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Short Communication

Hydrogen peroxide production in a pilot-scale microbial electrolysis cell

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ABSTRACT

A pilot-scale dual-chamber microbial electrolysis cell (MEC) equipped with a carbon gas-diffusion cathode was evaluated for H_2O_2 production using acetate medium as the electron donor. To assess the effect of cathodic pH on H_2O_2 yield, the MEC was tested with an anion exchange membrane (AEM) and a cation exchange membrane (CEM), respectively. The maximum current density reached 0.94–0.96 A/m^2 in the MEC at applied voltage of 0.35–1.9 V, regardless of membranes. The highest H_2O_2 conversion efficiency was only 7.2 ± 0.09% for the CEM-MEC. This low conversion would be due to further H_2O_2 reduction to H_2O on the cathode or H_2O_2 decomposition in bulk liquid. This low H_2O_2 conversion indicates that large-scale MECs are not ideal for production of concentrated H_2O_2 but could be useful for a sustainable in-situ oxidation process in wastewater treatment.

1. Introduction

Microbial electrochemical or electrolysis cells (MECs) are considered a potential sustainable platform for energy-efficient wastewater treatment, due to resource recovery and wastewater treatment. Because of the dual benefits, MECs have gained tremendous attention in the last decade [1,2]. Several studies have attempted pilot-scale MECs for either electricity or H_2 production [3–5] to deploy MECs in field. However, none of these studies provided significant benefits of the recovered resource against input energy and materials.

H_2O_2-producing MECs can give significant profits over other MECs due to high cost and demand of H_2O_2 [6]. In addition, the recovered H_2O_2 from organic waste or wastewater can be used as an in-situ oxidant in wastewater treatment, improving the sustainability of wastewater management. Similar to a conventional MEC system, H_2O_2-producing MECs comprise of two chambers separated by an ion exchange membrane. A solution containing dissolved organic matter is fed to the anode chamber where anode-respiring bacteria (ARB) such as *Geobacter* sp., *Pseudomonas* sp., *Shewanella* sp., etc. oxidize the organics and use the anode as the electron sink [7–10]. The electrons flow through an external circuit to the cathode where oxygen is electrochemically reduced to H_2O_2 at the cathode surface by the two-electron pathway shown in Eq. (1) below [11]:

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \quad (1)$$

All studies to date have examined H_2O_2-MECs at the lab scale, investigating H_2O_2 conversion efficiency, reactor design, electrode materials, and so on [11–14]. These lab-scale experiments have commonly showed high potential of H_2O_2-MECs, but scale-up tests are essential to demonstrate performance and benefits of the MECs; however, no large-scale MECs for H_2O_2 generation have been conducted yet.

This study is the first pilot-scale MEC (110 L) experiment for H_2O_2 production. The pilot MEC was featured with anode modulation for provision of high surface area for biofilm formation and passive oxygen diffusion to a non-Pt carbon cathode. To evaluate the effect of catholyte pH on H_2O_2 yield, the pilot-scale MEC fed with acetate medium was run using an anion exchange membrane (AEM) and a cation exchange membrane (CEM), respectively, as electrode separator. Performance of the MEC was summarized, focusing on electrode potential, current density, pH, and H_2O_2 yield.

2. Materials and methods

2.1. Reactor configuration

Fig. 1 presents the schematic diagram and the picture of the pilot-scale MEC. The system has a dual-chamber configuration equipped with bioanode modules and a gas diffusion cathode (the anode chamber 1 m × 0.5 m × 0.2 m and the cathode chamber 1 m × 0.5 m × 0.02 m). The volumes of the anode and a cathode chamber were 100 L and 10 L, respectively. To provide the large

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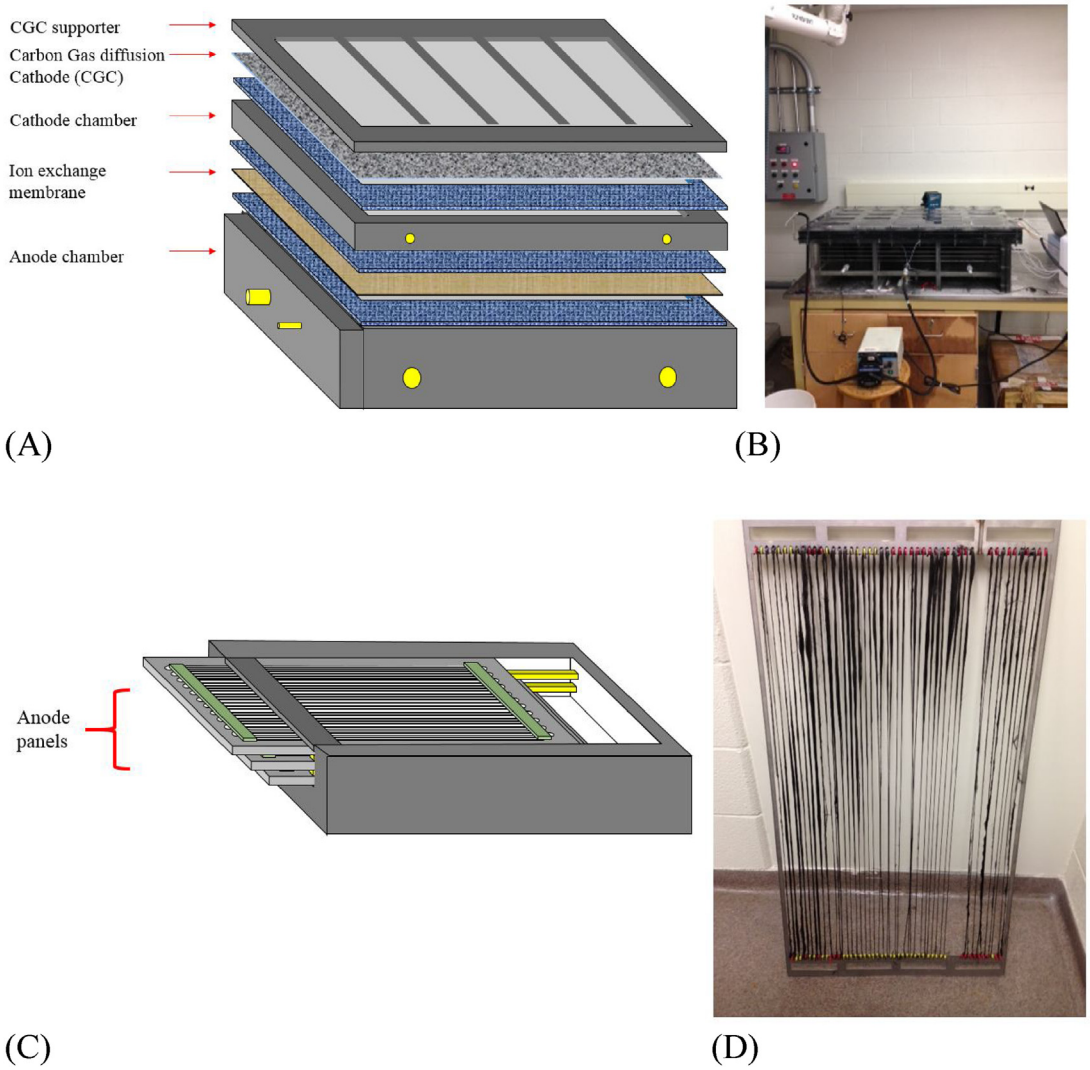

Fig. 1. Schematic diagram of a large-scale microbial electrochemical cell (MEC). (A) MEC components, (B) photo of the MEC, (C) anode modulation, and (D) photo of an anode module.

surface area for biofilm formation without increasing footprint of the MEC, the anode was fabricated by connecting carbon fibers (2293-A, 24A Carbon Fiber, Fibre Glast Development Corp., Ohio, USA) to a stainless current collector, as shown in Fig. 1B. The MEC was equipped with five anode modules (Fig. 1C), providing a specific surface area of 1.27 m^2/m^3 anode. The carbon fibers were pretreated with nitric acid (1 N), acetone (1 N), and ethanol (1 N), and finally washed with tap water before use [15]. Peristaltic pumps (Masterflex L/S Economy Drive 7554-90, Cole-Parmer, USA) were used to circulate both anolyte and catholyte at a flow rate of 2 L/min for mixing.

Cathode catalyst selection is one of the critical parameters in H_2O_2 producing MECs. Precious-metal-free carbon cathodes are preferred for H_2O_2 production [16,17]. When using precious metal-based catalysts like platinum, the four-electron oxygen reduction to water (Eq. (2)) many outcompete the two-electron reduction to H_2O_2 (as shown in Eq. (1)).

$$O_2 + 4H^+ + 4e^- \rightarrow 2\ H_2O \quad (2)$$

Due to advantages of high conductivity, low cost, long-term stability and low catalytic activity of H_2O_2 decomposition to water, carbon-gas diffusion electrode (GD2230, Fuel Cell Earth, USA) was used as the cathode (called, carbon gas-diffusion cathode (CGC) in this study. Passive diffusion of O_2 from atmosphere through the CGC, means no energy requirement for oxygen supply to the cathode in the MEC. An anion exchange membrane (AEM) (AMI-7001, Membranes International Inc., USA) having a surface area of 0.5 m^2 was used for the MEC, which was later replaced with a cation exchange membrane (CEM) (CMI-7000, Membranes International Inc., USA) for comparison.

2.2. Inoculation and operation

The pilot MEC equipped with AEM was inoculated with effluent from lab-scale MECs (3.5 L of anolyte) operated with acetate medium, and was fed with 20 mM acetate medium [15]. The medium was sparged with ultra-pure nitrogen (99.999%) for 30 min. Then, $FeCl_2 \cdot 2H_2O$ (20 mM) and $Na_2S \cdot 9H_2O$ (77 mM) were added to acetate medium (1 mL per L). The pH in acetate medium was constant at 7.3 ± 0.1. The cathode chamber was filled with tap water. The AEM-MEC had been run in batch mode (~4 months) until a peak current density of ~0.9 A/m^2 (~0.45 A) was repeatedly observed in the MEC. Then, experimental data was collected in the batch pilot MEC. AEM was replaced with CEM later, and the CEM-MEC was operated for comparison experiments.