge pellets were suspended in phosphoric buffer solution, stirred for 1 hour and centrifuged for 15 minutes at 4°C. The supernatant containing LB-EPS was separated from the pellets for analyses. Lastly, pellets were again re-suspended in buffer solution along with cation-exchange resin, stirred for 1 hour, and after centrifugation the supernatant was separated for TB-EPS. Dubois and Lowery methods were used for the quantification of PS and PN, respectively.

**Treatment performance parameters**

COD, MLSS and MLVSS were measured according to *Standard Methods* (*APHA et al. 2005*), while the ammonium-nitrogen and phosphate-phosphorus were measured using spectrophotometric Hach methods. COD was not possible for permeate water due to its high salinity; the total organic carbon (TOC) of permeate was measured using a TOC analyzer (Analytik Jena, multi N/C 3100, Germany).

**RESULTS AND DISCUSSION**

**Flux performance and salt accumulation at different OLRs**

Figure 2 illustrates the flux and salt accumulation variations over operation time. Flux generally dropped from 5.0 LMH (litres per square metre per hour) to approximately 3 LMH and the conductivity of the bioreactor increased from 3 mS/cm to approximately 31 mS/cm within a cycle of 14 days. After each cycle, supernatant was removed from the biotank and filled with feed water as the conductivity...
dropped back to 3 mS/cm at the start of a new filtration cycle. Initial flux decline from 5 to 3 LMH was observed within approximately 3 days of operation under all OLR conditions, followed by a stabilized flux at 3 LMH. Membrane fouling was observed earlier during OLR3 within 7 days of operation as compared to OLR2 which exhibited fouling after 10 days and OLR1 after 12 days of operation.

The initial flux drop was due to the reverse salt accumulation in the bioreactor, which decreased the osmotic pressure difference between the feed and the DSs. During this initial filtration stage, colloidal and soluble organic material accumulate on the membrane surface, creating an additional surface to support the adsorption of macromolecules and resulting in the formation of a dynamic layer which acts as a barrier between the biomass and the membrane surface, preventing it from further fouling (Lee et al. 2001; Lesjean et al. 2005; Sharp & Escobar 2006). The stable osmotic pressure difference and development of the dynamic layer after 3 days of operation can be the main reasons for the stabilized flux. It can be comprehended from Figure 2 that approximately 98% of the flux was recovered (after day 14: flux = 4.9 LMH; and after day 28: flux = 4.8 LMH) by backwashing, which shows that the type of fouling in OMBR is mostly reversible.

As compared to conventional MBR, fouling in the OMBR was less affected by the change of OLR. Trussell et al. (2006) found that membrane fouling in an MBR increased 20-fold when the food-to-microorganism ratio was enhanced four times. Conductivity trend in the biotank is shown in Figure 3. Initially conductivity increased from 3 to 26 mS/cm within 3 days of operation, followed by stable conductivity after the formation of the dynamic membrane. Conductivity increased at 7, 10 and 14 days for OLR1, OLR2 and OLR3, respectively, corresponding to decrease in membrane flux. Cornelissen et al. (2011) found that back solute transport was different for different sludge concentrations used in an OMBR for treating wastewater.

**MLSS variation at different OLRs**

MLSS variation at OLR1, OLR2 and OLR3 is shown in Figure 4. For each OLR, acclimatized sludge was taken from a semi-pilot-scale MBR with MLSS concentration of 6–7 g/L. Initially a fall in MLSS concentration from 5.9 to 2.5 g/L was observed after 10 days of operation for the first run during OLR1 and then it became stable for another two runs afterward. For OLR2, less reduction of MLSS concentration was found as compared to OLR1, from 6.7 to 5.3 g/L, due to maintenance of a relatively higher organic matter content. But at OLR3 with a higher COD concentration, the MLSS concentration increased from 7 to 8 g/L after 16 days of operation and the condition was suitable for biomass growth.

**Phosphate concentration in permeate and biotank**

Phosphate removal performance of OMBR at different OLRs was examined and the results obtained are shown in Figure 5. At all OLRs, more than 98% phosphate was rejected by the FO membrane and similar trends were
found in previous studies (Holloway et al. 2007; Yu et al. 2011; Qiu & Ting 2014). Accumulation of phosphate in the biotank was observed after completion of each filtration run and approximately 47 mg/L, 39 mg/L, and 20 mg/L phosphate was found in the bioreactor during OLR1, OLR2 and OLR3, respectively. As the supernatant was removed after every run, the amount of phosphate in the biotank reverses to the inflow phosphate concentration. Qiu & Ting (2014) removed accumulated phosphorus by precipitating it by changing the pH in the biotank. Compared to OLR1 and OLR2, significant amount of phosphate was removed by the biomass during OLR3 showing less accumulation of phosphate, due to higher biomass activity requiring higher concentration of phosphate to fulfill microbial nutrient demand.

**COD and TOC removal efficiencies at different OLRs**

Figure 6 demonstrates that the TOC removals in the permeate and COD removals in the supernatant at OLR1, OLR2 and OLR3 with the inlet COD of 200 mg/L, 600 mg/L, and 1,000 mg/L, respectively. Above 98% TOC removal was found in the permeate by OMBR under the OLR conditions because of the non-porous structure of the FO membrane, capable of high TOC rejection (Holloway et al. 2007; Achilli et al. 2009). Similar to the phosphates, organics accumulation in the biotank was also observed: 60 mg-COD/L, 98 mg-COD/L and 162 mg-COD/L as soluble (filtered) COD was observed at OLR1, OLR2 and OLR3, respectively. The increase in organic content accumulation with increase in OLR may be due to relatively lower biodegradation capacity of biomass with increasing OLR.

**Ammonium-nitrogen removals at different OLRs**

Compared to TOC and phosphate-phosphorus, removal of ammonium-nitrogen was relatively low. Ammonium-nitrogen removal at OLR1, OLR2 and OLR3 with average inlet concentrations of 27.3 mg/L, 47.8 mg/L and 86.2 mg/L, respectively, are shown in Figure 7. Initially 88% of ammonium-nitrogen was removed by the OMBR during OLR1, which was subsequently decreased to 84% after 10 days of operation. During OLR2, ammonium-nitrogen removal was decreased from 90 to 86% within 10 days of operation. Similarly, during OLR3 ammonium-nitrogen removal decreased from 96 to 91% over 10 days. The drop in the removal rates was due to the effect of salinity on biomass as the salinity increased from 3 to 31 mS/cm during each run. Salinity was reduced by removing supernatant from the biotank after each filtration run, because high salinity inhibits the growth of nitrifiers responsible for nitrification. These results are in agreement with the findings of Lay et al. (2008b). They found that ammonium removal was greatly affected by the increase of salinity, and nitrifiers regained their potential to remove ammonium-nitrogen when salinity was reduced. As compared to conventional MBRs high...
removal of ammonium-nitrogen was achieved in the OMBR. Nitrifiers typically have very slow growth rates; however, OMBR with a prolonged HRT of 12 hours provided better conditions for nitrifier growth, hence better removal efficiencies.

EPS results are summarized in Table 3, which shows high soluble and bound EPS at OLR3 as compared to OLR1 and OLR2 with less influent COD. Results revealed that high

**Figure 5** Phosphate removal at (a) OLR1, (b) OLR2 and (c) OLR3.
organic loading caused more EPS production which caused biofilm formation and floc agglomeration, although the effect of EPS increase on membrane fouling in OMBR was less significant as compared to conventional MBR.

Dewaterability was measured in terms of capillary suction time (CST), which shows how quickly wasted sludge can be dewatered. CST values of $81.6 \pm 5.1$, $91.6 \pm 4.5$ and $120 \pm 15$ seconds were found during OLR$_1$, OLR$_2$ and OLR$_3$, respectively. OMBR sludge during OLR$_1$ was easily dewatered as compared to the sludge during OLR$_2$ and OLR$_3$ because of lower biomass concentration. Despite high EPS and less dewaterability, which are normally considered as demerits of activated sludge in relation to membrane fouling, OMBR can treat high-strength wastewater with less membrane fouling.

Figure 6 | COD and TOC removal at (a) OLR$_1$, (b) OLR$_2$ and (c) OLR$_3$. 

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CONCLUSION

The finding of this research study indicates that OMBR is a feasible technology even for high-strength wastewater at high OLR. No substantial effects of OLR on membrane fouling were found with the increase in OLR. Elevated MLSS concentration makes wasted sludge less dewatered and produces more EPS. The high-rejection property of the FO membrane accumulated the nutrients and salts in the biotank, considered as a drawback. By operating the OMBR system at high-strength wastewater, one can investigate the influence of short SRT in the range of 10–15 days, which would help in the decrease of salinity in the biotank.

REFERENCES


