A breakthrough dynamic-osmotic membrane bioreactor/nanofiltration hybrid system for real municipal wastewater treatment and reuse

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Abstract

This study designed a Dynamic-Osmotic membrane bioreactor/nanofiltration (OsMBR/NF) system for municipal wastewater treatment and reuse. Results indicated that a continuously rotating FO module with 60 RPM in Dynamic-OsMBR system could enhance shear stress and reduce cake layer of foulants, leading to higher flux (50%) compared to Traditional-OsMBR during a 40-operation day. A negligible specific reverse salt flux (0.059 G/L) and a water flux of 2.86 LMH were recorded when a mixture of 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114 functioned as draw solution (DS). It was found that the Dynamic-OsMBR/NF hybrid system could effectively remove pollutants (~98% COD, ~99% PO_4^{3-}—P, ~93% NH_4^+-N, > 99% suspended solids) from wastewater. In short, this developed system can be considered a breakthrough technology as it successfully minimizes membrane fouling by shear force, and achieves high water quality for reuse by two membrane- barriers.

Keyword: Dynamic OsMBR, rotating FO module, draw solution, membrane fouling, surfactant

1. Introduction

Recently, Osmotic membrane bioreactor (OsMBR) has increased for wastewater treatment, especially green and sustainable water treatment (Wang et al., 2020; Xu et al., 2021; Yao et al., 2020). Compared to traditional membrane bioreactor (MBR), OsMBR has several obvious advantages including less fouling tendency, high contaminant removal, and low energy consumption (Morrow et al., 2018). Furthermore the treatability of concentrate sludge facilitates recovery of minerals and resources from wastewater (Viet et al., 2021; Wang et al., 2020; Xu et al., 2021). Indeed, OsMBRs combine a biological treatment with a forward osmosis (FO) process in which water is extracted by osmosis pressure gradient (Cornelissen et al., 2011; Qiu & Ting, 2014). The FO process provides an additional barrier against a wide range of pollutants, hence increasing the overall contaminant removal of OsMBR (Gwak et al., 2015; Tan et al., 2015).

Unlike in MBR, the water is extracted by FO process without requiring a high hydraulic pressure; therefore, the specific energy consumption of wastewater treatment by OsMBRs (0.12 Kwh/m^3) can be much lower than that of MBR (0.37 Kwh/m^3) (Miyoshi et al., 2018) when the energy needed for the regeneration of the diluted DS is waived. For example, seawater or liquid fertilizer can be used as the DS for the OsMBR treatment of wastewater so that the diluted DS can be beneficially used without the requirement for recovering DS (Kim et al., 2016). In addition, the FO membrane in OsMBRs is much less subjected to membrane fouling than the MF membrane in MBRs. As a result, OsMBRs are compatible with concentrated high nutrients in wastewater and minerals-containing sludge, and these nutrients and minerals can be reused. One notable example is OsMBR treatment of nutrient-rich centrate to extract fresh water and struvite (i.e. $MgNH_4PO_4.6H_2O$) as fertilizer for agriculture to achieve green and sustainable wastewater treatment (Cong Nguyen et al., 2020; Yang et al., 2021).

Although membrane fouling affects OsMBRs to a much lower extent compared to traditional MBRs, it is still a major challenge for OsMBRs operation, particularly in the treatment of concentrated sludge and wastewater with complex compositions (Cong Nguyen et al., 2020; Gu et al., 2015; Hosseinzadeh et al., 2021). Concentrated sludge and complex wastewaters often contain large amounts of inorganic and organic colloids together with soluble compounds (Wang et al., 2020). These foulants have high affinity to the FO membrane; hence, they are likely to adsorb into the membrane pores or onto the membrane surface, thus raising the internal concentration polarization (ICP) as well as resistance to the transfer of water through the membrane (Nguyen et al., 2020). This inevitably leads to decline in the OsMBRs' water flux. Gu et al (2015) showed that water flux dropped to 74% after 16 days operation despite continuous biogas bubbling during an anaerobic OsMBR treatment of a low-strength wastewater. Foulant layers were also attributed to 45% water flux reduction in the OsMBR system (Nguyen et al., 2016b; Zhang et al., 2012).

Several techniques have been explored on how to resolve membrane fouling in OsMBRs (Kastl et al., 2021; Li et al., 2020; Nguyen et al., 2016a). For example, in our previous study, combining an OsMBR and a sponge-based moving bed (SMB) was assessed (Nguyen et al., 2016a), and it emerged that the hybrid SMB-OsMBR system could mitigate the cake layer of fouling in the membrane's active layer. Clearly, a sponge moving around the FO tube module in the bioreactor based on intensive aeration constitutes the main mechanism for cleaning the FO membrane during 90-day AnOsMBR operation. However, serious ICP occurred in the support layer due to the immobile FO module, leading to reduced osmotic pressure gradient, and consequently, water flux declined from 11.30 to 9.83 LMH (Nguyen et al., 2016a). Innovative designs of FO modules for OsMBR plays a critical role in reducing fouling and extending the membrane's usable life.

To date, several FO membrane module configurations have been devised and trialed for OsMBR, such as spiral wound, plate, tubular, and hollow fiber membrane modules (Ali et al., 2021). These membrane modules offer a high packing density that allows for a large active membrane surface area to be packed into a small volume of membrane module. This serves to increase the treatment capacity of OsMBRs. However, one common feature of these FO membrane modules is that they are static and subsequently prone to fouling and ICP (Chang et al., 2019; Nguyen et al., 2013). Intensive aeration, air bubbling, or fierce stirring of the sludge is required to reduce this fouling. These methods are energy-intensive and inevitably elevate the energy footprint of OsMBRs' treatment of wastewater.

In this study, a submerged Dynamic-OsMBR with an innovative rotating FO membrane module was devised for treating real municipal wastewater. Instead of using a static traditional FO membrane module, the OsMBR system in this study deployed a rotating FO membrane module for enhanced membrane fouling mitigation and reduced energy consumption. The FO membrane is fixed on central hollow axes and rotated by an electric motor to increase shear stress force to minimize the membrane fouling. The aim is to curtail membrane fouling without the need for air bubbling or fierce sludge stirring.

Another innovative feature of this work is a new DS for the FO process to reduce salt accumulating in the OsMBR system. The new DS consisted of polyethylene glycol tert-octylphenyl ether (Triton114), high charge ethylenediaminetetraacetic acid disodium salt (EDTA-2Na), and Na_2CO_3. EDTA-2Na presents a highly negatively charged component (i.e. $HEDTA^{3-}$) and $NaEDTA^{3-}$ complexion at high pH value, which can reduce reverse salt flux. Moreover, the ideal Van't Hoff index is high (i = 4) when dissolving EDTA-2Na in alkalinity solution, leading to high water flux. Triton114 is a surfactant with a low concentration of critical micelle (i.e. 0.2 mM) and a large structure. Given this structure, Triton114 forms a layer on the membrane surface, constricts the membrane pores, and hence minimizes reverse EDTA-2Na. Triton114 also enlarges the size of EDTA-2Na, which promotes its effective recovery during a nanofiltration (NF) regeneration of the diluted DS. On the other hand, Na_2CO_3 offers the new DS the ability to maintain its high pH so that EDTA-2Na exists at high charge components (i.e., $HEDTA^{3-}$ and $NaEDTA^{3-}$); thus, together with Triton114, it helps prevent the reverse diffusion of EDTA-2Na. To the best of our knowledge, the mixture of EDTA-2Na/Triton114/Na_2CO_3 has not yet been used as the DS of a Dynamic-OsMBR/NF system to simultaneously attain low salt accumulating, minimal membrane fouling, and a stable flux.

2. Materials and methods

2.1. The Dynamic-OsMBR/NF hybrid system

A lab-scale Dynamic-OsMBR/NF hybrid system was employed in this work (Figure 1). The Dynamic-OsMBR/NF system included a bioreactor, a plate-and-frame NF module, and a rotating tubular FO module. The FO membrane was immersed in the bioreactor, whereas the module of NF membrane was put outside the bioreactor for simultaneous freshwater extraction and regeneration of the diluted DS. The active volume of reactor was 9.0 L (with length×wight×height of 30×20×15 cm) and air diffusers installed at its bottom to maintain the dissolved oxygen content of 3 mg/L. The polyvinylidene fluoride hollow fiber microfiltration (MF) membrane module (Figure 1) with effective membrane area of 0.2 m^2 and pore size of 0.45 µm for periodic extraction of dissolved salts from the sludge to prevent the effects of salt accumulation on microbial activity [15]. Prior to the Dynamic-OsMBR/NF hybrid process, the acclimation of activated sludge was conducted with Dalat municipal wastewater (located in Vietnam) for 10 days until a mixed liquor suspended solids (MLSS) in the bioreactor was retained at 12 g/L.

Figure 1

The most notable component of the hybrid Dynamic-OsMBR/NF system was the rotating FO membrane module (Figure 1). The rotating FO module was made-up by a tube configuration and enfolded in FO flat sheet membranes from HTI, Albany, OR in USA. The rotating FO membrane module with membrane area of 251 cm^2 had a central tube placed inside a membrane tube, and the center tube was used to keep FO tubular module balance and rotate around its axis. Two rotary sealing bearings were set up at two heads of the FO tube and the rotation of the FO module around its axis was driven by an electromotor. The center tube was fixed and connected with a union at two heads. One head of the center tube was connected to the influent flow of DS and the other head was connected to the effluent flow of diluted DS. At the middle location, the center tube was carved out of small holes (diameter of 0.25 cm) so that DS could flow out and contact the FO membrane. Lab-scale NF membrane modules (Delrin Acetal Crossflow Cell, USA) were operated under cross-flow mode to recover diluted DS. The NF membrane (manufactured by Trisep) was made of polyamide with a molecular weight cut-off of 150 Da. The polyvinylidene fluoride hollow fiber MF membrane module was conducted in this study and supplied by Ray-E Creative Co., Ltd.,Taiwan.

During the Dynamic-OsMBR/NF hybrid process, municipal wastewater was fed into the reactor and the water level remained constant using a float-controller in the reactor. Hydraulic retention time (HRT) was in the range of 35-55 h, which was calculated by the water fluxes of Dynamic-OsMBR/NF and permeate fluxes of MF. The sludge retention time in Dynamic-OsMBR/NF was fixed of 20 days. OsMBR process were employed with the membrane orientation of active layer facing feed solution and rotation speed of tubular FO module was 60 RPM.The peristaltic pump was used to pump a mixed DS of 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114 into the rotating FO membrane module with flow rate of 1500 mL/min. Because of different osmotic pressure through the FO module, water extracted from the bioreactor and diluted the DS. The NF-TS80 membrane module was employed to regenerate diluted DS under a hydraulic pressure of 8 bar for its recovery and freshwater extraction in tandem.

During the Dynamic-OsMBR/NF hybrid process, the submerged MF module was introduced for periodically extracting the accumulated nutrients as well as phosphorus from the sludge. In Dynamic-OsMBR/NF system, features (e.g. total dissolved solids (TDS), PO_4^{3-}-P, NH_4^+-N, ,and chemical oxygen demand (COD)) of the waters in the bioreactor and the final permeate tank were analyzed every day to calculate the efficiency of the treatment process. The digital balances were used for recording the variable weight of the permeate NF tank and wastewater feed tank to determine the permeate fluxes of the Dynamic-OsMBR and the NF process.

2.2. The property draw solution and wastewater feed

Dalat municipal wastewater in Vietnam was used as the feed water to the hybrid Dynamic-OsMBR/NF system. Properties of the real wastewater are listed in Table 1. The DS was made by mixing 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114 in DI water, and then stirred for 1 day prior to the Dynamic-OsMBR experiments. Lab-grade EDTA–2Na.$2H_2O$ and $Na_2CO_3.H_2O$ were supplied from Sigma-Aldrich Co., Ltd., Germany, while Triton114 was purchased by Scharlau Chemie, Spain.

Table 1

2.3. Methods of calculation

The reverse salt flux J_s *(GMH)*, specific reverse salt flux J_s/J_w (G/L), and permeate water flux J_w (LMH) were determined based on previous researches (Nguyen et al., 2015b; Nguyen et al., 2016a; Nguyen et al., 2020).

The NH_4^+-N, PO_4^{3-}-P, COD and TDS rejection in Dynamic-OsMBR/NF hybrid system was calculated by the equation as follows:

$$R = (1-\frac{C_{eff}}{C_{inf}})\,.\,100\% \quad (1)$$

where: R was the removal efficiency; C_{inf} and C_{eff} were the concentrations of TDS, NH_4^+-N, PO_4^{3-}-P, COD, and SS at the influent and the effluent, respectively.

2.4. Analysis methods

The TDS and pH was measured daily by conductivity meter (Sension156, Hach, China) and pH meter (HI 9025, Hanna Instruments), respectively. The concentration of MLSS as well as COD was analyzed by Standard Methods (Eaton et al., 2005). The concentration of NH_4^+-N and PO_4^{3-}-P was measured by an ultraviolet–visible spectrophotometer (DR-4000, Hach, Japan). The osmometric model (Advanced Instruments, Inc., USA) measured the osmolality of DS. Vibro Viscometer (AD Company, Japan) was used to determine the viscosity of solutions. Energy dispersive X-ray spectroscopy (EDS) and scanning electron microscopy (SEM) supplied by JSM-5600, JEOL, Tokyo, Japan was used to observe membrane fouling. MINEQL+V.4.6 software (Sawyer et al., 2003) was used to predict the charge species of the multi-ion solutions as a function of pH.

3. Results and discussion

3.1 Effect of the different DSs on specific reverse salt and flux water flux in rotating FO process

Figure 2a illustrates the specific reverse salt fluxes (J_s/J_w) and water fluxes of five DSs with various Triton114 concentrations and a fixed EDTA-2Na and Na_2CO_3 concentration of 0.1 M. This outcome showed that water flux fell slightly from 3.12 LMH to 2.62 LMH when increasing the Triton concentration in the mixed DS from 0.1 mM to 1.2 mM. This was due to increased viscosity from 1.08 Cp to 2.21 Cp (Table 2), which altered the water diffusivity through the FO membrane. However, the J_s/J_w reduced significantly (from 0.330 G/L to 0.059 G/L) when raising Triton114 from 0.1 mM to 0.9 mM due to the second layer on the membrane surface. The explanation may be that hydrophobic interaction force between Triton114 tails and membrane surface likely constricted the pore of FO membrane, leading to raising retention of ions such as CO_3^{2-}, $NaEDTA^{3-}$ when the surfactant was coupled to mixed EDTA-2Na/Na_2CO_3 DS. This observations agrees with Chekli et al.(Chekli et al., 2018) and Hau et al. (Nguyen et al., 2015a), that decrease in the reverse salt diffusion is due to constricted membrane pores based on hydrophobic interactions between FO membrane surface and the surfactant tails.

Figure 2

Nevertheless, the Js/Jw rose from 0.059 G/L to 0.061 G/L when raising concentration of Triton114 in mixed DS from 0.9 mM to 1.2 mM. A huge gel layer of micelle forming in DS at 1.2 mM Triton114 caused a significant increase in viscosity (2.21 Cp). This prevented water diffusion and reduced water flux (average of 8.4% decrease). Hence, adding 0.9 mM Triton114 to mixed 0.1 M EDTA-2Na/0.1 M Na_2CO_3 DS was the best possible scenario to achieve lowest Js/Jw and high water flux.

Moreover, Figure 2b compared between 0.1 M pure NaCl; 0.1 M pure EDTA-2Na and mixed 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114 as DS. The result showed that 0.1 M EDTA-2Na with osmolarity of 225± 5 mOsm/Kg and viscosity of 1.19± 0.11 Cp achieved the highest water flux (Jw= 2.98 LMH), following by mixed 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114 with osmolarity of 237± 4 mOsm/Kg and viscosity of 1.89± 1.23 Cp (Jw= 2.86 LMH) and the lowest water flux of 0.1 M NaCl with osmolarity of 170 ± 3 mOsm/Kg and viscosity of 1.08 ± 0.07Cp (Jw=2.63 LMH). However mixed DS achieved the lowest Js/Jw (Js/Jw = 0.059 G/L) due to pH 8 (Figure. 3): (i) Mixed DS presented high charged $HEDTA^{3-}$ (79.5%) and macromolecular $NaEDTA^{3-}$-complexes (20.5%), which reduced reverse salt flux; (ii) Adding 0.9 mM surfactant to the mixed DS formed micelle led to reduced mobility of ions (Na^+, $EDTA^{3-}$); (iii) Adding 0.1 M Na_2CO_3 in mixed DS created a buffer in DS and maintained pH at 8, leading to low Js/Jw. Moreover, since the higher negatively charged NF-TS80 membrane was recorded at higher pH value (Verliefde, 2008), this increased electrostatic repulsion between ions ($HEDTA^{3-}$, $NaEDTA^{3-}$) in diluted DS. It negatively charged the NF-TS80 membrane and subsequently enhanced the efficiency of the NF recovery process.

Figure 3

Hence, among draw solutes, mixed 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114 is selected to perform the best for the Dynamic-OsMBR system. Furthemore, diluted mixed DS was effectively recovered by using NF technology under an 8-bar pressure with the high TDS rejection of 96%. The water flux of the NF-TS80 membrane was reported as 3.32 ± 0.45 LMH with TDS of permeate stream less than 500 mg/L.

3.2. Comparing the Dynamic and Traditional OsMBR systems

The above exploration of novel DS comprising mixed 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114 in the OsMBR system could reduce salt accumulation in the bioreactor significantly. However, for an OsMBR system in a real scenario application, a new

Dynamic – OsMBR module for minimizing membrane fouling is necessary for long-term viability. Figure 4a shows both Dynamic and Traditional - OsMBR water flux declined with time because of membrane fouling and salt accumulation inside the bioreactor in long-term operation. This phenomena approved by Ricci et al (2021), who demonstrated that external polarization concentration (ECP) and foulants contributed key mechanisms for declined water flux in long-term OsMBR operation. It caused an increasing filtration resistance and reduced the coefficient of mass transfer (Wang et al., 2016). However, the water flux of the Dynamic-OsMBR system dropped slightly while the Traditional-OsMBR system reduced quickly with time. The reason may be that continuous rotation of the FO tube module in a Dynamic-OsMBR system could prevent solids/foulants attaching to both sides of surfaces, thus minimizing membrane fouling and maintaining water flux (Figure.4b). Consequently, the Dynamic-OsMBR system retained stable water flux and achieved an average water flux higher in the Traditional-OsMBR system of approximately 50%. This was an interesting outcome of the Dynamic-OsMBR system.

Figure 4

In 40 days of operation, the OsMBR water flux changed and this followed 3 stages. In the first 5 days, both modules had water fluxes diminished quickly due to macromolecule adsorption. The water flux of Traditional-OsMBR fell from 2.59 to 1.96 LMH while Dynamic-OsMBR declined from 2.67 to 2.55 LMH. At the second stage (from day 5 to day 30), the decrease in water flux was because of a formed cake layer of biomass on the active layer surface of FO membrane. Figure. 4d shows that a thick cake layer of fouling formed on the active layer of FO tube membrane in the Traditional-OsMBR system, leading to significantly reduced water flux (Jw reduced from 1.96 to 0.98 LMH). This was due to fouling/activated sludge being easily attached to the immobile FO tube. Meanwhile, the continuously rotating FO module (60 RPM) in the Dynamic-OsMBR system created the high shear cross-flow and prevented the foulant layer forming on the membrane surface (Figure.

4b). This issue is explained that the average shear stress τ_a (Pa) depends on rotational speed ω (RPM) as follows (Limjeerajarus et al., 2018):

$$\tau_a = -1.9902 \times 10^{-9} \omega^3 + 4.3457 \times 10^{-6} \omega^2 + 0.00059221\omega - 0.017086 \quad (2)$$

Clearly, the higher rotational speed was applied in Dynamic-OsMBR system the higher shear stress was achieved for reducing membrane fouling. Hence, water flux of the Dynamic-OsMBR system reduced slightly (Jw reduced from 2.55 to 2.29 LMH), which was a good contribution of this study. In the third stage (from day 35 to day 40), the permeate flux of both OsMBR systems increased slightly at day 35 (average of 5% increase) due to using MF extraction of rich nutrient stream in the bioreactor. This in turn reduced the salt accumulation in the reactor from 2116 to 1671 mg/L.The rotating FO module in the Dynamic-OsMBR system could reduce the cake layer of fouling significantly during 40 days of operation, sop this represented a clear advantage compared to the Traditional-OsMBR system. Indeed, the water flux of the Dynamic-OsMBR system decreased by 13.5% only due to better membrane associated transport phenomena, while the water flux in the Traditional-OsMBR system decreased by 60.6% after 40 days of operation.

The surface SEM photos of the used membrane in Dynamic-OsMBR system, and Traditional-OsMBR system are depicted in Figure. 4c and 4d. For the used membrane in the former system, a thin cake layer of contaminants was observed on the surface as seen in Figure. 4c. However, a thick cake layer of foulants was observed on the membrane surface in the latter system (Figure. 4d). This observation could be explained by the rotating FO module creating a large shear force on the surface of the FO membrane. Consequently, any cake layer of fouling was significantly eliminated.

Moreover, EDS analysis results showed that as compared to the Dynamic-OsMBR system, an additional peak of Na appeared on the structural support layer of used membrane in the Traditional-OsMBR system, which caused concentration polarization phenomenon and led to rapid water flux decline. The reason is because free Na^{+} ions in mixed draw solution (EDTA-2Na and Na_2CO_3) faced the support side and attached to the used membrane in Traditional-OsMBR system due to attractive electrostatic force between negatively charged FO membrane and positively charged Na^{+}. Meanwhile, there is no peak of Na on the used membrane in the Dynamic-OsMBR system. The reason may be because the shear stress force based on the rotating FO module in the Dynamic-OsMBR system is larger than electrostatic attraction force between negative charge of FO membrane and positive charge of Na^{+}. Clearly, the continuously rotating FO module in Dynamic-OsMBR system could simultaneously reduce the ICP on the support layer and foulants on the active layer. This is a good exploration to maintain water flux during Dynamic-OsMBR operation process.

3.3 Dynamic-OsMBR/NF hybrid system's performance

Figure 5 illustrates that the Dynamic-OsMBR/NF rejected nearly 98.6% of PO_4^{3-}-P, that is better than what the MBR can do (typically 93%) (Guo et al., 2011).The high PO_4^{3-}-P rejection in the Dynamic-OsMBR/NF can be explained by 3 main reasons: (i) phosphorus accumulation in organisms; (ii) steric effect; and (iii) electrostatic repulsion. Firstly, since high biomass concentration (MLSS =12g/L) was used in the Dynamic-OsMBR/NF system without forming a cake layer of foulants, according to Guo et al. (Guo et al., 2011) the higher microorganism biomass accumulated more phosphorus, leading to superior phosphorus removal. Secondly, Kiriukhin & Collins (2002) recorded the large hydrated radius of PO_4^{3-} (0.34 nm) while the CTA-ES FO membrane had a small pore radius (average of 0.37 nm)), which resulted in reducing PO_4^{3-} through FO membrane due to the steric effect. Finally, pH of Dalat municipal wastewater was about 7.3, and Cartinella et al (2006) observed a negatively charged FO membrane at pH > 7, and the negatively charged PO_4^{3-} repulsed negatively charged FO membrane. In addition, the rejection of PO_4^{3-}-P tended to fall from day 1 to day 32 (dropped off from 99.81% to 99.62%) because of the high PO_4^{3-}-P accumulation causing high diffusion of PO_4^{3-}-P through the FO membrane and then to NF membrane. However, at day 35, the removal efficiency of PO_4^{3-}-P improved slightly due to using MF extraction of a nutrient-rich solution derived from the bioreactor.

Figure 5

Figure 5 indicates that high NH_4^+-N rejection (92.87%) was attained in the Dynamic-OsMBR/NF system; the average effluent concentration of NH_4^+-N was 3.57 mg/L. This result agreed with previous studies that the OsMBR process could retain high NH_4^+ efficiency (Achilli et al., 2009). As can be observed in Figure. 5, the Dynamic-OsMBR/NF hybrid system achieved a stable COD and SS removal during 40- operational days. The average removal of COD amounted to 98.35% when the effluent concentration of COD was 16.2 mg/L. The Dynamic-OsMBR/NF hybrid system achieved high SS removal of 99.68% due to two-barrier membranes (FO and NF membranes) with the SS effluent concentration of 0.9 mg/L. This corresponded to the SS concentration in influent DaLat municipal wastewater being 288.3 mg/L.

Figure 6 displays various salts accumulating in the reactor of the Dynamic-OsMBR/NF system with time. After 32 operational days of Dynamic-OsMBR/NF, the salinity in the reactor raised from 972 to 2116 mg/L. The reasons for increasing TDS were due to (i) the accumulation of salts (NH_4^+, PO_4^{3-}, Ca^{2+}) from the influent Dalat municipal wastewater; (ii) the salt leakage the DS into the bioreactor. Nevertheless, the salt accumulating in the reactor was low, which encouraged the normal development of microorganism communities because of the low Js/Jw ratio (<0.059 G/L). As shown in Figure.6, mixed 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114 as the novel DS in the Dynamic-OsMBR/NF achieved lower salt accumulating (<2200 mg/L) than that of using NaCl DS (>8000 mg/L) (Holloway et al., 2014). Indicated here is favorable DS for Dynamic-OsMBR/NF application to reduce the effects of salt accumulation on microbial communities. Moreover, novel Dynamic-OsMBR/NF was employed to not only minimize fouling but also generate high water quality for reuse. The concentration of TDS in the effluent stream changed within the 403-445 mg/L range during 40 operational days (Figure. 6). Overall, the Dynamic-OsMBR/NF hybrid system achieved a high level of contaminant removals from Dalat municipal wastewater. Clearly, the average concentrations of COD , NH_4^+-N, PO_4^{3-}-P, and TDS in the final permeate during the Dynamic-OsMBR/NF system were as low as 16.21 ± 0.58 mg/L, 3.57 ± 0.28 mg/L, 0.25 ± 0.03 mg/L, and 429 ± 6 mg/L, respectively, which was suitable for water reuse as compared to WHO standard (Agriculture et al., 1989).

Figure 6

4. Conclusion

In this work, a novel Dynamic-OsMBR/NF system was successfully devised to treat wastewater using a mixed 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114 as the suitable DS. Doing so helped to obtain a high water flux and a negligible Js/Jw (0.059 G/L).The Dynamic-OsMBR hybrid system could mitigate cake layer fouling significantly due to continuously rotating FO module leading to enhanced shear stress. Finally, the proposed system exhibited not only an excellent ability to reject PO_4^{3-}-P, COD and SS (>98%), and confirm good reuse of water. It also greatly diminished membrane fouling in sustained OsMBR operations.

Acknowledgements

This research is funded by Vietnam National Foundation for Science and Technology Development (NAFOSTED) under grant number 105.08-2017.311.

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

As Huu Hao Ngo, a co-author on this paper, is Editor of Bioresource Technology, he was blinded to this paper during review, and the paper was independently handled by Christian Larroche as editor.

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Highlights

- A new Dynamic-OsMBR/NF system was designed for wastewater treatment and reuse.
- Continuously rotating FO module in Dynamic-OsMBR removed cake layer of foulants.
- 50% more water flux was observed in Dynamic-OsMBR compared to Traditional-OsMBR.
- Specific reverse flux of mixed DS was 8-fold lower than when using NaCl only.
- Dynamic-OsMBR/NF hybrid system achieved high contaminant removal (almost > 98%).

42.

Figure Captions

Figure. 1. A 3D illustration of the lab scale Dynamic-OsMBR/NF hybrid system.

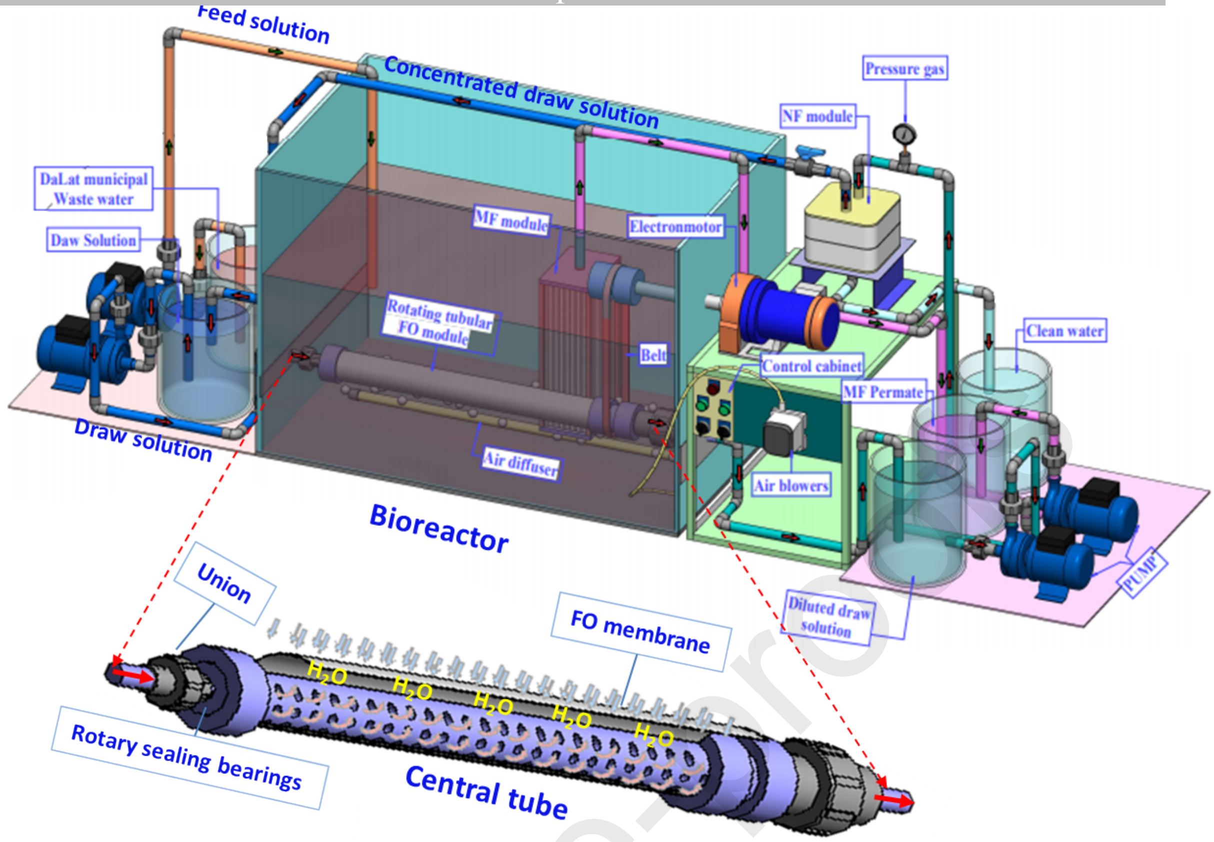
Figure. 2 **(a).** Various water flux, reverse salt flux and specific reverse salt flux of different DSs; **(b).** Comparison water flux and specific water flux of 3 kinds of DSs [Rotating tubular FO module: 60 RPM, FO membrane: CTA-ES, Membrane orientation: active layer facing feed solution, Feed solution: DI water; All experiments were run in 2 h and error bars were based on the standard deviation of three replicate tests after 2h].

Figure. 3. Multifunctional DS reduced specific reverse salt flux in FO process.

Figure. 4. (a).Comparison of permeate flux of Dynamic and Traditional OsMBR systems [Membrane orientation: active layer facing feed solution; MLSS: 12 g/L; Draw solution: Mixed 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114; Feed solution: Dalat municipal wastewater; Dynamic-OsMBR: 60 RPM; Traditional-OsMBR: 0 RPM]; (b). Reduced cake layer of foulants based on rotating FO module in Dynamic-OsMBR system; (c).SEM photos of active layers of used FO membrane in Dynamic-OsMBR system; (d) used FO membrane in Traditional-OsMBR system.

Figure. 5 Variations of organic and nutrient removal during Dynamic-OsMBR/NF system operation for wastewater treatment (Draw solution: Mixed 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114, Feed solution: Dalat municipal wastewater, Membrane orientation: active layer facing feed solution, MLSS: 12 g/L; rotating tubular FO module: 60 RPM).

Figure. 6. Variations in salt accumulation and TDS of effluent during Dynamic-OsMBR/NF system wastewater treatment (Draw solution: Mixed 0.1 M EDTA-2Na/0.1 M Na_2CO_3/0.9 mM Triton114; Feed solution: Dalat municipal wastewater, Membrane orientation: active layer facing feed solution, MLSS: 12 g/L; rotating FO module: 60 rpm).



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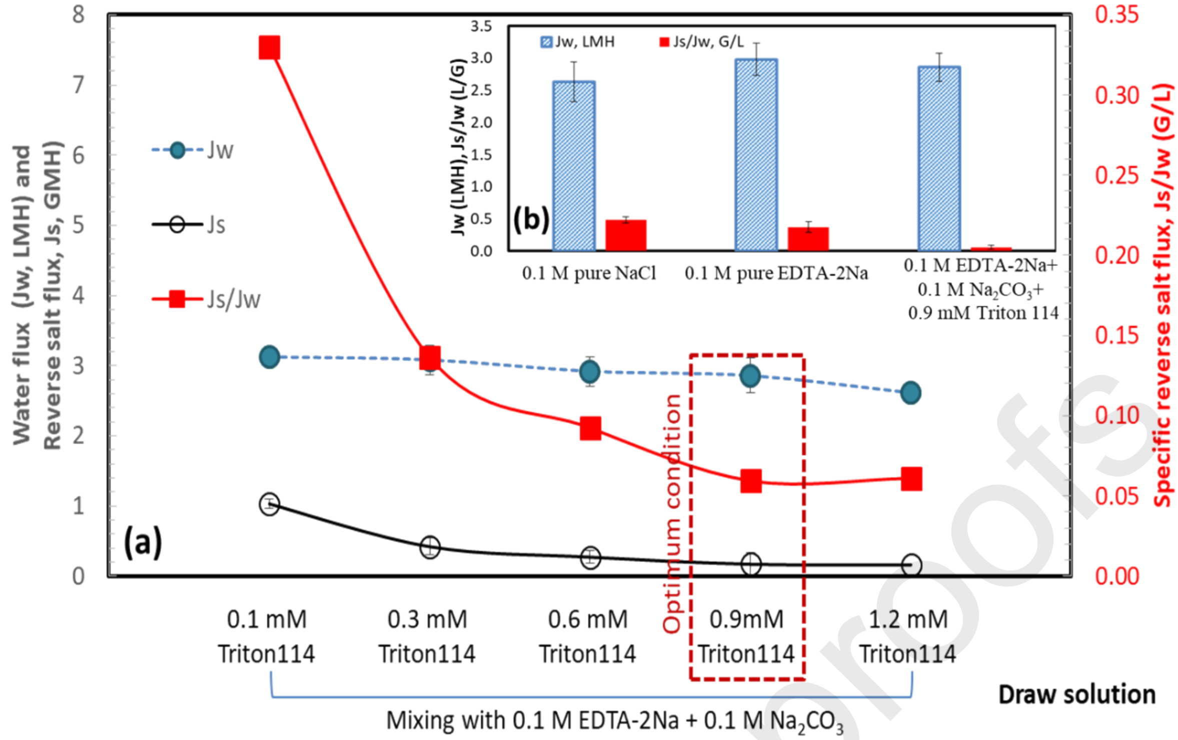

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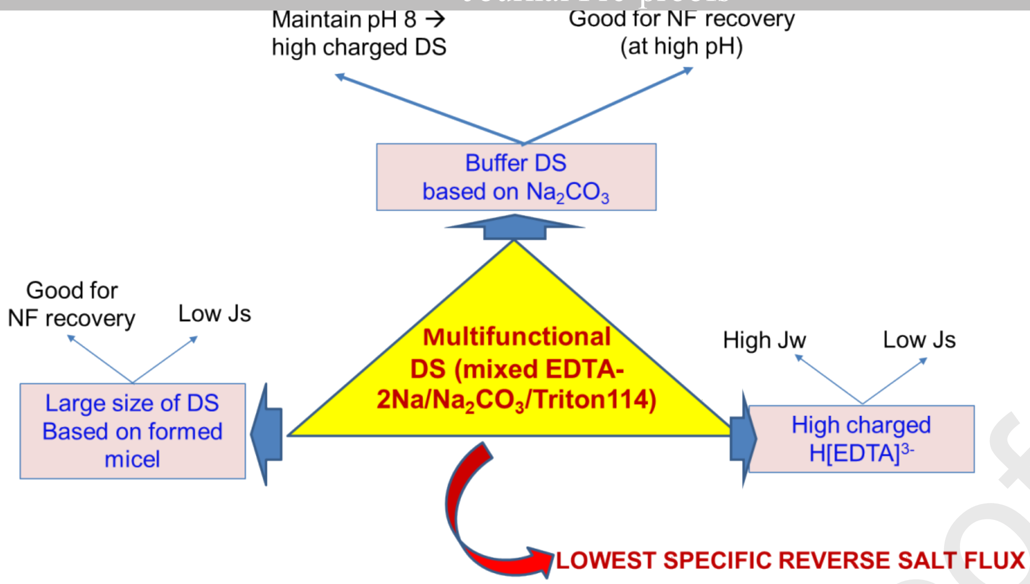
587 

588 **Figure. 3**. Multifunctional DS reduced specific reverse salt flux in FO process.

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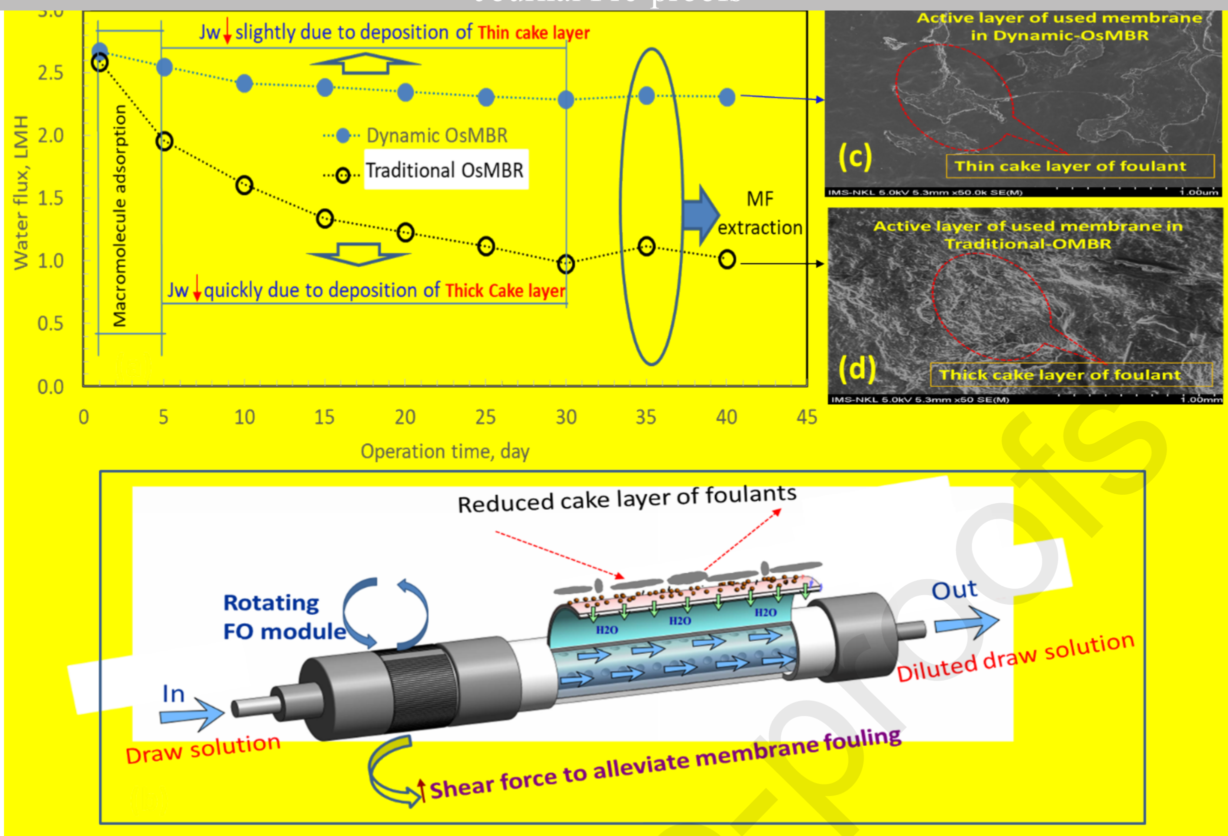

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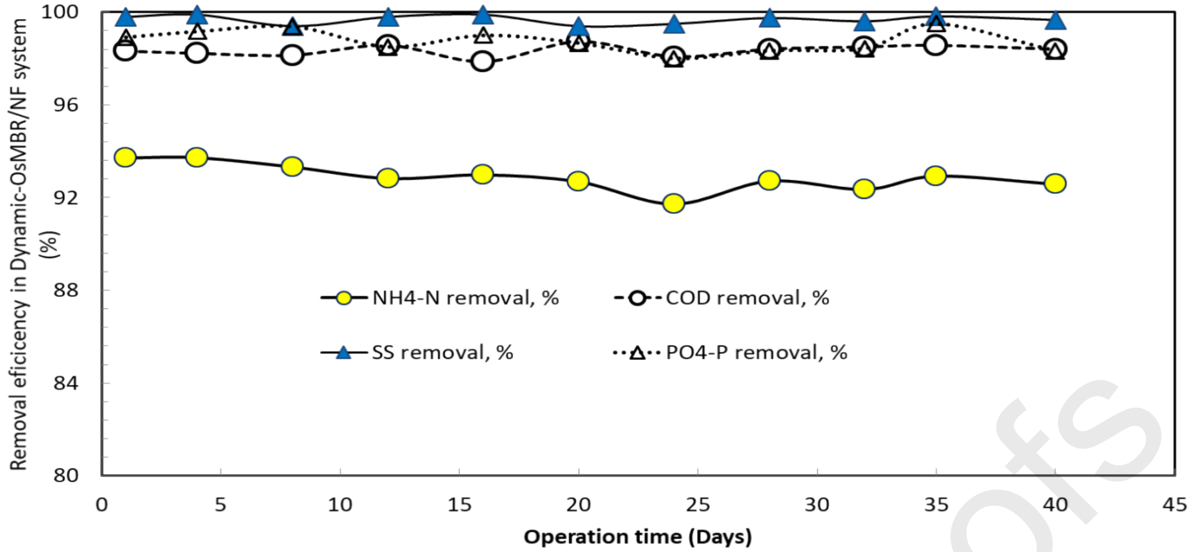

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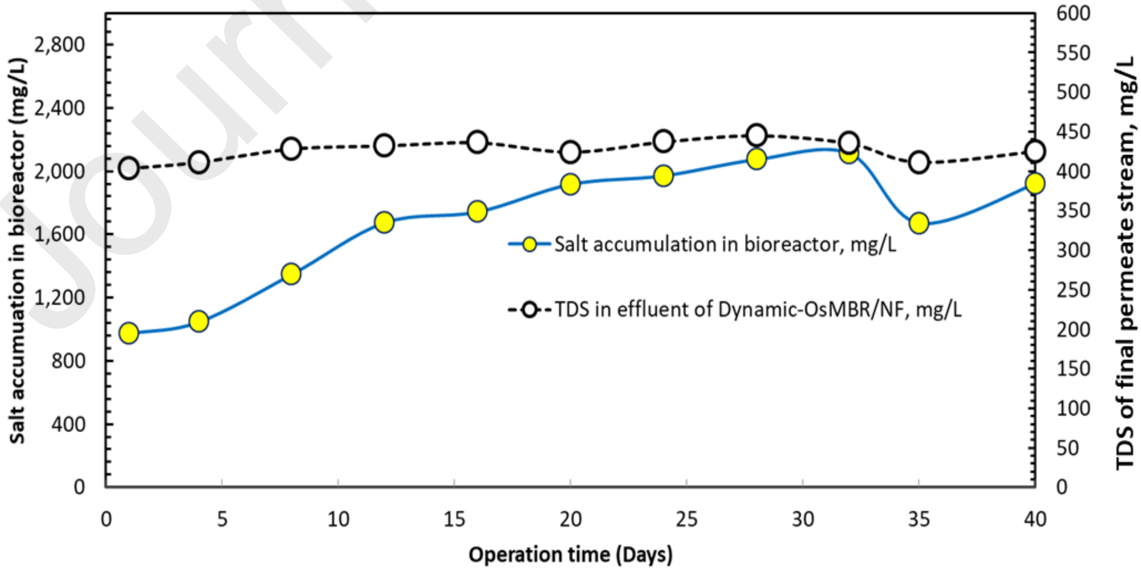

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Table captions

Table 1. Key characterictics of the municipal wastewater used as the feed to the Dynamic-OsMBR system

Table 2. Osmolarity,viscosity and pH of different DSs

Table 1. Key characterictics of the municipal wastewater used as the feed to the Dynamic-OsMBR system

Parameter	Unit	Value
COD	mg/L	880 ± 2
NH_4^+-N	mg/L	47.25 ± 0.75
PO_4^{3-}-P	mg/L	16.32 ± 0.18
SS	mg/L	280 ± 5
TDS	mg/L	825 ± 3
Mg^{2+}	mg/L	25.4 ± 0.3
pH	-	7.3 ± 0.5

Table 2. Osmolarity,viscosity and pH of different DSs

Draw solution		**Osmolarity, mOsm/Kg**	**Viscosity, Cp**	**pH**
0.1 M pure NaCl		170 ± 3	1.08 ± 0.07	6.3± 0.2
0.1 M pure EDTA-2Na		225± 5	1.19± 0.11	4.7± 0.1
0.1 M EDTA-2Na/0.1 M Na_2CO_3 mixed with	0.1 mM Triton114	238± 3	1.25± 0.14	8.0± 0.2
	0.3 mM Triton114	242± 7	1.37± 0.09	8.0± 0.2
	0.6 mM Triton114	239± 2	1.67± 1.12	8.0± 0.3
	0.9 mM Triton 114	**237± 4**	**1.89± 1.23**	**8.0± 0.2**
	1.2 mM Triton114	231± 5	2.21± 1.25	8.0± 0.2

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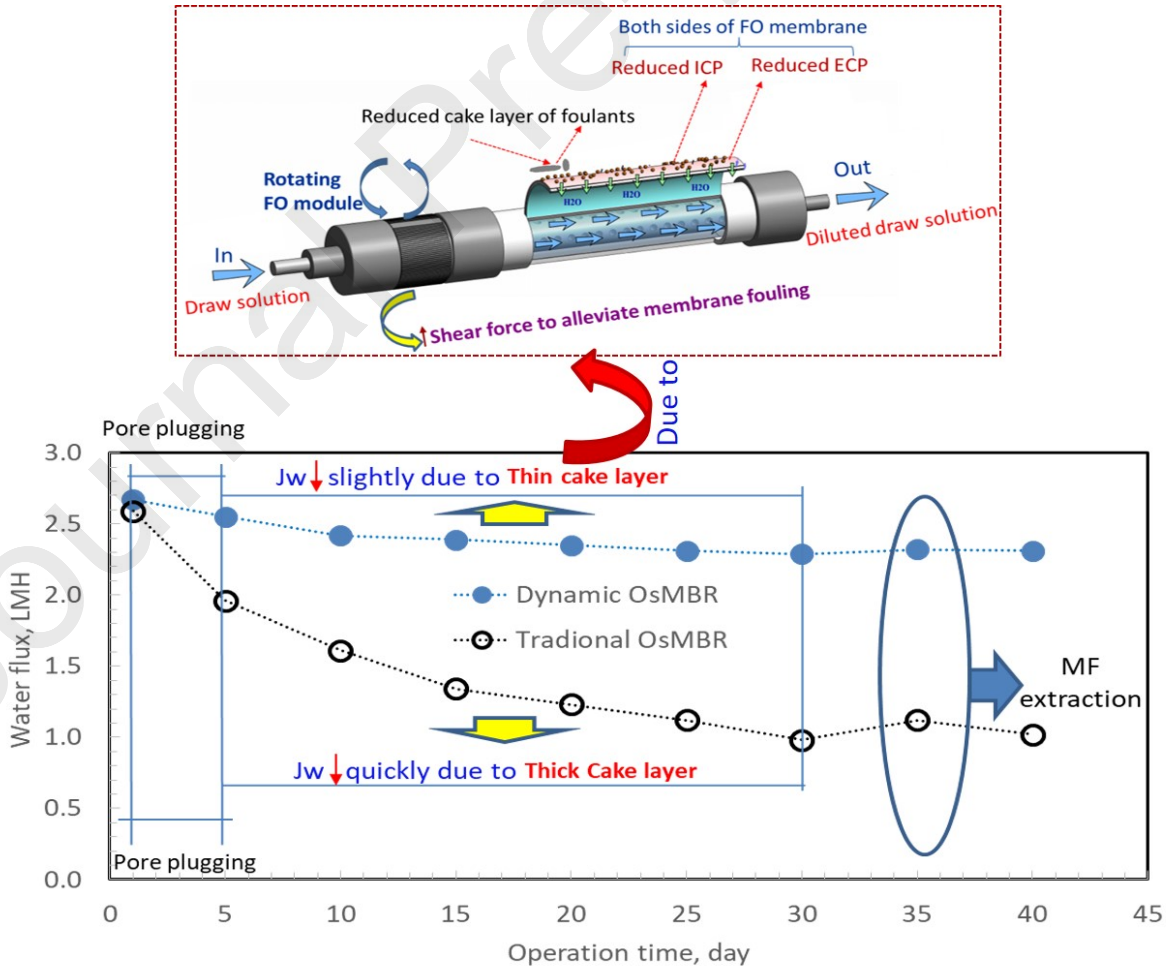

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