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Hydrogen production in microbial reverse-electrodialysis electrolysis cells using a substrate without buffer solution

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HIGHLIGHTS

- Using a substrate without buffer solution in MRECs achieved hydrogen production.
- Increasing in anolyte HRT and OLR increased cell current with stable anode potential.
- Hydrogen production rate was 0.61 m^3-H_2/m^3-V_{an}/d (yield = 0.51 mol-H_2/mole-COD).
- Efficiencies of Coulombic and COD removal were 41% and 81% (rate: 1.55 g-COD/L/d).

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ABSTRACT

The aim of this work was to use substrate without buffer solution in a microbial reverse-electrodialysis electrolysis cell (MREC) for hydrogen production under continuous flow condition (10 cell pairs of RED stacks, HRT = 5, 7.5, and 15 h). Decreasing in the HRT (increasing in the organic matter) made cell current stable and increased. Hydrogen gas was produced at a rate of 0.61 m^3-H_2/m^3-V_{an}/d in H-MREC, with a COD removal efficiency of 81% (1.55 g/L/d) and a Coulombic efficiency of 41%. This MREC system without buffer solution could successfully produce hydrogen gas at a consistent rate.

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1. Introduction

The world energy consumption rate is projected to grow with both population and economic increases in the 21st century (Lewis and Nocera, 2006). Oil prices are also increasing due to limited fossil energy and the continuous consumption of fossil fuel. As a result, sustainable and renewable alternative energies are of great interest and are being studied in a wide range of areas including solar-thermal, wind power, and hydrogen. Solar-thermal and wind power energies, however, are non-continuous energy sources that can only produce power with sun and wind, respectively. In contrast, hydrogen can be produced from inexhaustible water sources, and use in transportation burns cleanly compared with traditional fuel sources. Thus, hydrogen has been widely discussed in the context of energy as a possible future alternative energy carrier.

Hydrogen gas as sustainable energy from an inexhaustible source is important for avoiding the environmental problems associated with energy depletion from the use of fossil fuels (Ho et al., 2012). Although water electrolysis is a simple and reliable method for generating high-purity hydrogen, it requires energy input to sustain the process.

MREC can electrochemically generate hydrogen gas at the cathode from the current produced from exoelectrogenic bacteria at the anode; this oxidizes organic matter and can transfer electrons to the anode. However, the electrical power (−0.30 V using acetate; pH 7) from exoelectrogens at the anode is not enough to split water (>1.2 V) for hydrogen production. Thus, an additional voltage is required to overcome the thermodynamic threshold (Nam et al., 2012). In order to cover the need for electrical energy, reverse electrodialysis (RED) stacks, which provide electrical energy from salinity gradient energy (theoretically ~0.1–0.2 V per membrane pair based on the open circuit potential), are placed between the

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anode and cathode chambers of an MREC (Veerman et al., 2009; Dlugolecki et al., 2009).

In many studies related to energy production, high-concentration buffer solutions are used to maintain a neutral pH condition for exoelectrogens and to increase electrolyte conductivity. Examples include microbial fuel cells (MFCs), microbial electrolysis cells (MECs), and MRECs (Zhang et al., 2015; Ambler and Logan, 2011; Luo et al., 2013). However, the addition of buffer solution is expensive for wastewater treatment applications. Phosphate buffer solution (PBS) in particular can contribute to eutrophication as the effluents are discharged without phosphate removal.

In this study, we demonstrated the possibility of using a substrate without buffer solution on an MREC for hydrogen gas production. The experiments were conducted under continuous flow for the anode, and in fed-batch mode for the cathode. The anolyte was complied with only a ratio of 300:5:1 = COD:N:P for anaerobic conditions. The RED stack had thin flow channels to improve the driving force in comparison with previous MRECs (Luo et al., 2013; Kim and Logan, 2011) using NaCl as a high-concentration (HC) solution. Tests were initially conducted to evaluate the effect of the anolyte HRT on current generation using substrates with PBS. Following these tests, the substrate without buffer solution was used in the MRECs. Using a substrate without buffer solution has not been investigated for hydrogen production in an MREC, and the performance of this MREC was discussed using hydrogen production, yield, Coulombic efficiency, and COD removal rate.

2. Methods

2.1. MREC setup

The rectangular parallelepiped acrylic reactor consisted of anode and cathode chambers with a working volume of 36 mL each (cross section area: 12 cm^2, 3 cm length) (Fig. 1). An inlet and outlet were drilled perpendicularly in the bottom and top on the wall of the anode chamber for continuous flow. A pH probe was placed in the anode chamber to continually monitor and control the pH of the anolyte. A carbon fiber brush was used as the anode electrode (2.5 cm diameter, 3 cm length; T700 SC-12000, Toray, Japan), which was pretreated by heating in a furnace at 450 °C for 30 min. A titanium mesh (width: 3.5 cm, length: 2 cm, thickness: 0.2 cm) plated with Pt (2 μm of thickness) was used as a cathode electrode. A Tedlar bag was inserted in the top of the cathode chamber for gas collection with a silicon stopper. Each chamber was equipped with an Ag/AgCl reference electrode (RE-5B, BASi) to measure the electrode potential.

A RED stack was located between the anode and cathode chambers that consisted of 10 pairs of high-concentration (HC) and low-concentration (LC) cells made with 11 anion- and 10 cation-exchange membranes (Selemion AMV and CMV, Asahi glass, Japan). Silicon gaskets, which have a rectangular open section of 12 cm^2 (3 × 4 cm) for flow, were placed between the anion and cation exchange membranes with a nylon mesh spacer (0.18 mm), and the empty bed volume of the stack was 4.3 mL. HC and LC solutions flowed continuously through each cell, but in opposing directions. Each solution was provided into the RED stack at a fixed flow rate of 1.2 mL/min.

2.2. MREC operation

The anode was pre-acclimated with exoelectrogen in an acetate-fed single air-cathode MFC (Song et al., 2015). The acclimated anode was transferred to the MREC when the MFC produced the same stable maximum voltages repeatedly for three cycles.

The anolyte complied with a ratio of COD:N:P = 300:5:1 for anaerobic microorganisms using CH_3COONa, NH_4Cl, and $NaH_2PO_4 \cdot 2H_2O$ (Tchobanoglous et al., 2004). 20 mM NaOH was used to maintain the anolyte at a neutral pH. The catholyte was synthetic HC solution degassed using sonication and a vacuum pump. The synthetic HC solution was 600 mM NaCl, and the LC solution was 12 mM NaCl created using a salinity ratio of 50. The total volume of the MREC was 36 (anode) + 36 (cathode) + 4.3 (RED stack) = 76.3 mL. The anolyte was continuously supplied and stirred at 80 rpm with a magnetic bar to reduce concentration loss. The catholyte was replaced every fed-batch cycle at room temperature when the current decreased to less than 1.0 mA. The anode and cathode were connected to a fixed external resistance of 10 Ω.

2.3. Experimental measurements and calculations

The cell and electrode voltages were monitored and recorded every 10 min with a voltage recorder (VR-71, T&D Corporation) connected to a computer. Current was calculated based on the measured cell voltage across the 10 Ω resistor using Ohm's law.

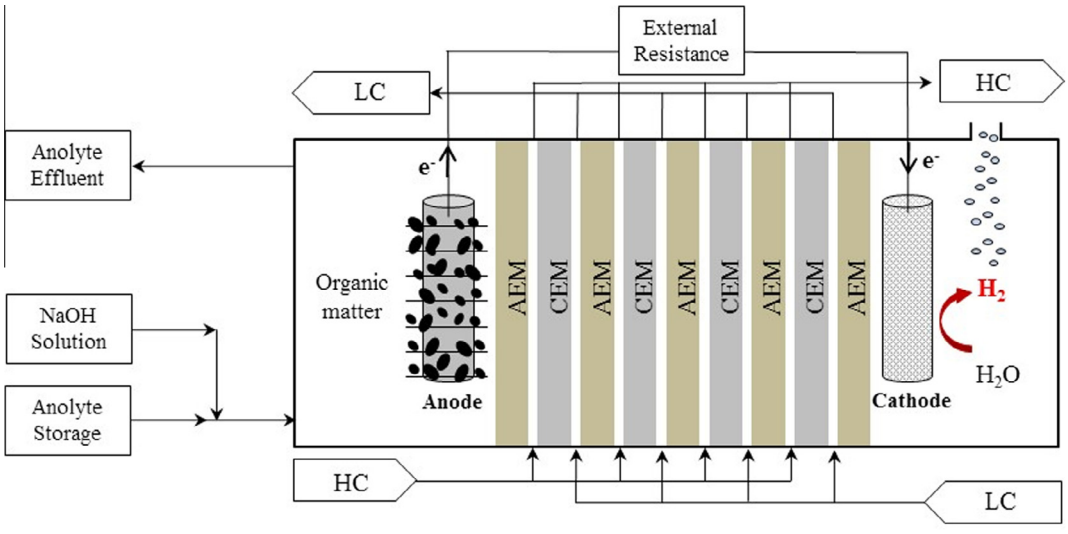

Fig. 1. Schematic diagram of MREC process set-up showing flow paths through the RED stack.

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