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## Microbial fuel cells and osmotic membrane bioreactors have mutual benefits for wastewater treatment and energy production



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#### ABSTRACT

This study demonstrates that microbial fuel cells (MFCs) and osmotic membrane bioreactors (OMBRs) can be mutually beneficial when integrated together for wastewater treatment. When connecting MFCs with OMBRs, the solute buildup increased conductivity and buffer capacity, which greatly increased MFC power density from 3 W/m<sup>3</sup> up to 11.5 W/m<sup>3</sup>. In turn, the MFCs conditioned and reduced sludge production and therefore reduced forward osmosis (FO) membrane fouling. The MFC-OMBR equipped with new thin-film composite (TFC) membrane showed excellent organic (>95%) and phosphorus removal (>99%) and therefore maintained effluent sCOD below 20 mg/L. However, the nitrogen removal was limited due to the negative surface charge of the thin-film composite membrane and solution chemistry, which led to higher flux of ammonium toward the OMBR draw solution. Further studies are needed to improve nitrogen removal, reduce fouling, and optimize system integration.

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#### 1. Introduction

Microbial fuel cells (MFCs) and osmotic membrane bioreactors (OMBRs) are two emerging technologies for sustainable wastewater treatment. While most studies focus on individual technology development, we hypothesize that these two processes can be mutually beneficial. MFCs use electrochemically active microorganisms to produce direct current from wastewater with less sludge production, but the current density is low from municipal wastewater due to its low conductivity and buffer capacity (Nam et al., 2010; Wang and Ren, 2013; Logan et al., 2015; Zhang et al., 2015). Another challenge of MFCs is that the effluent quality (e.g., chemical oxygen demand (COD) and turbidity) generally cannot meet the discharge standard and therefore requires post-treatment (Logan et al., 2015; Zhang et al., 2015). Membrane processes such as ultrafiltration (UF), microfiltration (MF), and FO have been incorporated into MFCs, and the effluent quality has been significantly improved (Wang et al., 2011; Ge et al., 2013a; Ren et al., 2014; Tian et al., 2014; Yuan and He, 2015; Zuo et al., 2015). Most previous research focused on this effluent quality aspect with good success, but in this study we hypothesize that the accumulation of FO solute (e.g., NaCl,  $PO_4^{3-}$  and  $HCO_3^{-}$ ) similar to high contaminants retention improves the solution salinity and buffer capacity and therefore will improve MFC power production.

On the other hand, because MFCs remove organic matters with small biomass production, we anticipate that they may reduce FO membrane fouling as pre-treatment by improving the mixed liquor properties (Tian et al., 2014). FO is an osmotically-driven membrane process where water flows from a low-salinity feed solution (FS) to a high-salinity draw solution (DS) through a semi-permeable membrane (Cath et al., 2006; Achilli et al., 2009; Lay et al., 2010; Chen et al., 2014; Holloway et al., 2014, 2015; Gu et al., 2015). The non-porous FO membrane with lower fouling propensity acts as a barrier to the contaminants so provide a high-level wastewater treatment and reclamation (Lay et al., 2010; Chen et al., 2014; Holloway et al., 2014, 2015; Gu et al., 2015). Although OMBRs are likely to have lower fouling propensity compared to the conventional pressure-driven MBRs with MF or UF membranes, fouling is still a major challenge where the concentrations of foulants are high (Lay et al., 2010; Chen et al., 2014; Holloway et al., 2014, 2015; Gu et al., 2015). Another unknown factor is the FO membrane materials. Most existing OMBR and/or osmotic microbial fuel cell (OsMFC) studies were performed with cellulose triacetate (CTA) based FO membranes (Achilli et al., 2009; Lay et al., 2010; Zhang et al., 2011; Ge et al., 2013b; Chen et al., 2014). However, it was shown that CTA membranes are vulnerable to hydrolysis and biological degradation (Geise et al., 2010; Chen et al., 2014). Thus, in this study we investigated the thin film composite (TFC) polyamide



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FO membranes, which has higher water flux, better solute rejection, and biodegradation resistance (Yip et al., 2010; Wei et al., 2011). Despite the great potential of TFC membranes for FO applications, their performance and fouling behavior in MBRs are rarely reported in the literature.

In this study, we investigated the mutual benefits between MFCs and OMBRs with new TFC FO membrane for low strength wastewater treatment. The central hypothesis is that by connecting MFC with OMBR, the accumulated solutes in OMBR increases solution conductivity and alkalinity and therefore improve ion transfer and MFC power output. The MFCs pretreat the wastewater with reduced sludge production, so it reduces membrane fouling in OMBRs. We investigated the MFC-OMBR system performance under different conditions in terms of TFC FO membrane flux level, solute transport, and nutrient removal, as well as organic removal and power production from MFCs. We also characterized TFC membrane fouling behaviors in the MFC-OMBR system and analyzed the potential mechanisms of nutrient transfer.

#### 2. Material and methods

#### 2.1. Membranes and chemicals

A commercial TFC FO membrane (Hydration Technology Innovations, Albany, OR) was used in this study. The membrane has a water permeability of 7.49  $\times$  10<sup>-12</sup> m/s.Pa, a NaCl permeability of  $7.40 \times 10^{-8}$  m/s, and a structure parameter of 0.70 mm (Coday et al., 2015; Wang et al., 2016). Membrane coupons were soaked in MilliQ water (18.2 M $\Omega$  cm) at room temperature for over 24 h before use (She et al., 2013a; Zhang et al., 2014). Glucose-based defined medium was used as the synthetic feed wastewater, so degradation and mass transfer mechanisms can be understood (Huggins et al., 2013; Gu et al., 2015; Lu et al., 2015). The medium contained (mg/L): glucose, 250; yeast extract, 100; NaCl, 400; NaHCO<sub>3</sub>, 150; NH<sub>4</sub>Cl, 80; KH<sub>2</sub>PO<sub>4</sub>, 50; MgCl<sub>2</sub>·6H<sub>2</sub>O, 5; and CaCl<sub>2</sub>, 10. The total soluble chemical oxygen demand (sCOD), NH<sub>3</sub>-N, PO<sub>4</sub>-P, electrical conductivity and pH of the feed solution were 400  $\pm$  10 mg/L,  $21.6 \pm 0.5 \text{ mg/L}$ ,  $13.0 \pm 0.2 \text{ mg/L}$ ,  $1.20 \pm 0.05 \text{ mS/cm}$  and  $7.25 \pm 0.05$ , respectively.

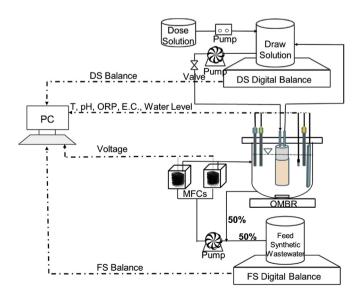


Fig. 1. Schematic diagram of the two-stage MFC-OMBR system.

#### 2.2. Reactor configuration and operation

The schematic diagram of the two-stage MFC-OMBR system is shown in Fig. 1. The system consists of two parallel MFCs and one OMBR, which are hydraulically connected in series. The singlechamber MFCs used carbon-brush anodes and air-cathodes. The volume of the MFCs was 110 and 120 mL respectively, due to the size differences of the anode brushes. The MFCs were inoculated with the anaerobic digested sludge collected from Boulder Wastewater Treatment Plant and fed with the synthetic wastewater. In the OMBR, a tubular membrane module (effective membrane area,  $A_m$ : 81 cm<sup>2</sup>) with active layer facing feed orientation was fully submerged into a bioreactor (effective volume, V<sub>R</sub>: 1.5 L). A magnetic stirrer bar was used to mix the solution in the OMBR at a speed of 400 rpm. In order to study the performance and fouling behavior of the MFC-OMBR system, different operation practices were conducted (Table 1). In Run 1, fresh medium was used after system inoculation, and no membrane cleaning was performed during the run. In Run 2, the same operation was performed, but the membrane was chemically cleaned each day to investigate the difference in membrane fouling behavior. The chemical cleaning procedure included 30 min alkaline (0.2% NaOH/0.2% EDTA) wash followed by 30 min acid (2% citric acid) wash (Wang et al., 2015). In Run 3, similar operation was used as Run 1, except 2000 mg total suspended solids (TSS)/L anaerobic sludge was added together with the medium into the MFC-OMBR system to understand if MFCs could serve as a pre-treatment to reduce membrane fouling. The performance under these operations was compared with a control conventional anaerobic OMBR (Run 4, 2000 mgTSS/L anaerobic sludge) without MFCs. Each run was performed three cycles. In Run 1-3, the same membrane module was used and chemical cleaning was performed to restore the membrane flux after each cycle; however, due to membrane damage in Run 3, a fresh membrane module was employed in Run 4. All reactors were maintained at anaerobic condition and 28  $\pm$  0.5 °C.

During operation, the solution volume of the MFC-OMBR system was monitored by a water level sensor. The flow was controlled by the volume of permeate extracted from the OMBR. When the water level of the OMBR dropped below the designated value, fresh feed wastewater converged with the same amount of bulk solution from the OMBR (Recycle Ratio = 100%) was firstly transferred into the two MFCs in parallel then into the OMBR using a peristaltic pump. Conductivity, temperature, pH, oxidation-reduction potential (ORP) values in the OMBR were monitored and logged using Lab-VIEW. In the meanwhile, draw solution (DS, 0.5 M NaCl) was recirculated at a cross flow velocity of 8.3 cm/s (equivalent to a flow rate of 230 mL/min). The concentration of DS was monitored by conductivity measurements and maintained constant through dosing a 5 M NaCl stock solution. The water flux was determined gravimetrically by weighing the mass of permeate water collected at predetermined time intervals with a digital balance (VWR, Radnor, PA). The FO membrane rejection rate  $(R_m)$  and the overall removal efficiency ( $\eta_R$ ) of the MFC-OMBR system were calculated using Eqs. (1) and (2), respectively.

$$R_m = 1 - \frac{C_P}{C_R} \approx 1 - \frac{C_{DS}}{C_R} f_D \tag{1}$$

$$\eta_R = 1 - \frac{C_{DS} V_{DS}}{C_{FS} V_{FS} - V_R \Delta C_R} \tag{2}$$

where  $C_B$ ,  $C_R$ ,  $C_{DS}$  and  $C_{FS}$  are the solutes and/or contaminants concentrations in the permeate, bulk reactor, draw solution and feed solution, respectively; and  $V_{DS}$  and  $V_{FS}$ , are the volume of draw solution and feed solution, respectively;  $\Delta C_R$  is the concentration

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