



Enhancing the performance of an osmotic microbial fuel cell through self-buffering with reverse-fluxed sodium bicarbonate



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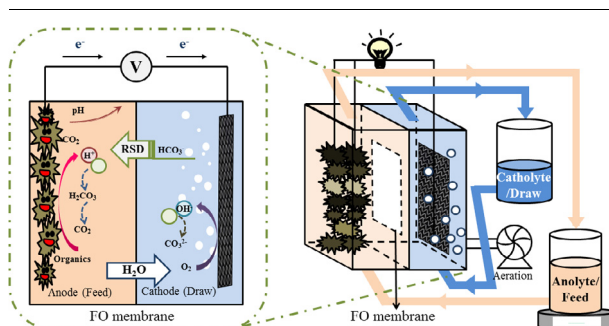
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HIGHLIGHTS

- The analyte of an OsMFC requires buffer for maintaining electricity generation.
- Reverse-fluxed bicarbonate buffers the analyte of an OsMFC.
- Reverse-fluxed bicarbonate enhances electricity generation.
- A concentration gradient is the major driving force for bicarbonate migration.
- Current generation also promotes reverse movement of bicarbonate ions.

GRAPHICAL ABSTRACT



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ABSTRACT

Osmotic microbial fuel cells (OsMFCs) combine the merits of microbial fuel cell (MFC) and forward osmosis (FO) for simultaneous contaminant removal, electricity generation, and high-quality water extraction. As an FO based technology, reverse solute flux (RSF) is one of the key challenges for its operation. Herein, RSF was converted into a positive effect on the system performance by using NaHCO_3 solution as a draw solution (DS)/catholyte. It was found that reverse-fluxed NaHCO_3 helped buffer the analyte pH and thus enhance electricity generation, compared to the OsMFC using the NaCl DS/catholyte. At the same concentration, the NaHCO_3 DS/catholyte achieved a higher Coulomb production of 1349.2 ± 80.3 C and higher analyte pH of 6.48 ± 0.19 than those of the NaCl DS/catholyte. At the same conductivity, the NaHCO_3 DS/catholyte exhibited better electricity generation performance with a comparable recovered water volume of 417.7 ± 13.7 mL to that of the NaCl DS/catholyte. As the NaHCO_3 concentration increased from 0.1 M to 0.75 M, the OsMFC electricity generation was enhanced due to the increased RSF from 19.2 ± 2.3 to 210.8 ± 17.5 $\text{mmol m}^{-2} \text{h}^{-1}$. In the anode, $92.0 \pm 0.8\%$ to $97.1 \pm 0.9\%$ of reverse-fluxed NaHCO_3 was used to neutralize protons. These results have demonstrated a new strategy that uses the bicarbonate migration driven by both a concentration gradient and electricity generation to successfully raise the alkalinity of the analyte towards enhancing electricity generation.

1. Introduction

Osmotic microbial fuel cells (OsMFCs), which are the integration of microbial fuel cell (MFC) and forward osmosis (FO), can simultaneously

accomplish contaminant removal, electricity generation, and high-quality water extraction [1]. This integration generates a number of benefits such as reduced internal resistance and enhanced current generation [2]. It could also enable wastewater treatment along with

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recovery of the desalinated water from seawater or oilfield produced water [3,4]. When an external voltage is applied, an OsMFC is converted to an osmotic microbial electrolysis cell, which exhibits enhanced hydrogen production by facilitating proton transport through FO membrane [5]. It was reported that the unavoidable membrane fouling in an OsMFC might benefit the current generation due to the decrease of internal resistance despite of the loss of water flux [6]. Appropriate membrane modification, for example with layer by layer polyelectrolyte, can increase water flux [7]. Coating silver nanoparticle-polydopamine onto FO membrane has achieved substantial reduction in biofouling as well as further improvement in current generation [8]. To help understand OsMFCs, a mathematical model was developed and used to reveal that higher electricity generation in an OsMFC than a conventional MFC was related to its lower membrane resistance [9].

A special requirement for OsMFC operation is the use of an appropriate draw solute that can create a large osmotic pressure difference across FO membrane for water extraction. In an FO system, extensive studies have been performed to select and investigate a wide range of draw solutes such as various inorganic salts, chlorhexidine gluconate, ferric-lactate complex, NH_4HCO_3 , and fertilizer [10–13]. In the presence of bioelectrochemical reactions, draw solution (DS, i.e. the liquid containing draw solute) in an OsMFC system also needs to function as a catholyte, which must have a high conductivity to reduce electrolyte resistance and high pH buffering ability in response to pH increase due to cathodic reduction reactions [14]. Because of inherent limitation with FO membrane, draw solutes can move in a direction against water flux from the draw into the feed due to a concentration gradient, called reverse solute flux (RSF) [15]. RSF could result in continuous loss of draw solutes, rendering reduced osmotic driving force (lower water flux), increased membrane fouling propensity, and elevated operation cost due to periodical replenishment of draw solutes [16,17].

There have been various efforts to reduce RSF, e.g., via membrane modification, use of larger molecule chemicals as draw solute, and electrolysis-assisted reduction [18–21], or recovery of reverse-fluxed draw solute via electrodialysis [22]. In particular, bioelectricity generation in an OsMFC was able to inhibit solute movement and thus decrease RSF [23]. However, no matter what methods are used, there are always some draw solutes migrating into the feed side. It will be of strong interest to turn this “negative effect” (i.e. draw solute intrusion) into a “beneficial effect”. Recently, we demonstrated the feasibility of using reverse-fluxed magnesium ions to accomplish *in situ* struvite formation in an FO system treating digested swine wastewater [24]. It was also found that the reversed-fluxed organic ionic draw solutes, e.g. sodium acetate and magnesium acetate, could have a positive effect on methane production when integrating FO with anaerobic treatment [25].

Herein, we proposed a new OsMFC system that took advantage of reverse-fluxed draw solute to achieve anolyte buffer for enhanced electricity generation. NaHCO_3 was selected as a draw solute due to its high buffer capacity and solubility to secure high-quality/quantity water reclamation, efficient electricity production, and pH stabilization of both the catholyte and the anolyte [26,27]. The specific objectives of this study were to (1) examine the feasibility of bicarbonate solution as a draw solution/catholyte in an OsMFC that had no buffer addition into its anode; (2) investigate the OsMFC performance affected by concentration and conductivity of bicarbonate; and (3) evaluate the possible mechanism of bicarbonate movement and transformation.

2. Materials and methods

2.1. Setup of a lab-scale OsMFC

The OsMFC system consisted of two identical compartments ($14 \times 7 \times 3.7$ cm, 360 mL/each) separated by a thin film composite (TFC) FO membrane with a surface area of 98 cm^2 (Hydration Technology Innovations, LLC, Albany, OR, USA) and with its active

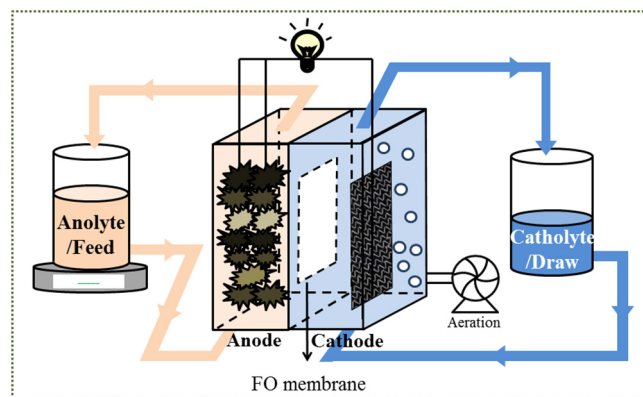


Fig. 1. Schematic of the osmotic microbial fuel cell used in this study.

layer facing the anode (feed side) (Fig. 1). Each compartment was connected to an external 1000-mL bottle (i.e. reservoir), and the mass change of the anolyte (the feed) was measured by placing its reservoir bottle on a digital balance that was monitored via computer. Two pieces of plastic mesh were placed on both sides of the FO membrane to provide sufficient physical support and avoid potential membrane swelling. Two carbon brushes (Gordon Brush Mfg. Co. Inc., Commerce, CA, USA) pretreated by acetone and 30-min heating (450°C) were served as the anode electrodes. One piece of carbon cloth (Zoltek Corporation, St. Louis, MO, USA) coated with 0.5 mg cm^{-2} activated carbon (AC, Thermo Fisher Scientific, Bridgewater, NJ, USA) was used as the cathode electrode. Both electrodes were connected by copper wire to an external resistor (10Ω). Two Ag/AgCl reference electrodes (0.197 mV vs. standard hydrogen electrode, SHE) were installed close to corresponding electrodes.

The anode chamber of the OsMFC was first inoculated with the anaerobic sludge (mixed liquor suspended solids (MLSS), 5.98 g L^{-1} ; mixed liquor volatile suspended solids (MLVSS), 4.52 g L^{-1} ; MLVSS/MLSS ratio, 75.6%) obtained from the Peppers Ferry Regional Wastewater Treatment Plant (Radford, VA, USA) and then was fed with a synthetic municipal wastewater containing (per liter deionized water (DI)): NaAc, 0.25 g; NH_4Cl , 0.3 g; NaCl, 1.0 g; MgSO_4 , 0.03 g; CaCl_2 , 0.04 g; and trace element, 1 mL [28]. The cathode was supplied with aeration of air to provide oxygen for cathodic reduction reaction. The OsMFC was operated in a batch mode under a room temperature of $\sim 20^\circ\text{C}$, and with a hydraulic retention time (HRT) of 22 ± 2 h. The anolyte and catholyte were both recirculated at 100 mL min^{-1} to provide mixing. At the end of a batch test, both chambers were drained and washed with 1000-mL DI water for 0.5 h to remove membrane foulants.

2.2. Experiment procedures

The experiment started with examining the impact of buffer on the anode performance by adding NaHCO_3 to the anolyte at various concentrations (0, 1, 3, 5, and 7 mM). For comparison, the OsMFC was also operated with dosing phosphate buffer solution (PBS) to the anolyte at the same concentration as that of NaHCO_3 . The initial anolyte conductivity with either NaHCO_3 or PBS (same mole concentration) was adjusted with NaCl to achieve same conductivity. In this test, the DS/catholyte was 0.5-M NaCl solution. Next, NaHCO_3 was used in the OsMFC cathode, serving as both draw solute and catholyte, while the anode was fed with buffer-free synthetic wastewater. To examine the OsMFC without the anode buffer, 0.75 M NaHCO_3 solution was applied as DS/catholyte, and compared with NaCl solution at the same concentration (performance benchmark). To reduce the effect of conductivity, both the NaHCO_3 and NaCl solutions were adjusted to have the same conductivity of 50 mS cm^{-1} in the additional tests. The effect of the NaHCO_3 concentration in the DS/catholyte was studied by varying it to 0.10, 0.25, 0.50, 0.75 M. Note that the catholyte

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