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**Evaluating the effect of different draw solutes in a baffled osmotic
membrane bioreactor-microfiltration using optical coherence tomography
with real wastewater**

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Abbreviations

AL-FS	Active layer feed side	RWW	Real wastewater
bEPS	Bound extracellular polymeric substances	SMP	Soluble microbial products
COD	Chemical oxygen demand	SND	Simultaneous nitrification–denitrification
CTA	Cellulose triacetate	SOA	Ammonium sulfate
DO	Dissolved oxygen	SRSF	Specific reverse salt flux
DS	Draw solution	SRT	Sludge retention times
FO	Forward osmosis	TDS	Total dissolved solids
FOG	Fat, oil and grease	TFCPA	Thin film composite polyamide
HRT	Hydraulic retention time	TN	Total nitrogen
HTI	Hydration Technology Innovations	TOC	Total organic carbon
ICP	Internal concentration polarization	TSS	Total suspended solids
KAUST	King Abdullah University of Science and Technology	UF	Ultrafiltration
MBR	Membrane bioreactor	WWTP	Wastewater treatment plant
MF	Microfiltration		
MLSS	Mixed liquid suspended solids		
MLVSS	Mixed liquid volatile suspended solids		
OCT	Optical coherence tomography		
OMBR	Osmotic membrane bioreactor		
RSF	Reverse salt flux		

Abstract

This study investigated the performance of an integrated osmotic and microfiltration membrane bioreactor for real sewage employing baffles in the reactor. To study the biofouling development on forward osmosis membranes optical coherence tomography (OCT)

technique was employed. On-line monitoring of biofilm growth on a flat sheet cellulose triacetate forward osmosis (CTA-FO) membrane was conducted for 21 days. Further, the process performance was evaluated in terms of water flux, organic and nutrient removal, microbial activity in terms of soluble microbial products (SMP) and extracellular polymeric substance (EPS), and floc size. The measured biofouling layer thickness was in the order sodium chloride (NaCl) > ammonium sulfate (SOA) > potassium dihydrogen phosphate (KH_2PO_4). Very high organic removal (96.9 ± 0.8 %) and reasonably good nutrient removal efficiency (85.2 ± 1.6 % TN) was achieved. The sludge characteristics and biofouling layer thickness suggest that less EPS and higher floc size were the governing factors for less fouling.

Keywords: OMBR; optical coherence tomography (OCT); microfiltration (MF); biofouling; salinity build-up.

1. Introduction

Currently, wastewater is increasingly considered as a source of water, nutrients and energy rather than a waste (Wang et al., 2016b). Wastewater recycling and reuse in domestic, manufacturing and agriculture applications is therefore gaining increasing attention around the world (Pathak et al., 2017). In recent years, osmotic membrane bioreactors have gained more popularity for sewage treatment applications (Wang et al., 2016b). OMBR has many advantages such as excellent permeate water for reuse, low biofouling than conventional membrane processes and better energy efficiency. However, the salinity build-up in the reactor due to the reverse salt flux from the draw solution and the osmotic concentration of the feed solution can reduce the driving force for water transport (Nguyen et al., 2016; Pathak et al., 2017).

Nutrient removal in domestic wastewater treatment is one of the main goals of biological wastewater treatment since excess of nutrients in the final treated water may cause accelerated eutrophication (Randall et al., 1998). For removal of nitrogen, anoxic and aerobic zones are applied where the mixed liquor is circulated between the anoxic tank and the aerobic MBR tank. Kimura et al. (2007) proposed a baffled membrane bioreactor that can eliminate the energy needed for the mixed liquor circulation. In our previous work, Pathak et al. (2017) have successfully demonstrated the performance of a novel baffled osmotic membrane bioreactor in which baffles are inserted in the reaction tank and feed water is drawn through the FO membrane. By utilizing this method, both nitrification and denitrification can be promoted in a single reaction tank. As a result, sludge recirculation pump and anoxic tank mixer can be omitted.

However, in membrane applications fouling is considered as main concern for a wide application of OMBRs, shortening the membrane life, decreasing water production, and

increasing operating costs (Bell et al., 2016). FO membrane biofouling is a dynamic, and relatively slow process manifested as an attachment of self-organized bacterial cells to the membrane surface by gel-like, self-produced extracellular polymeric substances (EPS) (Inaba et al., 2017). EPS originated from biomass are the main components which contribute to biofouling and cause membrane flux decline over time (Wang et al., 2016b).

In order to operate conventional MBRs, hydraulic backwashing or relaxation can be applied. However, in OMBRs with FO membrane modules chemical cleaning or osmotic backwashing is feasible alternatives (Achilli et al., 2009). One of the major issue limiting membrane fouling control is the inadequate understanding of the basic mechanisms governing adhesion and deposition of foulants inside the pores as well on membrane surface. To assess and mitigate membrane biofouling, it is important to explore FO fouling mechanisms during OMBR applications and to develop competent control strategies (Inaba et al., 2017). Different tactics are proposed to study the biofilm architecture in the literature. The majority of techniques requires displacing the membrane from the reactor for sampling and those techniques are destructive. Hence, non-destructive direct observation of biofilm formation and development is vital (Fortunato & Leiknes, 2017; Inaba et al., 2017). Optical techniques for studying biofilms are gaining more attention because they are non-destructive and capable of non-contact and in situ measurements as compared to conventional destructive methods (Fortunato et al., 2016).

This paper proposes for the first time to evaluate the biofilm growth on FO membrane in the novel baffled OMBR-MF hybrid system treating real wastewater. On-line monitoring of biofouling growth and changes in morphology on a flat sheet CTA-FO membrane was performed non-destructively with optical coherence tomography (OCT), allowing an *in-situ* and real-time examination of the biofilm structure over 21 days.

Thus, the present study systematically investigate the complex fouling phenomenon of FO membranes in a baffled OMBR employing an OCT technique. Furthermore, the majority of FO fouling tests were performed with simulated wastewater and a few studies have been reported employing real sewage in OMBR membrane fouling examinations. In order to overcome this major shortcoming, real wastewater was used in order to assess its influence on membrane fouling. Moreover, other sludge characteristics like soluble microbial products (SMP), bound extracellular polymeric substances (bEPS) and floc size were measured employing three different draw solutes in an OMBR-MF hybrid system.

2. Materials and methods

2.1 FO and MF membrane characteristics

The HTI (Albany, OR, USA made cellulose triacetate (CTA) FO membranes was used in this study (Kim et al., 2017). FO membrane area was 0.0264 m^2 . The submerged FO membrane module was custom designed and fabricated using stainless steel (SS). The hollow-fiber micro-filtration (MF) membrane (Uniqflux Membranes LLP, India) was employed in this study. The MF membrane had 0.1 m^2 area and a nominal pore size of $0.33 \mu\text{m}$ respectively.

2.2 Feed solutions characteristics

Real wastewater (RWW) was sampled from the flow equalisation tank at the wastewater treatment plant (WWTP) of King Abdullah University of Science and Technology (KAUST, Thuwal, Saudi Arabia). The WWTP is a conventional AS-MBR (activated sludge- membrane bioreactor) designed to treat an average daily flow of $9,500 \text{ m}^3/\text{d}$ (Jumat et al., 2017). The characteristics of real sewage used in this study were: pH 7.45 ± 0.2 , TSS 65 ± 24 , TDS 645 ± 32 , FOG 9.8 ± 2.1 , COD 180 ± 29.5 , TN 14.7 ± 3.4 , $\text{NH}_4\text{-N}$ 12.6 ± 0.5 , $\text{PO}_4\text{-P}$ 3.1 ± 0.3 .

2.3 Draw solutions

The DS employed in this study were prepared by dissolving respective salts of three different chemicals sodium chloride (NaCl), ammonium sulphate (SOA) and potassium dihydrogen phosphate (KH_2PO_4) in deionized (DI) water. The chemicals used in this study were reagent grade purchased from Sigma-Aldrich, Saudi Arabia. Thermodynamic properties of DS were determined at temperature of 20 °C using OLI Stream. The osmotic pressure of 1 M NaCl, ammonium sulphate ($(\text{NH}_4)_2\text{SO}_4$) and KH_2PO_4 were 46.8, 46.1 and 36.5 atm respectively. The corresponding diffusivities were $1.32 \times 10^{-9} \text{ m}^2/\text{s}$, $1.14 \times 10^{-9} \text{ m}^2/\text{s}$ and $1.02 \times 10^{-9} \text{ m}^2/\text{s}$ for NaCl, ammonium sulphate ($(\text{NH}_4)_2\text{SO}_4$) and KH_2PO_4 respectively.

2.4 Baffled osmotic membrane bioreactor-microfiltration (OMBR-MF)

system and operation

2.4.1 Set-up and operation

The schematic of the lab-scale baffled OMBR-MF system used in this study is shown in **Fig. 1**. This baffled OMBR-MF system consisted of a feed tank, a plate-and-frame FO membrane cell submerged in a plexiglass bioreactor and a hollow fibre MF membrane module, a concentrated DS reservoir and a diluted DS reservoir. The effective volume of the bioreactor tank was 10.5 L (*i.e.* 24.5 (L) x 15.5 (W) x 40.0 (H) cm). On the three inside walls of the tank, plexiglass partition of 25 cm (height) was running from top to 5 cm above the bottom of the tank thus making hollow baffle box inside the tank with the size of 18.5 cm length, 12.5 cm width and 25 cm height. The details of baffled tank can be seen elsewhere (Pathak et al., 2017). The seed sludge was collected from the AS-MBR (activated sludge-membrane

bioreactor) wastewater treatment plant (WWTP) at KAUST (Thuwal, Saudi Arabia). The bioreactor was acclimatized for two weeks prior to adding into the baffled OMBR-MF system. Also, real wastewater (RWW) was employed to perform acclimatization and when evaluating different draw solute performances. Real wastewater was collected in clean containers (20 L) which were rinsed in the wastewater sample prior to sampling. Wastewater was transported from KAUST AS-MBR plant to KAUST laboratory using dedicated cart. Wastewater collection was performed twice a week and it was stored at 4 °C in cold storage. This acclimatization time for 15 days at infinite SRT (except sample collection) prior to starting the experiments might have helped achieve steady performance during test runs. During acclimatization steady state reached when more than 90 % TOC removal efficiency was obtained.

By employing the level controller, the oxic cycle time was set 0.5 h (level controller leg 0.5 cm above the baffle tip) and anoxic cycle time 1.5 h (level controller leg 1.5 cm below the baffle tip) can be adjusted accordingly. The hydraulic retention time (HRT) and solid retention time (SRT) of the OMBR-MF system was 34 h and 70 d respectively. The air diffuser was installed inside the oxic zone and 3 LPM airflow rate was maintained. The concept and operating details of the baffled reactor are discussed elsewhere (Kimura et al., 2007; Pathak et al., 2017). The continuous-flow experiments were conducted in active layer facing feed side (AL-FS) mode using the CTA-FO membrane.

2.4.2 In-situ biofilm monitoring

In order to measure *in-situ* biofouling growth on the FO membrane optical coherence tomography (OCT) technique was used for the OMBR-MF hybrid system. Thus FO membrane fouling characterization was facilitated without disturbing FO module from the reaction tank (Fortunato et al., 2016). Biofouling monitoring was performed twice a day for each DS for 7 days of operation. While performing OCT analysis, air scouring was stopped

for about 15 minutes to allow the sludge to settle down prior to image analysis. The fouling monitoring was performed with an OCT (Thorlabs GANYMEDE SD-OCT, Thorlabs, GmbH, Dachau, Germany) equipped with a 5 × telocentric scan lens (Thorlabs LSM 03BB). The OCT scan lens was mounted on a 3-axis motorized stage slide (Velmex) (Fortunato et al., 2016). The OCT scans had a resolution of (3500 x 1024 pixel) corresponding to 7.00 mm x 2.05 mm (width x depth). The images were cropped and filtered (Li et al., 2017). The fouling layer thickness was calculated using a customized MATLAB code (Fortunato et al., 2017).

2.5 Analytical methods

2.5.1 Water quality parameters

Total organic carbon (TOC) analysis was carried out with a TOC analyzer (TOC-V-CSH, Shimadzu, Japan). The MLSS and MLVSS in the OMBR were measured as per standard methods (AWWA, WEF (1998)). The concentration of dissolved oxygen was measured by using a DO meter (Vernier, USA). COD of real wastewater was analysed according to standard methods (AWWA, WEF (1998)). Hach TNTplus™ reagent vials and HACH® Spectrophotometer DR5000 used for NH₄-N, TN and PO₄-P analysis. Malvern Mastersizer, UK was used for mean particle size analysis of the sludge flocs.

2.5.2 EPS extraction and quantification

The formaldehyde-NaOH method was used to extract the extracellular polymeric substances from the sludge. The modified Lowry method (Sigma, Australia) and Anthrone-sulfuric acid method was used for protein and polysaccharide analysis respectively (Raunkjær et al., 1994). Standard curves of protein and polysaccharide were developed using various concentrations of Bovine Serum Albumin (BSA) and glucose, respectively.

3 Results and discussion

3.1 Process performance of OMBR system

Fig. 2 illustrates the FO and MF water fluxes as a function of time for each DS as well as the TDS concentration inside the bioreactor. Among the three different DS, NaCl produced the highest initial flux followed by SOA and KH_2PO_4 , respectively, at a constant molar concentration. A notable decrease in water flux was observed by the higher salinity (means low osmotic pressure difference) of the reactor mixed liquor in the presence of inorganic ions (Na, $\text{NH}_4\text{-N}$, $\text{PO}_4\text{-P}$) (Zou & He, 2016). The significant flux decline of NaCl and SOA than the KH_2PO_4 DS, which can be attributed to the higher initial flux and reverse solute flux specifically for NaCl DS, both contributing to decreased driving force and hence permeate flux declined. Thus, SRSF (defined as the ratio of reverse salt flux to water flux in FO process) of NaCl led to higher salt accumulation in the mixed liquor than the other two draw solutes. These results correlated well with previous findings (Kim et al., 2016; Wang et al., 2017; Wang et al., 2014a). In addition, salinity build up in the reactor mixed liquor could increase viscosity and decrease oxygen solubility then aggravates membrane fouling and flux decline (Lay et al., 2010). Xiao et al. (2011) noted that in OMBR operation the steady state is reached when the solutes entering into the bioreactor is balanced by the solutes discharge via waste sludge. Holloway et al. (2015b) reported that salt accumulation in OMBRs was modelled by different modelling studies. It was deduced that the steady state salt concentration in the osmotic membrane bioreactor would reach a constant value depending on the solids retention time (SRT).

However, an alternative strategy for minimizing salt concentrations in the bioreactor without changing the SRT would be to operate MF/UF membrane parallel to the FO membrane in the bioreactor. Wang et al. (2014b) and Holloway et al. (2015b) kept 10 d and 30 d single SRT in their MF/UF-OMBR study, respectively. In our study by incorporation of MF membrane

even at higher SRT of 70 d salinity build up was mitigated. This salinity level (2.5 g/L) does not affect biomass activity and sludge characteristics as reported by other studies (Nguyen et al., 2015). This situation in combination with other constant operating parameters (HRT, SRT, F/M ratio) might have facilitated to operate baffled OMBR-MF in reasonably equilibrium conditions within short time operations of 7 d employing different DS.

Other OMBR studies suggest that salinity concentration of > 2 g/L has adverse impact on biomass and specifically result in inhibition of ammonia-oxidizing bacteria (AOB) responsible for nitrification (Nguyen et al., 2015). In this study except NaCl DS (2.5 g/L) very less salinity build up for SOA (1.3 g/L) and KH_2PO_4 (1.2 g/L) DS was observed. Due to N and P nutrient content in SOA and KH_2PO_4 DS it is believed that it has favourable impact on the biomass evidenced by increase in floc size (section 3.5.2).

Wang et al. (2014a) also reported that flux quickly reduced from the initial value of about 9 LMH to the steady value of approximately 3.5 LMH during the first week operation of OMBR at 1 M NaCl DS. In another OMBR study, (Wang et al., 2014b) incorporated MF membrane to alleviate salinity build up and they achieved 2.5 times higher water flux as compared to OMBR study performed without MF membrane by Wang et al. (2014a). This indicates that it is feasible to maintain stable FO flux in the OMBR by adding a MF membrane under similar operating conditions (Zou & He, 2016). Incorporation of a MF module in this study helped reduce the salinity build up and maintain a constant hydraulic retention time (HRT), where the flux decline for NaCl is attributed to the accumulation of salt, increasing from 0.7 to 2.5 g/L within the bioreactor. In another study employing 1 M SOA as DS, a rapid flux decline from 6.9 to 2.5 LMH was observed in four days using a similar HTI CTA FO membrane without incorporation of a MF/UF membrane (Wang et al., 2017). In another recent OMBR study using fertilizer DS the water flux dropped sharply from 5.8 to 2.8 LMH within three days without combining MF/UF membrane (Zou & He,

2016). In contrast to the reports in previous studies, higher water flux was achieved in the present study from 7.8 to 5.5 LMH in the first three days working with 1 M SOA as DS. This was achieved by integrating a MF module that reduced salt accumulation in the osmotic reactor. Salinity build up was much lower for SOA (TDS 1.3 g/L) compared to NaCl (TDS 2.5 g/L). KH_2PO_4 fertilizer DS showed a lower initial flux (7 LMH) compared to the other two DS, and less salinity build up, thus a much lower flux decline was observed. In case of KH_2PO_4 DS, TDS increased up to 1.2 g/L. This TDS rise was two times higher than initial TDS value in the beginning of KH_2PO_4 DS run (day 18-25). Lower TDS rise with KH_2PO_4 DS can be related to the very low specific reverse salt flux (SRSF) (0.45 g/L of KH_2PO_4 DS). Also, initial lower flux with KH_2PO_4 DS required MF membrane to operate at relatively higher MF flux to maintain constant HRT as compared to other two DS test runs. MF membrane had withdrawn more permeate in order to operate OMBR-MF hybrid system at constant HRT and reduced salt concentration in bioreactor when KH_2PO_4 was employed as a DS.

It should be noted that after each run with different DS, MF membrane solely was operated for 48 h (**Fig. 2**). This allowed OMBR-MF hybrid system to resume with almost initial value of TDS concentration (0.6 g/L) when different DS was employed. The rapid flux decline observed with SOA, similar to that of NaCl, is therefore attributed to the biofilm formation on the FO membrane in the presence of nutrients. In order to investigate the effect of biofouling on the FO membrane, *in-situ* observation in real time was performed and is deliberated in following section.

3.2 *In situ* biofouling examination using OCT

The adhesion and growth of bacterial community on the membrane surface is a dynamic process. Most of the imaging techniques employed for the biomass characterization are destructive; enabling the imaging of the fouling only at the end of the operation. The OCT

allows acquiring information about the morphology online non-invasively. In this study, the biomass morphology formed on CTA-FO membrane was acquired *in-situ* through on-line OCT scanning during the entire period of operation over 21 d. The OCT scan at time 0, where no biomass was adhered on the membrane surface; the depth-resolved study allowed a cross-section imaging of the clean membrane. The thick porous layer of around 100 μm , with an embedded woven mesh, corresponds to the support layer, while the upper thin bright layer corresponds to the skin layer for the salt separation. Over time, accumulation of the fouling layer on the membrane surface led to a decrease of the signal intensity corresponding to the membrane.

Fig. 3 presents a set of 2D scans of the biomass development (bright orange) with time. The amount of biomass increased over time for all three different DS. As first, the *in-situ* observation enabled to observe and to depict the difference in biomass morphologies deposited on the membrane surface among the different DS. In the first 2 weeks of operation, where NaCl and SOA were used as DS, the presence of an irregular and loose structure corresponding to sludge flocs was observed above a thick, dense and compact fouling layer. In the last week, where KH_2PO_4 was used as DS, the amount of flocs deposited decreased due to the improvement of the sludge characteristics. This can be ascribed to the better acclimation of biomass with time as well as to the decrease in salinity with KH_2PO_4 as DS. The result was confirmed by the amount of EPS content in the reactor that corresponds to the higher flocs size. When KH_2PO_4 was used, a more stabilised water flux was observed compared to the other two DS. It has been noted that some microbes and EPS dislodged from membrane surface under steady water flux (Yuan et al., 2015). This could be a possible reason for thin biofouling layer observed for KH_2PO_4 . A more detailed discussion regarding floc size and its implications is presented in section 3.5.2. Beside the possibility of visualizing the biomass deposited in the system under continuous operation, the image

analysis performed on the OCT scans allowed the assessment and quantification of the fouling deposition over time for each DS. The MATLAB code employed for the thickness calculation enabled the selection of the fouling layer, without considering the flocs deposited above the layer. As shown in **Fig. 4**, using MATLAB coding, it is possible to calculate the thickness of the fouling layer over time for each DS. Moreover, the in-situ observation is capable of providing an insight into the fouling mechanism and the impact of the biomass on the membrane performances (Fortunato et al., 2017).

In comparison with the high pressure driven systems, the FO process is usually defined as a low fouling process due to the absence of hydraulic pressure and the lower flux conditions. However, in the case of the OMBR treating wastewater, the scenario is more complex. The flux decline over time is due to the increase of the hydraulic resistance and/or the concentration polarization effect. In this study, the thickness of the biomass deposited on the membrane in an OMBR treating real wastewater was correlated to the permeate flux. As shown in **Fig.4**, the flux decline corresponds to the increase of biomass thickness over time. By comparing the different DS, SOA and NaCl showed a similar trend in fouling thickness, which is in good agreement with the similar flux trend measured for the two DS. In the case of KH_2PO_4 that showed less flux decline over time and the highest flux at the end of the observation period, a lower amount of fouling was observed on the membrane surface compared with the other DS. After 7 days of operation, a fouling layer of 163 μm , 147 μm and 100 μm , and permeate flux of 3.7 LMH, 3.9 LMH and 5.5 LMH, were observed respectively for NaCl, SOA and KH_2PO_4 . Hence, the amount of biomass deposited on the membrane can be inversely correlated with the permeate flux. Operating the OMBR for 7 days with a different DS, the flux decrease measured is mainly affected by the thickness of the fouling layer formed on FO membrane. This implies that over time, generation of a cake-layer is the dominant fouling mechanism. In fact, the increment in biomass thickness also led

to an increase of water diffusion resistance from the feed to the draw side (Zhao et al., 2016).

In OMBR-MF operation with all three DS, the online non-destructive analysis performed in real time on information on the biofouling was linked to the variation in flux trend. This further explains the successful monitoring of the dynamic evolution of the biofilm.

In this study OCT technique was employed for the first time to assess real time fouling analysis. So, system was operated for relatively shorter time. Therefore this study is valuable as a preliminary test employing OCT technique since it revealed some interesting findings. Thus, having performed successful monitoring of the dynamic evolution of the biofilm in this study the future scope of the work should be to operate OMBR in longer run employing OCT technique to assess biofouling.

It should be noted that new FO membrane was substituted when changing the draw solution in this OMBR-MF study. However, in long-term operation, biofouling layer on FO membrane could largely affect system performance. Researchers suggested different cleaning techniques for FO fouling mitigation in long-term OMBR-MF operation. Physical cleaning of the FO membrane can be performed with deionised water and then by gentle scrubbing with sponge ball (Wang et al., 2017). When desired flux recovery cannot be obtained with physical cleaning then Holloway et al. (2015a) have proposed chemically enhanced osmotic backwashing. Backwashing is performed by circulating a very low strength acid or base solutions inside the draw channel. The main advantages of chemically enhanced osmotic backwashing include simplicity, less amount of cleaning solution as compared to external cleaning and direct exposure of cleaning agents at the membrane foulant interface. Moreover, in long-term OMBR-MF hybrid study to examine FO fouling employing OCT technique, we shall focus in detail on fouling issues, cost-effective cleaning strategy and flux recovery.

3.3 Organic matter and phosphorous removal

Regardless of the DS, the high retention characteristics of the FO membrane, longer HRT and SRT in OMBR-MF hybrid system and integration of MF to control salinity build-up all conditions showed an impressive removal in terms of TOC (**Fig. 5 (a)**). The average concentration of TOC in the NaCl, SOA and KH_2PO_4 DS was 1.3 ± 0.3 , 1.2 ± 0.4 and 1.2 ± 0.3 mg/L, respectively. Overall, 96.6 ± 0.5 , 97 ± 0.9 and 96.6 ± 0.6 % TOC was removed from the influent to the FO permeate with three DS respectively. In previous OMBR studies, an excellent TOC removal efficiency (96%) was reported in the bioreactor (Luo et al., 2015a; Wang et al., 2014a). Qiu and Ting (2013) also demonstrated more than 90% TOC removal in reactor mixed liquor during moderate salinity conditions. In this OMBR-MF hybrid study, similar high TOC removal performance by OMBR process was achieved.

The OMBR performance in terms of phosphate ($\text{PO}_4\text{-P}$) removal was evaluated for NaCl and SOA DS (**Fig. 5 (b)**). Due to the phosphate ion presence in KH_2PO_4 DS $\text{PO}_4\text{-P}$ accumulation took place in the bioreactor from RSF and hence the removal was not possible. It was anticipated that high rejection of FO membrane would allow substantial enrichment of phosphate within the bioreactor. An accumulation was reported in other OMBR studies and related to the effective FO membrane rejection based on a negatively charged and larger hydrated diameter of the ortho-phosphate ion (Luo et al., 2016; Luo et al., 2015b). With an average influent phosphate concentration of 2.9 ± 0.3 mg/L, the concentration in the mixed liquor was found to be < 1 mg/L with NaCl and SOA as DS (**Fig. 5 (b)**). However, the salinity build-up in the case of inorganic DS generates a detrimental condition for phosphorous accumulating organisms (PAOs). It is generally hypothesized that salt accumulation within the cells not only adversely affects the sensitivity of PAOs but also reduces the phosphate accumulating ability of microbes increasing the osmotic stress within the cells (Lay et al., 2010; Luo et al., 2016). A reasonable phosphate removal (67.4 %) with

NaCl and SOA under strict anoxic conditions was the result of higher biomass activity. In the baffled OMBR-MF hybrid system, mixed liquor suspension is cyclically exposed to aerobic and anoxic conditions, and enhanced biological phosphorus removal might have occurred to some extent. Further, oxic-anoxic cycle variation must have created a pseudo-anaerobic condition in the baffled reactor with extended anoxic cycle time. During aerobic condition, phosphorous uptake by microbial consortia could have happened. Thus, phosphorus removal could be achieved in similar way which was observed by (Kimura & Watanabe, 2005). In our study, possible phosphorous release in anoxic zone could also be linked to the existence of attached biomass on to the outer baffle wall as well to the inside wall of the reactor in anoxic chamber. During extended anoxic cycle (1.5 h), the diffusion limitation in the anoxic chamber might have created anaerobic condition into the biofilm that was adhered to the baffle wall. This situation had possibly stimulated phosphorous release. Thus, during aerobic cycle, phosphorus uptake followed by regular sludge discharge possibly managed phosphorus removal through biomass. Aftab et al. (2015) also reported a lower accumulation of phosphate due to increased microbial activity resulting in higher phosphate consumption. The FO process achieved more than 98 % removal of phosphate for each DS because of the high rejection of the FO membrane.

3.4 Total nitrogen removal

Nitrification of ammonia in domestic wastewater is first required before it can be further denitrified into molecular nitrogen. However, nitrification remains a limiting step in simultaneous nitrification-denitrification (SND) process due to high DO and longer SRT requirement (Kimura & Watanabe, 2005). Almost complete nitrification was achieved in this study can be attributed to sufficient oxygen availability and maintaining longer SRT of 70 d and hence limitation in nitrification was not observed. **Fig. 6 (a) and (b)** show the removal of $\text{NH}_4\text{-N}$ and TN for NaCl and KH_2PO_4 at the same molar concentration. For SOA, $\text{NH}_4\text{-N}$ and

TN accumulation was noticed during OMBR-MF operation (data not shown), which can be ascribed to the reverse salt transfer from draw solution to the mixed liquor. The TN level in the reactor increased considerably to 50 mg/L. The trend of $\text{NH}_4\text{-N}$ build-up was also observed for inorganic draw solutes in previous studies with high saline stress in the bio-tank (Aftab et al., 2015; Luo et al., 2015a; Qiu & Ting, 2013). Nitrification is a very sensitive process, as the nitrifying microbes involved in the process are slow growing and highly susceptible to conditions such as DO concentration and salt concentration (Lay et al., 2010). Compared to other OMBR studies, incorporating a MF membrane in the bioreactor mitigated excessive salinity build-up, and good nitrification was achieved with the high SRT and sufficient aeration as provided in the present study. Permeate quality showed more than 98% of $\text{NH}_4\text{-N}$ removal was achieved for both NaCl and KH_2PO_4 DS. In contrast to previous results for inorganic DS (Aftab et al., 2015; Holloway et al., 2015b; Siddique et al., 2017; Zou & He, 2016), a high removal of $\text{NH}_4\text{-N}$ within the bioreactor was observed (**Fig. 6 (a)**) throughout the operation with both the NaCl and KH_2PO_4 DS.

TN removal efficiencies in the reactor were 72.5 ± 1.7 % for NaCl and 74.6 ± 0.7 % for KH_2PO_4 at a low aeration rate of 2-3 L/min. With both DS more than 85 % total nitrogen removal in FO permeate was observed. Remarkable denitrification achieved in the baffled OMBR-MF hybrid system was possibly related to the NO_x ions exclusion by FO membrane which prolonged NO_x retention in the bioreactor, thus facilitating their removal during the anoxic cycle under very low DO (< 0.5 mg/L). A high initial C/N ratio in the reactor (around 11) also gave favourable conditions for an efficient simultaneous nitrification-denitrification (SND) process that might have occurred when the nitrification and denitrification rates were possibly in a stable equilibrium (Chiu et al., 2007). Dey (2010) also reported that sufficient electron donor is necessary for sustaining denitrification, where a C/N ratio above 10 is needed to have a significant effect on overall nitrogen. Holloway et al. (2014) in their UF-

MBR study reported that high nitrate concentrations were detected in the bioreactor. They deduced that elevated nitrate might have been either due to a carbon limitation or an insufficient hydraulic retention time (HRT) in the anoxic zone.

In the present work, the influent was fed to the anoxic zone of the reactor to supply COD to achieve a good C/N ratio for better denitrification to occur. In contrast, TN accumulation was found in the reactor with SOA, which is most likely due to the RSF of SOA from the DS to reactor mixed liquor. The nitrogen build-up occurred gradually in the reactor, increasing the ammonium-loading rates due to RSF from the DS side at constant COD loading. Subsequently, a lower C/N ratio in the reactor represents less carbon source available for biomass. Hence, the lower C/N ratio caused TN accumulation in the reactor.

Lay et al. (2010) noted that especially some microorganisms, like denitrifying bacteria, are apparently more sensitive under osmotic stress. In the current study, combining MF membranes with FO in the osmotic reactor helped alleviate salinity build up in the hybrid OMBR-MF system thereby providing favourable conditions for growth of denitrifying bacteria to achieve SND. In other studies carried out by Zajzon (2012) in laboratory-scale MBR system with higher SRT showed SND to become more significant, supporting the microenvironment theory outlined in section 3.5.2.

Finally, impact of different DS was investigated in a baffled OMBR-MF hybrid system. We could see that the fertilizer DS did not affect the bioreactor performance seriously.

Nevertheless, one week operation time was a little short with individual DS yet was sufficient to achieve carbonaceous and total nitrogen removal. It might be due to MF membrane incorporation which controlled salinity build up and the bacterial community structures were not significantly changed with different DS. Therefore, future studies will focus on long term operation of hybrid OMBR-MF system to investigate the impact of fertilizer DS on the

process performance via the analyses of bacterial community structures in order to understand how these DS influence the oxic-anoxic process (which is one of the most important parts in this hybrid system).

3.5 Biomass characteristics

Biomass characteristics play an important role in MBR operations, which governs the efficiency of the biological treatment as well as affects the membrane filtration processes (Lay et al., 2010; Siddique et al., 2017).

3.5.1 Organic foulants behaviour

It is well known that the SMP and EPS play important roles in membrane fouling. Understanding the characteristics of SMP and EPS in the hybrid OMBR-MF system is therefore helpful, to give a better insight into the membrane fouling mechanisms of FO processes (Qiu & Ting, 2014a). Both SMP and EPS are heterogeneous and generally comprise a range of biological origin substances like proteins, humic acid, triglycerides, carbohydrates and nucleic acid (Wang et al., 2016a). Proteins and carbohydrates are the dominant components found naturally in extracted SMP and EPS, and their respective concentrations were used to represent SMP. bEPS has been normalised as the sum of protein and carbohydrate concentrations to the sludge concentration in the reactor. The concentrations of initial SMP and bEPS in the reactor and at the end of experiments for each of the DS were measured in the OMBR-MF system. Salt accumulation within the bioreactor increases the osmotic stress on microbial activity, which translates to a noticeable increase in the SMP concentration, particularly with NaCl. These observations are consistent with previously reported results, where salt accumulation in the mixed liquor resulted in an increased SMP concentration in the OMBR (Luo et al., 2015b). The SMP are in direct contact with membrane surface thus protein and polysaccharide fraction of SMP has pronounced impact on flux profile as compared to EPS component (Johir et al., 2013). Initially, the

reactor had 30.4 mg/L SMP increasing to 61.6 mg/L over a week when NaCl was employed. Increased SMPs production, mainly as a carbohydrate fraction, facilitates the formation of a gel layer on the membrane surface that is not readily removed by physical cleaning (Di Bella et al., 2013). The increase in SMP is mainly due to plasmolysis and release of intracellular constituents as well as accumulation of intermediate and unmetabolised fractions and microbial produced polymers (Johir et al., 2013).

After each test run with SOA and KH_2PO_4 DS, the final SMP was observed to be lower than with NaCl; 52.6 mg/L and 51.4 mg/L, respectively. This is possibly because of less saline stress in comparison to NaCl DS. The decreases in SMP employing SOA and KH_2PO_4 can also be attributed to the different characteristic of the DS, both fertilizer DS having nutrient components as well as less RSF compared to NaCl DS. This provides a more favourable environment to the biomass resulting in less SMP released as the system becomes more stabilised.

At the same time, increase in the EPS concentration was also found with all three DS compared to the initial reactor EPS. However, the experiment with NaCl showed more EPS mass in comparison to the other two DS. After 7 days of operation with NaCl, bound EPS values increased due to increase in salt concentration, probably as a protective response by mixed bacterial population in sludge with high salinity (2.5 g/L). Reid et al. (2006) evaluated short term effect of salinity build up in pilot-scale experimental campaign. They reported immediate flux decline using NaCl and when chloride (Cl^-) concentration was raised to 4.5 g/L respectively. Also, both SMP and EPS significantly increased to 24.9 g/L and 86.7 g/kg MLSS respectively. In this study, at the end of experiments with SOA and KH_2PO_4 DS, measured EPS concentration was higher than initial reactor EPS content. However, measured EPS was less in the runs carried out under low salinity conditions (1.3 g/L for SOA and 1.2 g/L KH_2PO_4) than that with NaCl DS.

3.5.2 Floc size

The variation in mean floc size (d_{50}), evaluated in terms of volume of particles. The measured floc size for reactor mixed liquor was 22 μm . The floc size for all three DS was found to increase. In other OMBR studies, floc size was found to decrease with salinity build up in the reactor, for example, the mean floc size decreased slightly from 45 μm to 42 μm with an increase in salinity by NaCl from 0 to 2.5 g/L (Qiu & Ting, 2014b). This could be due to a decrease in the amount of filamentous bacteria in activated sludge flocs with increased salinity. These filamentous organisms are the backbone of sludge flocs, contributing to the development of larger flocs (Johir et al., 2013). Zhang et al. (2014) also reported that release of EPS from flocs, because of cell lysis, and the production of more SMP in the supernatant with the increased salinity, may also deteriorate the settling and flocculating properties of mixed liquor. On the contrary, Moussa et al. (2006) observed larger floc size and better sludge settling properties with increased salinity. In addition, average particle size improved from 100 μm to 200 μm at increased salinity conditions as compared to the fresh water environment. However, in this OMBR-MF study, increase of floc size was observed for NaCl DS. The slow and gradual increase of salinity over time may have provided time for the biomass to acclimatise with the changing environment thus not adversely affecting the floc size.

It was reported that higher EPSs content corresponded to higher floc size and stability (Lin et al., 2014). EPS increased with NaCl DS from 23.1 to 43.5 mg/g VSS, which is assumed to have affected the EPS content of sludge flocs and thus allowed floc size to increase from 22 to 64 μm when NaCl DS was employed. In this study the maximum salt load was 2.4 g/L with NaCl DS, increased gradually over one week and therefore did not adversely affect the floc size.

Many reports on conventional MBR studies show that membrane filtration realised with the small floc/particles in the activated sludge may have higher fouling potential than the larger sludge floc/particles (Sun et al., 2011). In an OMBR study Wang et al. (2014a) reported decrease in floc size from 137 μm to 116 μm with decreasing EPS in the reactor mixed liquor. This was due to increase in salinity up to 25 g/L. However till 15 d operation they found increased in EPS with increasing salinity (20 g/L). This does not adversely affect floc size which was reported to 137 μm very much similar to initial floc size 136 μm in the reactor. In our study the higher EPS contents in the reactor mixed liquor for all three DS compared to initial reactor mixed liquor EPS content should therefore result in greater floc size. Furthermore, higher SRT in the OMBR-MF reactor might have resulted in an increase in EPS in sludge samples that has been shown to enhance floc formation (Zajzon, 2012).

The floc size can also explain the SND phenomena observed in the baffled hybrid OMBR-MF system. SND occurs on account of DO concentration gradients within microbial flocs due to diffusional limitations. The nitrifiers will survive in the outer regions of the flocs where there is a higher DO concentration, while the denitrifiers will preferentially be active in the inner zones of the floc with very low DO concentrations (Münch et al., 1996). Results obtained in this study support the microenvironment theory and the mechanism of SND.

MLSS and MLVSS concentrations as a function of time was measured and it clearly revealed that MLSS and MLVSS concentration was not affected much with all three DS (data not shown). The MLVSS/MLSS ratio decreased from 0.84 to 0.77 when NaCl DS was used. Luo et al. (2015a) also reported that, due to increased osmotic stress on the feed side, a reduction in MLVSS/MLSS ratio was observed which decreased the concentration of active biomass in the sludge. In the current work, due to very low salinity build up in the reactor (due to MF membrane) no more decrease of biomass was observed for each inorganic draw solute. The

average MLVSS/MLSS ratio above 0.80 suggests an abundance of living microorganism, except slightly less (0.77) with NaCl DS.

4. Conclusions

This study investigated the flux performance and fouling behaviour of three different draw solutes in a hybrid OMBR-MF system. Biofouling behaviour was monitored using the OCT technique treating real sewage. The maximum biofilm thickness was observed with NaCl (163 μm) due to its higher specific RSF, resulting in elevated salinity (2.5 g/L). The lower fouling layer thickness measured for SOA and KH_2PO_4 DS is attributed to the lower salinity (1.2 g/L SOA and 1.2 g/L KH_2PO_4) and lower EPS concentrations. Incorporating a MF membrane helped control the salinity which also improved sludge characteristics such as EPS and enhanced flocculation characteristics.

Appendix A. Supplementary data

“E-supplementary data for this work can be found in e-version of this paper online”

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Figure Captions

Fig. 1. Schematic of lab scale baffled OMBR-MF combined system 1. Feed tank 2

Concentrated draw solution 3. Diluted draw solution 4. Oxidic zone 5. Anoxic zone 6

FO membrane 7 MF membrane 8 Level controller 9. Air diffuser 10. Weighing

balance with data logger 11 Pressure gauge 12 Pump 13 MF permeate 14 OCT probe

integrated with video camera 15 SD-OCT engine.

Fig. 2. Variation of FO and MF water flux and reactor salinity using different draw solutions

Fig. 3. Variation in biofilm morphology acquired by OCT scan with time

Fig. 4. Variation of fouling thickness and flux with time for (a) NaCl, (b) SOA and (c)

KH_2PO_4 DS

Fig. 5. (a) TOC (b) $\text{PO}_4\text{-P}$ removal in OMBR-MF hybrid system

Fig. 6 (a) $\text{NH}_4\text{-N}$ (b) TN removal in OMBR-MF hybrid system

Fig. 1.

Fig. 2.

Fig. 3.

Fig. 4

Fig. 5. (a) and (b)

Fig. 6 (a) and (b)

HIGHLIGHTS

- An OMBR-MF hybrid system performance was examined using CTA- FO membrane.
- A novel method to assess biofouling in OMBR is presented for the first time.
- *In-situ* and non-destructive examination of the biofilm acquired using OCT.
- Performance of three different draw solutions in OMBR-MF system were assessed.
- Simultaneous nitrification denitrification achieved with baffles in reactor.

Graphical abstract

Baffled OMBR-MF set-up with OCT Camera

Biofilm morphology acquired by OCT scan over time