



Integrating microbial fuel cells with anaerobic acidification and forward osmosis membrane for enhancing bio-electricity and water recovery from low-strength wastewater



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ABSTRACT

Microbial fuel cells (MFCs) and forward osmosis (FO) are two emerging technologies with great potential for energy-efficient wastewater treatment. In this study, anaerobic acidification and FO membrane were simultaneously integrated into an air-cathode MFC (AAFO-MFC) for enhancing bio-electricity and water recovery from low-strength wastewater. During a long-term operation of approximately 40 days, the AAFO-MFC system achieved a continuous and relatively stable power generation, and the maximum power density reached 4.38 W/m³. The higher bio-electricity production in the AAFO-MFC system was mainly due to the accumulation of ethanol resulted from anaerobic acidification process and the rejection of FO membrane. In addition, a proper salinity environment in the system controlled by the addition of MF membrane enhanced the electricity production. Furthermore, the AAFO-MFC system produced a high quality effluent, with the removal rates of organic matters and total phosphorus of more than 97%. However, the nitrogen removal was limited for the lower rejection of FO membrane. The combined biofouling and inorganic fouling were responsible for the lower water flux of FO membrane, and the *Desulfuromonas* sp. utilized the ethanol for bio-electricity production was observed in the anode. These results substantially improve the prospects for simultaneous wastewater treatment and energy recovery, and further studies are needed to optimize the system integration and operating parameters.

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1. Introduction

As a new emerging membrane technology, forward osmosis (FO) has attracted growing interests in the field of wastewater treatment and reclamation in recent years (Achilli et al., 2009; Chung et al., 2012; Cornelissen et al., 2011; Gu et al., 2015; Wang et al., 2014a). Owing to the driving force of an osmotic pressure rather than a hydraulic pressure, FO is considering as a low fouling process resulting in a cost-saving and energy-efficient process. Besides that, FO membrane can achieve a higher quality pure water due to its high rejection for a wide range of contaminants (Coday et al., 2014; Linares et al., 2011; Xie et al., 2012; Zhang et al., 2014a). However, FO process is actually concentrating wastewater

instead of degrading contaminants. Thus, the remaining concentrates from the feed side of FO and the energy stored in organic contaminants are not recovered (Achilli et al., 2009).

Microbial fuel cells (MFCs) are a promising technology for sustainable energy production and energy-efficient wastewater treatment (Li et al., 2014; Logan et al., 2006). Despite the MFCs can effectively treat various contaminants and generate bioelectricity without any requirement for electrical grid energy, the effluent of MFCs generally cannot be directly discharged or reused without further treatment (Zhang et al., 2015). Thus, it is necessary to improve the effluent water quality for further applications of MFCs in wastewater treatment.

Although MFC and FO were two distinctly different technologies, they have some features that may complement each other. In fact, previous studies have demonstrated that integrating FO into an MFC for simultaneous water extraction, wastewater treatment and energy recovery is technically feasible (Ge and He, 2012; Ge et al., 2013; Hou et al., 2016; Ismail and Ibrahim, 2015; Warner

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et al., 2013; Zhang et al., 2011). In these reports, the configuration of MFC and FO (named as OsMFC) was same, i.e., the FO membrane was applied as a separator between the anode and the cathode instead of the proton exchange membrane. In this case, water through an FO membrane driven by the osmotic pressure difference will accelerate ion transport (e.g., protons), and thus the bio-electricity generated from MFCs is improved (Qin et al., 2015; Zhu et al., 2015). Although the electricity generation in the OsMFC is much better compared to the conventional MFC, the water flux level of FO membrane is lower due to the severe flux decline induced by internal concentration polarization (ICP), salinity build-up and membrane fouling. In addition, FO membrane fouling is still a great challenge to these internal configuration for difficultly applying an in-situ membrane cleaning. All the above mentioned problems associated with the OsMFC will eventually lead to the operation in a short time. This is why the continuous operation of OsMFC for a long time was not well investigated in previous studies (Werner et al., 2013; Zhang et al., 2011). Based on these facts, more studies are needed for better understanding the combination of MFC and FO.

In the present study, a novel combination of MFC and FO (named AAFO-MFC) was proposed for enhancing bio-electricity and water recovery from low-strength wastewater. In the AAFO-MFC system, the organic matters in wastewater were converted into the short-chain fatty acids (SCFA) and alcohols during the process of anaerobic acidification controlled at alkaline condition, and then the high rejection of FO membrane would absolutely result in their accumulation in the reactor. Based on the fact that the small molecule organic matters are much easier used by the exoelectrogens for producing electricity, the AAFO-MFC system will enhance the bio-electricity production. Furthermore, the effluent of the AAFO-MFC system was from the FO membrane, thus its water quality was much better than the conventional MFC for low-strength wastewater treatment. Compared with the OsMFCs, the side-stream configuration of the AAFO-MFC is more easy to control the salt accumulation and apply the in-situ cleaning of FO membrane, thus more possible for achieving a long continuous operation. To the best knowledge of the authors, this is the first study for simultaneously applying anaerobic acidification and FO membrane for enhancing the bio-electricity production of MFCs. Thus, the objectives of this study are 1) to evaluate the feasibility of using anaerobic acidification and FO membrane for accelerating the bio-electricity production in MFCs, 2) to investigate the performance of AAFO-MFC including power production, nutrient and organic matters removals, water flux, salt accumulation, fouling behaviors and microbial communities.

2. Materials and methods

2.1. Experimental set-up and operating conditions

A bench-scale AAFO-MFC (see Fig. 1) including a bioreactor and an MFC was applied in this study. With regard to the bioreactor with an effective volume of 4.5 L, the anaerobic acidification was achieved by keeping the pH value at 9.5 ± 0.2 through continuously dosing the sodium hydroxide (0.2 mol/L and pH 13.5) according to previous studies (Xie et al., 2014; Yuan et al., 2006). Two membrane modules (i.e., an MF and an FO) were submerged into the bioreactor. The FO membrane module with an effective membrane area of 0.025 m^2 was made of polyethersulfone (PES) (Aquaporin, Denmark), and the orientation of active layer facing the feed solution (AL-FS) was applied in this study. The properties and surface morphology of the PES membrane are summarized in Table S1 and Fig. S1, respectively. The MF membrane was made of polyvinylidene fluoride (PVDF) with a nominal pore size of $0.20 \text{ }\mu\text{m}$ and an

effective surface area of 0.025 m^2 (Zizheng Environment Inc., China).

A single-chamber MFC with carbon felt anode (40 cm^2 , Sanye Carbon Co., Beijing) and air-cathode (40 cm^2) was used in this study. The carbon felt was successively treated use 1M HCl and 1M NaOH, and then heated at $450 \text{ }^\circ\text{C}$ for 30 min before using. The activated carbon air-cathodes were made by rolling-press method according to a previous study (Dong et al., 2012). The working volume of the MFC was 160 mL. The anode and cathode electrodes were connected through a $500 \text{ }\Omega$ resistor, and the voltages were recorded automatically every 30 min using a data acquisition system (34972A, Agilent, China).

During the operation of AAFO-MFC, the synthetic wastewater was continuously pumped into the bioreactor, and the influent pump was controlled by a water level sensor to maintain a constant water level in the reactor. In order to mix the biomass intensively and alleviate the membrane fouling, gas produced from the bioreactor was recycled through a gas diffuser located below the two membrane modules with a recirculation rate of 2 L/min. The mixed liquor in the bioreactor was recirculated from the bioreactor to the MFC with a flow rate of 75 mL/min. A 0.5 M NaCl was used as the draw solution, which was monitored by a conductivity control system and maintained constantly through a concentrated salt adding device with 5 M NaCl. The draw solution was recirculated at the flow rate of 0.4 L/min with a corresponding cross-flow velocity of 1.7 cm/s. With regard to the MF membrane module, its effluent flow rate was changed according to the salinity in the reactor and controlled by a peristaltic pump.

The synthetic wastewater contained 230 mg/L glucose, 60 mg/L peptone, 40 mg/L CH_3COONa , 20 mg/L beef paste, 198 mg/L NaHCO_3 , 12 mg/L KH_2PO_4 , 170 mg/L NH_4HCO_3 , 2.4 mg/L $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$, 1.2 mg/L CaCl_2 and 1 mg/L $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ was used as the feed water. Its concentrations of chemical oxygen demand (COD), total organic carbon (TOC), ammonia nitrogen ($\text{NH}_4^+\text{-N}$), total nitrogen (TN) and total phosphorus (TP) were $373.3 \pm 17.2 \text{ mg/L}$, 146.3 ± 2.69 , 28.1 ± 1.19 , 31.7 ± 1.90 and $2.2 \pm 0.11 \text{ mg/L}$, respectively. The seeded sludge was taken from a local domestic wastewater treatment plant (Taihu Xincheng Wastewater Treatment Plant, Wuxi, China). Before the seeded sludge was put into the bioreactor, it was cultivated in a fermentation flask with an effective volume of 5 L by the synthetic wastewater for about 2 months at the temperature of $28 \pm 0.5 \text{ }^\circ\text{C}$ and pH of 9.5 ± 0.2 . The initial mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) of sludge in the bioreactor were 3.2 and 2.6 g/L, respectively. The MFC directly used the mixed liquor in the bioreactor as the inoculum and substrate. When the constant electric power was achieved, the MFC finished the start-up. The AAFO-MFC system was maintained at the temperature of $28 \pm 0.5 \text{ }^\circ\text{C}$. During the whole experiment, the sludge retention time (SRT) was maintained at 80 d, and the hydraulic retention time (HRT) varied in the range of 22–50 h due to the flux decline of FO membrane.

2.2. Analytical methods

2.2.1. General analytical methods

Water flux through the FO membrane was calculated based on the weight change of the draw solution, and the conductivity and pH of the mixed liquor were monitored and recorded by a conductivity device (OKD-650, Shenzhen OK Instrument Technology Co., Ltd., China) and a pH meter (DELTA 320, METTLER TOLEDO, Shanghai), respectively. COD, $\text{NH}_4^+\text{-N}$, TN and TP in the influent, sludge supernatant and permeates of FO and MF membranes were conducted according to the Chinese NEPA standard methods (2002), and the TOC concentration in these samples was

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