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Innovative sponge-based moving bed–osmotic membrane bioreactor hybrid system using a new class of draw solution for municipal wastewater treatment

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A B S T R A C T

For the first time, an innovative concept of combining sponge-based moving bed (SMB) and an osmotic membrane bioreactor (OsMBR), known as the SMB-OsMBR hybrid system, were investigated using Triton X-114 surfactant coupled with $MgCl_2$ salt as the draw solution. Compared to traditional activated sludge OsMBR, the SMB-OsMBR system was able to remove more nutrients due to the thick-biofilm layer on sponge carriers. Subsequently less membrane fouling was observed during the wastewater treatment process. A water flux of 11.38 L/(m^2 h) and a negligible reverse salt flux were documented when deionized water served as the feed solution and a mixture of 1.5 M $MgCl_2$ and 1.5 mM Triton X-114 was used as the draw solution. The SMB-OsMBR hybrid system indicated that a stable water flux of 10.5 L/(m^2 h) and low salt accumulation were achieved in a 90-day operation. Moreover, the nutrient removal efficiency of the proposed system was close to 100%, confirming the effectiveness of simultaneous nitrification and denitrification in the biofilm layer on sponge carriers. The overall performance of the SMB-OsMBR hybrid system using $MgCl_2$ coupled with Triton X-114 as the draw solution demonstrates its potential application in wastewater treatment.

1. Introduction

Advances in wastewater treatment technology have facilitated increasing the pollutant removal efficiency and meeting stringent effluent regulations. However, there are still many challenges faced in wastewater treatment processes, especially in relation to nutrient and trace organic removal, which necessitate improving existing wastewater treatment processes for achieving higher removal efficiency (Sayi-Ucar et al., 2015). Currently, membrane technology is employed to augment water supplies, and it is crucial for sustainable water production. Among the membrane processes, membrane bioreactor (MBR) technology has become one of the most effective options for improving water sustainability; this technology encourages wastewater reuse, requires less space and produces less sludge (Guo et al., 2012; Ramesh et al., 2006). However, conventional activated sludge-based MBRs pose operational and R&D problems such as membrane fouling, high energy consumption, and limited nutrient removal capability (Nguyen et al., 2012).

To overcome these problems, a novel osmotic membrane bioreactor (OsMBR) with the following unique features was developed: (i) osmotic pressure is used as the driving force instead of hydraulic pressure, (ii) forward osmosis (FO) membranes show high rejection for a wide range of contaminants, and (iii) the membranes have a low fouling tendency (Cornelissen et al., 2011; Gwak et al., 2015; Qiu and Ting, 2014; Tan et al., 2015). 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;

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Kim, 2014). 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 bioreactor. Increased bioreactor salinity can severely impact on microbial viability and membrane performance because some functional bacteria are more sensitive to high salinity conditions (Moussa et al., 2006; 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 (Dinçer and Kargi 2001, Uygur and Kargı 2004). In addition, the salinity stress enhanced the release of both soluble microbial products and extracellular polymeric substances, leading to severe membrane fouling (Park et al., 2015).

Moreover, an increase in the total dissolved solid (TDS) concentration in the bioreactor tank can reduce the osmotic pressure difference across the FO membrane, causing the water flux to decrease rapidly (Uygur, 2006; Ye et al., 2009). For example, Holloway et al. (2014) used 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 high water flux (5.72 L/(m^2 h)). However, because monovalent ions (Na^+ with a hydrated radius of 0.18 nm and Cl^- 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 (a)), the TDS concentration in the bioreactor increased by approximately 8 g/L after 40 days (Holloway et al., 2014). To minimize salt leakage, Qiu and Ting (2013) demonstrated that using a divalent salt such as $MgCl_2$ (Mg^{2+} with a hydrated radius of 0.3 nm (Kiriukhin and Collins, 2002)) in the draw solution in a submerged OsMBR could help increase organic matter removal to 98% and reduce salt leakage compared with an NaCl draw solution. However, the mixed liquor conductivity in the OsMBR was still high, ranging from 2 to 17 mS/cm for a 80-day operation, because of the reverse transport of $MgCl_2$ from the draw solution and the rejection of dissolved solutes in the feed by the FO membrane.

A mixture of Ethylenediaminetetraacetic acid disodium salt (EDTA-2Na) and Triton X-100 was used as the draw solution in an OsMBR in our previous study. Although it can reduce the reverse salt flux appreciably and minimize salt accumulation in the bioreactor for a 60-day operation (Nguyen et al., 2015a), the water flux was relatively low because of the limited solubility of EDTA-2Na salt in water. Meanwhile, the solubility of $MgCl_2$ is high (up to 5 M) so as it can produce a high osmotic pressure and high water flux. Therefore, to achieve a high water flux and minimal salt leakage, a mixture of Polyethylene glycol *tert*-octylphenyl ether (Triton X-114) and $MgCl_2$ was used as the draw solution in the current study. The advantage of using the non-ionic Triton X-114 surfactant is that it has a large structure involving a long straight carbon chain and a low critical micelle concentration (CMC) of 0.2 mM. This structure leads to the formation of second layers on the membrane surface, constricting the membrane pores and minimizing reverse salt diffusion. Moreover, the high water solubility of $MgCl_2$ can produce high osmotic pressure as well as a high water flux in an OsMBR system.

Up to this date, the major technical challenges to OsMBR application are the build-up of salinity in the bioreactor, the membrane fouling in long-term operation and limited nutrient removal in single reactor, which motivated the author to carry out this work. To the best of our knowledge, a draw solution containing a mixture of Triton X-114 surfactant and $MgCl_2$ salt has not been used for a sponge-based moving bed (SMB)-OsMBR hybrid system to simultaneously achieve a low salt accumulation, a low fouling and high nutrient removal efficiency. Hence, this study systematically investigated the performance of the mixture as the draw solution in an SMB-OsMBR system for municipal wastewater treatment. First, the effect of the Triton X-114 concentration 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. The nutrient removal efficiency was then determined in the SMB-OsMBR hybrid system for the proposed draw solution. Finally, the membrane fouling characteristics were analyzed using scanning electron microscopy and energy dispersive x-ray spectroscopy (SEM–EDS), and fluorescence excitation-emission matrix (FEEM) spectrophotometry.

2. Materials and methods

2.1. Description of SMB-OsMBR

A laboratory scale SMB-OsMBR system is shown in Fig. 1. The FO module with an effective membrane area of 120 cm^2 was fabricated with a tube configuration and wrapped in OsMem™ cellulose triacetate with embedded polyester screen support (CTA-ES) flat sheet membranes (Hydration Technologies, Inc., Albany, OR, USA). It was then immersed in the vertical position in the bioreactor tank (6 L), with the active layer of the membrane facing the feed solution. Sponge biocarriers (Table 1) were added to the bioreactor tank after acclimatization, with a filling rate of 40% (by volume of the bioreactor). Air diffusers were installed at the bottom of the bioreactor for moving the biocarriers and reducing membrane fouling. In the SMB-OsMBR system, synthetic wastewater was continuously pumped into the bioreactor tank from a feed tank (6 L), and the liquid level in the bioreactor tank was maintained at a constant level by connecting the overflow pipe to the feed tank. The hydraulic retention time (HRT) was determined by the SMB-OsMBR water flux and was in the range of 40–51 h.

The draw solution was pumped into the FO membrane tube and this caused water from the feed solution to permeate through the membrane to dilute the draw solution. Constantly maintaining the draw solution concentration was achieved by using a conductivity controller connected to a concentrated draw solution reservoir. The feed tank was placed on a digital scale (BW12KH, Shimadzu, Japan), and the water flux was calculated according to changes in the feed tank weight.

The amount of salt accumulation in the bioreactor was determined by monitoring the conductivity of the mixed liquor with a conductivity meter (Oakton Instruments, USA). The fluctuation in the room temperature during the experiment was in the 26–29 °C range. Samples were collected from the bioreactor and draw solution tank for measuring the dissolved organic carbon, NH_4^+–N, NO_3^-–N, NO_2^-–N, and PO_4^{3-}–P. Throughout SMB-OsMBR operation, 200 mL of mixed liquor was withdrawn daily (every 24 h) from the bioreactor and allowed to settle for 30 min. The clarified supernatant was discarded. Water from the mixed liquor was used as a sample for analysis.

2.2. Feed and draw solutions

Synthetic wastewater simulating domestic wastewater served as the inoculum for the sponge carriers and as the feed solution for the SMB-OsMBR. It contained glucose, ammonium chloride, potassium phosphate, trace elements as shown in Table S1, which has 150 ± 8 mg/L dissolved organic carbon (DOC), 30 ± 2 mg/L NH_4^+–N, and 6 ± 1 mg/L PO_4^{3-}–P. In addition, deionized (DI) water was used as the feed solution to determine the reverse salt flux. $MgCl_2$ was purchased from Imperial Chemical Corp, Taiwan. Triton X-114 with a CMC of 0.2 mM was supplied by Scharlau Chemise, Spain. The