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# A novel osmosis membrane bioreactor-membrane distillation hybrid system for wastewater treatment and reuse



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# HIGHLIGHTS

- Highly charged EDTA was investigated as a draw solution in AGB-OsMBR/MD hybrid system.
- Low salt accumulation (<1.5 g/L) was observed during a 60-day AGB-OsMBR operation.
- AGB integrated with an OsMBR system could reduce membrane fouling of FO membrane.
- AGB-OsMBR achieved high nutrient removal (99.94% of NH4-N and 99.73% of PO<sub>4</sub>-P).
- Diluted draw solution could be effectively recovered (100%) by PTFE MD membrane.

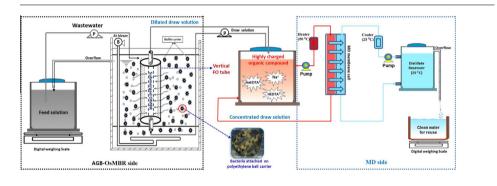
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#### G R A P H I C A L A B S T R A C T



## ABSTRACT

A novel approach was designed to simultaneously enhance nutrient removal and reduce membrane fouling for wastewater treatment using an attached growth biofilm (AGB) integrated with an osmosis membrane bioreactor (OsMBR) system for the first time. In this study, a highly charged organic compound (HEDTA<sup>3-</sup>) was employed as a novel draw solution in the AGB-OsMBR system to obtain a low reverse salt flux, maintain a healthy environment for the microorganisms. The AGB-OsMBR system achieved a stable water flux of 3.62 L/m<sup>2</sup> h, high nutrient removal of 99% and less fouling during a 60-day operation. Furthermore, the high salinity of diluted draw solution could be effectively recovered by membrane distillation (MD) process with salt rejection of 99.7%. The diluted draw solution was re-concentrated to its initial status (56.1 mS/cm) at recovery of 9.8% after 6 h. The work demonstrated that novel multi-barrier systems could produce high quality potable water from impaired streams.

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#### 1. Introduction

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Of all the sustainable water reuse technologies, membrane bioreactor (MBR) is the most feasible and has been employed in real applications (Guo et al., 2012; Luo et al., 2015). More attention is being placed on MBR systems because of their distinctive advantages, such as high quality of product water, low footprint, short hydraulic retention time, and reduced sludge production due to high biomass concentration in the bioreactor, and very high rejection of suspended solids (rejection >99%) and particles (turbidity rejection >98%) (Kraume and Drews, 2010; Wang et al., 2016). However, the widespread application of MBRs is challenged by the high operational costs associated with membrane fouling (Kraume and Drews, 2010; Qiu and Ting, 2013). Membrane fouling lowers productivity, increases energy requirements, and increases frequency of membrane cleaning and replacement, and may result in deterioration of treated water quality (Le-Clech et al., 2006). Hence, various technologies are currently being developed to overcome these limitations of conventional MBRs.

In recent years, an innovative MBR process using a forward osmosis (FO) membrane instead of a microporous membrane (conventional MBR) has been developed: the osmosis membrane bioreactor (OsMBR) (Achilli et al., 2009; Cornelissen et al., 2008) to reduce fouling and enhance rejection of dissolved species and small particles. FO membrane has a few advantages, these being: (i) low energy consumption because of the use of osmotic pressure instead of hydraulic pressure as the driving force; (ii) high rejection of various contaminants, thus increasing the quality of the product water; and (iii) low fouling propensities resulting from the dense and tight surface structure of the FO membrane (Nguyen et al., 2015, 2016; Wang et al., 2016; Yin Tang and Ng, 2014). Therefore, OsMBR is considered as a multiple-barrier technology, well suited for indirect and direct potable water reuse applications (Achilli et al., 2009; Alturki et al., 2012).

Nevertheless, a major technical challenge to OsMBR application was the lack of appropriate draw solutions that could reduce salt accumulation and membrane fouling during long-term operation (Ge et al., 2012; Holloway et al., 2015a; Wang et al., 2016). Yap et al. (2012) demonstrated that the reverse salt flux from the draw solution into the bioreactor and the high salt rejection by the FO membrane caused the build-up of salinity in the in OsMBR systems. Increased bioreactor salinity can severely impact on microbial viability and membrane performance because some functional bacteria are more sensitive to high salinity conditions (Osaka et al., 2008). Kinetics studies have suggested that nitrogen and phosphorus removal efficiency dropped to 20% and 62%, respectively, when salt concentration was 5% NaCl in the bioreactor. In addition, the salinity stress enhanced the release of both soluble microbial products (SMP) and extracellular polymeric substances (EPS), leading to severe membrane fouling (Park et al., 2015). Furthermore, an increase in the total dissolved solid (TDS) concentration in the bioreactor tank can reduce the osmotic pressure gradient across the FO membrane, causing the declined water flux (Qiu and Ting, 2013).

Among suggested draw solutions (Table1), NaCl was widely used as an inorganic draw solution in OsMBR systems because it exhibits high water flux at moderate draw solution concentrations, high solubility, and easy storage. For example, Holloway et al.

(2015b) used 0.5 M NaCl salt as the draw solution in an OsMBR system with mixed liquor suspended solids (MLSS) of 5 g/L and achieved high removal efficiencies for phosphate and chemical oxygen demand (96%) for a water flux of  $4.3 \text{ L/m}^2$  h. However, because monovalent ions (Na<sup>+</sup> with a hydrated radius of 0.18 nm and Cl<sup>-</sup> with a hydrated radius of 0.19 nm (Kiriukhin and Collins, 2002)) could easily pass through the FO membrane (membrane pore size: 0.37 nm) (Xie et al., 2012), the TDS concentration in the bioreactor increased by approximately 20 g/L after 126 days. To minimize salt leakage, Qiu and Ting (2013) demonstrated that using a divalent salt such as MgCl<sub>2</sub> (Mg<sup>2+</sup> with a hydrated radius of 0.3 nm (Kiriukhin and Collins, 2002)) in the draw solution in a submerged OsMBR could reach organic matter removal to 98% and reduce salt leakage compared with an NaCl draw solution. However, the salt accumulation in the OsMBR system was still high (>9 g/L) for an 80-day operation, because of the reverse transport of MgCl<sub>2</sub> from the draw solution and the rejection of dissolved solutes in the feed by the FO membrane. Furthermore, Ansari et al. (2015) used glycine and glucose as the organic draw solutions in OsMBR system to achieve low salt accumulation but the water flux was relatively low (Table 1).

Up to this date, new OsMBR configurations such as UF-OsMBR, NF-OsMBR, and anaerobic-OsMBR have been studied and obtained the promising results (Holloway et al., 2015b; Yin Tang and Ng, 2014). However, limited nutrient removal in single reactor, reduced salt accumulation, and the membrane fouling in longterm operation is still the major technical challenges to OsMBR application, which motivated the author to carry out this work. To the best knowledge of the authors, this is the first approach to use highly charged organic compound of Ethylenediaminetetraacetic acid disodium (EDTA-2Na) as a draw solution in an attached growth biofilm - OsMBR (AGB-OsMBR) system to simultaneously reduce salt accumulation and membrane fouling. Compared with activated sludge OsMBR, low suspended solids in AGB-OsMBR may cause low viscosity, less cake deposition, and less biofouling of the FO membrane. The high charge and large molecular size of EDTA draw solution has high potential to obtain a low reverse salt flux. Moreover, this diluted draw solution was easily recovered using MD membrane since the water flux in MD is not significantly affected by the salt concentration in the solution.

This study aimed to systematically investigate the performance of AGB-OsMBR/MD hybrid system for wastewater treatment using highly charge EDTA as a draw solution. First, the effect of the flow rate on the water flux and reverse salt flux was evaluated using deionized (DI) water as the feed solution. Next, the variation of the water flux and amount of salt accumulation with the operating duration was examined using synthetic wastewater as the feed solution. Then, the nutrient removal efficiency was then determined in the AGB-OsMBR system and the recovery of diluted draw solution was conducted using polytetrafluoroethylene (PTFE, pore size of 0.45  $\mu$ m) membrane in MD process. Finally, the membrane fouling characteristics were analyzed using scanning electron

Table 1

Comparison	of	different	draw	solutions	in	OsMBR	system.
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Draw solution	Concentration, M	Increased bioreactor salt concentration, g/L	Declined water flux, $L\!/m^2h$	Operation time, day	References
Sodium chloride	0.5	0.5–20	4.3-1.6	126	Holloway et al. (2015b)
Sodium chloride	1	0.14-4.1	12-3	7	Alturki et al. (2012)
Sodium chloride	1	0.4–12	9–5	16	Lay et al. (2011)
Magnesium chloride	0.5	0.3–9.7	7.50-5.62	80	Qiu and Ting (2013
Sodium acetate	0.72	2.41	3.5-ND	24	Ansari et al. (2015)
Glycine	1.13	3.46	3.2-ND	24	Ansari et al. (2015)
Glucose	1.31	1.48	2.3-ND	24	Ansari et al. (2015)

Note: All OsMBR experiments used CTA FO membrane and orientation membrane of active layer facing the feed solution, ND (no data) - References did not provide data.

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