



Full length article

Enhancement of bioelectricity generation for an air-cathode microbial fuel cell using polyvinyl alcohol-membrane electrode assemblies

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ABSTRACT

This study applied polyvinyl alcohol (PVA) film as the air-cathode membrane electrode assembly (MEA) of the microbial fuel cell (MFC) to estimate the electricity generation at various ethyl acetate (EA) concentrations. PVA-MEA can be produced by compressing carbon cloth with PVA hydrogel fabricated by freezing and thawing, and further drying the compressed product. The PVA-MEA thus prepared could serve as an air-cathode. The self-developed PVA-MEA exhibited excellent water absorption (water uptake: $149.4 \pm 0.04\%$) and an excellent swelling ratio ($12.15 \pm 0.17\%$), capable of accelerating proton transmission through water molecules to the cathode to participate in the reduction reaction. When the concentration of EA increased from 26.2 mg/L to 1048 mg/L, the biofilm on the anode surface increased from 8 mg biomass/L to 274 mg biomass/L, attaining an average closed-circuit voltage (CCV) of 382–425 mV, an average open-circuit voltage of 468–682 mV, and a maximum power density of 59–143 mW/m²; moreover, the Coulombic efficiency (E_c) was elevated from 0.22% to 13.57%, by approximately 62 times. For the Monod kinetic parameters, the half-saturation constant was 3.74 mg/L, and the maximal CCV output was 425 mV. The PVA-MEA developed in the study could serve as an air-cathode MEA of an AC-MFC without having any modification material added to the cathode surface, achieving maximum voltage output with low concentrations of EA.

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1. Introduction

Ethyl acetate (EA) is commonly used as a pharmaceutical extracting agent, a diluent used in chemical synthesis-based pharmaceutical factories, or a food additive that provides artificial flavor in the food industry [1]. EA-containing wastewater is produced from washing the equipment used for EA-related processes. Substantial EA-containing wastewater can enter wastewater-processing plants and burden them [2]. Exhaust emissions of EA from various industrial processes cause environmental damage and are hazardous to human health. However, because EA is a highly biodegradable compound, biological treatment is a favorable and economical technique for EA-processing. Although EA biodegradation and the concurrent bioelectricity generation in an air-cathode microbial fuel cell (AC-MFC) that is equipped with a polyvinyl

alcohol and membrane electrode assembly (PVA-MEA) have been reported [3], the characteristics of the electricity production kinetics by the AC-MFC have not been explored. Therefore, we examined the extent of EA removal and concurrent bioelectricity generation by an AC-MFC in this study.

A microbial fuel cell (MFC) is a device that transforms chemical energy to electricity by employing microorganisms to catalyze organic matter. This innovative biological treatment technology is effective in simultaneously removing pollutants and generating electricity [4,5]. The main components of MFC comprise anodes, cathodes, proton exchange membranes (PEMs), organic matter that serves as fuel, and microorganisms. All these components can affect the efficacy of electricity production. The air-cathode microbial fuel cell (AC-MFC) has the capacity to directly use oxygen in the atmosphere as the terminal electron acceptor, and thus does not require the establishment of a cathode chamber to facilitate oxygen transportation [6]. Therefore, the conventional double-chamber MFC has been gradually replaced by the single-chamber AC-MFC [7,8]. The key to influencing the electricity production efficiency of AC-MFCs is through the function of PEMs. The main purposes

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of PEMs are to deliver protons, thus preventing fuel diffusion from the anode chamber and oxygen diffusion into the anode chamber. The mechanism to maintain the humidity of the PEMs and facilitate proton delivery is the key factor influencing the output voltage of MFCs [9,10]. Currently, numerous scholars have attempted combining a PEM with cathodes to produce one-pieced membrane electrode assemblies (MEAs), which have been observed to lower the proton–oxygen mass transfer limitation, shorten the reaction time of the cathodes, and thus enhance electricity production efficiency [11–13].

In recent years, a number of materials replacing the costly Nafion or nonfluorinated polymer membranes to serve as the PEM of MFCs have emerged, including polyvinyl alcohol (PVA), PVA/sulfosuccinic acid [14], polytetrafluoroethylene [15], and perfluorosulfonic acid [16]. Among the aforementioned materials, PVA exhibits the most favorable film formability and water absorption. In a hydrogel state, a crosslinking reaction can be induced between the hydrogen bond and hydroxide (–OH) of the PVA structure through repeated procedures of freezing and thawing according to the principles of material expansion under heat and contraction under coldness [17]. Stable bonding can be formed after sufficient reactions between the hydrogen bond and hydroxide are conducted. After a drying procedure, film products are completed [18]. Self-developed PVA-MEAs were used in the present study as the cathodes of the AC-MFC. The characteristics of EA electricity production at various concentrations were assessed. To identify the characteristics of the biodegradation kinetics of the AC-MFC, the output voltage produced after the degradation of EA by microorganisms was employed to obtain the kinetic parameters; moreover, the Monod model was used to explain the characteristics of the electricity production kinetics of the AC-MFC [19,20].

2. Material and methods

2.1. Consortia, inoculation and biomass measurement

Bacterial strains were collected from the activated sludge in the petroleum refining wastewater treatment plant affiliated with the Nan Ya Plastics Corporation (Yunlin Plant, Taiwan). The collected anaerobic bacterial strains were inoculated in a 3-L AC-MFC and connected a 1000 Ω external resistor for an extended period of time, in which EA served as the only carbon source. The AC-MFC was incubated in the room temperature. After 6 months of acclimation, mixed bacterial strains that could degrade EA were obtained. In the process of bacteria acclimation, the EA injection volume was enhanced by 50 μL every 7 d until the injection volume reached 2000 μL , by which time, the EA concentration in the solution was 720 mg/L. The concentration was used to maintain culture sludge that had an EA-degrading capability. The composition of the anolyte/mineral medium solution was described in Wu et al. [21]. For biomass measurement, the anode was sonicated at 70 kHz at 37 °C in a test tube that was submerged in a Sonicator SC-100 sonicating bath (Sonicor Instrument Co., Copiaque, NY) for 10 s to remove adherent bacteria, and rinsed with sterile medium. Suspended solid in the solution was determined.

2.2. Construction of AC-MFC

In a conventional AC-MFC, costly Nafion was commonly used as the PEM. Self-developed PVA-MEAs showing high water retention and water absorbency, which accelerate proton transport in the PEM, were used in the present study as the cathodes of the AC-MFC. The AC-MFC was assembled using acrylic sheets of 8 cm \times 8 cm \times 5.5 cm. The total chamber volume was 352 mL (Fig. 1). The cathode and PEM were fabricated through press form-

ing, in which pieces of carbon cloth that were 5 cm \times 5 cm in length and width (0.7 mm thickness, CW1001, KoTHmex, Taiwan) were pressed on denatured 10% (wt) PVA hydrogel that had underwent freezing (–20 °C, 6 h) and thawing (room temperature). PVA (BF-26, Chang-Chun Group, Taiwan) is a straight-chain polymer with excellent hydrophilicity and water absorption. At a high temperature, PVA is miscible with water and forms a PVA–hydrogel solution. The preparation method of the PVA hydrogel was referred to in a study by Wu et al. [21]. The carbon cloth pieces and PVA hydrogel were compressed, fully bonded, and completely dried in an oven at 50 °C to produce the PVA-MEA used in the present experiment. The anode was fabricated by graphite felt of 5 cm \times 5 cm in length and width (10 mm thickness, Hongye Vacuum Technology, Taiwan). A corrosion resistant, highly conductive titanium wire was applied to connect the anode and cathode (0.4 mm, Hengde metal Co., Ltd., China). An external resistance (1 k Ω) was set between the anode and cathode to facilitate electrode transmission.

Synthetic wastewater with EA concentrations of 26.2–1048 mg/L was prepared by adding EA solvent to the mineral medium solution. In a batch experiment, 352 mL of inorganic solution in the anode chamber of a MFC was periodically inoculated with a microbial consortia suspension that contained 4335 \pm 29.7 mg/L of biomass.

2.3. EA and its byproduct analysis

A gas chromatography–flame ionization detector (GC-FID, Shimadzu, GC-2014, Japan) was used to analyze the liquid-phase EA, ethanol, acetic acid, and acetaldehyde concentration. Sample extraction was conducted using a 1- μL airtight injector to extract liquid from the anode chamber, which was then injected to the GC-FID for analysis. The temperature parameters for the GC-FID were as follows: injector 150 °C, initial column temperature was held at 60 °C for 2 min, then programmed at 20 °C/min to 140 °C for 2 min, and detector 180 °C. Capillary columns of 30 m \times 0.53 mm \times 1 μm were used (Stabliwax-DA, Restek, USA). In a sample analysis, high-purity nitrogen (purity >99.98%) was used as the carrier gas (3 kg/cm²), and air (5 kg/cm²) and hydrogen (3 kg/cm²) were employed as the combustion gases.

2.4. Measurement of water uptake and swelling ratio of PVA-MEA

Wet and dry weight measurements were used to calculate the water uptake of the PVA-MEA in this study. The PVA-MEA was dried in an oven at 50 °C for 24 h in advance; its weight was then obtained as the dry weight. Subsequently, the PVA-MEA was soaked in deionized water for 24 h and then retrieved; after residual water was absorbed using an absorbent paper, the remaining weight was determined as the wet weight of the PVA-MEA. Eq. (1) presents the calculation of the water uptake [22].

$$W_{\text{wateruptake}} = \frac{W_{\text{wet}} - W_{\text{dry}}}{W_{\text{dry}}} \times 100\% \quad (1)$$

where $W_{\text{wateruptake}}$ represents the water uptake of the PVA-MEA (%), W_{wet} represents the wet weight of the MEA, and W_{dry} represents the dry weight of the MEA.

The swelling ratio of the PVA-MEA was acquired using the volume method. The PVA-MEA was first dried in an oven at 50 °C for 24 h and then retrieved; the length, width, and thickness of this sample were measured to obtain the dry volume of the PVA-MEA. The dried PVA-MEA was then soaked in deionized water for 24 h, retrieved, and had its residual water absorbed using an absorbent paper. The length, width, and thickness of this sample in a wet con-

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