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Feasibility study of electricity generation and organics removal for a molasses wastewater by a waterfall-type microbial fuel cell

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ABSTRACT

A membrane electrode assembly (HEM) was incorporated with microbial fuel cell (MFC) to create a HEM-MFC for treating molasses wastewater and converting the pollutant to electrical energy. The most important novelties in this work include the use of polyvinyl alcohol-hydrogel (PVA-H) to replace the less permeable and more expensive proton exchange membrane and the placement of the reactor at an adjustable tilt angle which enabled a proper connection of independent MFC units of the reactor in series to maximize its output voltage, organics removal efficiency, and decolorization. The results showed that the PVA hydrogel introduced into the HEM sustained the moisture for the membrane electrode, thus facilitating proton transmission. When the HEM-MFC was arranged parallel to the wastewater surface and formed a tilt angle (θ) of 0°, the reactor generated electricity by using one MFC only. Increasing the tilt angle enabled inflow wastewater to create differences in the water displacement heights between each chamber of the reactor, thereby forming a set of MFCs connected in series. At tilt angle of 25°, the proposed reactor attained a COD removal efficiency of 95.6% and decolorization of 60.1%. In addition, a tilt angle of 25° yielded a maximum open-circuit voltage (OCV_{max}) of 2130 mV and power density (PD) of 16.1 mW/m^2, which were higher than those achieved through a tilt angle of 0° (the OCV_{max} and PD increased 3.9 and 18.7 fold, respectively). Overall, the proposed waterfall-type MFC increased its voltage output by connecting the MFCs in series, effectively treated and decolorized molasses wastewater, and demonstrated considerable potential for scale-up development.

1. Introduction

Molasses wastewater is a type of organic wastewater commonly produced by the food industry. Such wastewater contain high concentrations of organic substances, such as sugar, pectin, and protein, and expends a high amount of dissolved oxygen (DO) when released to rivers directly, thus creating putrid water and deteriorating the water quality [1]. Because molasses wastewater features high organic loading (OL) and intense chroma and is biodegradable, biological treatment methods can be applied to remove the organic pollutants [2–4]. Previous research applied a baffled reactor (BR) to treat molasses wastewater and effectively reduced the COD of a wastewater sample with an OL of 1.5–3.6 kg/m^3/d by $>$90% [5]. First proposed by Bachmann et al. [6], a BR is a highly effective anaerobic reactor that involves multiple separation baffles to divide the reactor into a number of chambers. When wastewater flows through the reactor, the separation baffles disturb the flow of the water, enabling it to be evenly mixed in each chamber. In addition, the baffles decrease the horizontal flow rate of the sludge, thus facilitating sludge deposition in the chambers and preventing it from being mixed with the outflow water. Consequently, the hydraulic retention time (HRT) of the wastewater is increased. This enables the microbes in the chambers to fully react with and decompose the pollutants, thereby increasing the wastewater removal efficiency [7,8].

A microbial fuel cell (MFC) adopts microbial catalytic reactions to convert the chemical energy of organic matter to electrical energy; hence, it is an emergent technology that functions as both a pollutant remover and power generator [9,10]. An MFC is mainly composed of an anode, a cathode, and a proton exchange membrane (PEM) and involves oxidation–reduction reactions between organic matter and microbes [11]. A PEM transmits protons and blocks fuels from the anode and oxygen from the cathode [12,13]. Therefore, how to maintain the moisture of a PEM and facilitate proton transmission is a key concern of MFC technology [14,15]. Currently, most MFCs adopt Nafion® as the PEM. However, this poly-

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mer is costly (*e.g.*, 1733 US$/m^2 for Nafion 117) and thus might impede the applications and development of MFC technology [16,17].

In recent years, less expensive alternatives have been developed to replace Nafion, including polyvinylidenefluoride (PVDF)-based [18] and polytetrafluoroethylene (PTFE)-based [19,20] ion exchange membranes. Because PVDF and PTFT are hydrophobic, they are converted to hydrophilic substances through chemical modification in order to facilitate proton transmission, thereby increasing the manufacturing costs. Numerous researchers have attempted to combine a PEM with a cathode to form a member electrode assembly (MEA), which has been reported to increase the power generation efficiency by decreasing the mass transfer limitation of protons and air and by reducing the cathode reaction time [17,21–22]. Moreover, the MFC characteristics and assembly methods are essential to the power generation efficiency [23,24]. For example, Zhong et al. [7] utilized the height differences between the separation plates to create varying water displacement heights in an anaerobic baffled stacking MFC, enabling each chamber of the MFC to function as an independent cell. The independent cells were then connected in series to enhance the voltage output of the MFC.

In this study, a polyvinyl alcohol hydrogel (PVA-H), which exhibits excellent insulating, water-absorption, and water-retention capabilities, was converted to a PVA-hydrogel elastomer (PVA-HE) through the freeze–thaw method [25]. Subsequently, it was combined with carbon cloth (serving as the cathode) to press-form an MEA, named the PVA-hydrogel elastomer membrane electrode assembly (PVA-HEM). This membrane was used to block fuels from the anode and facilitate the oxygen transfer from atmosphere to the cathode. An existing BR [26] was integrated with multiple units of PVA-HEM and MFC to develop a novel PVA-HEM-BR-MFC reactor (called HEM-MFC). Moreover, the tilt angle of the reactor was adjusted to create water displacement differences between each reactor chamber, thereby enabling each chamber to function as an independent MFC. The MFCs were then connected in series to increase the overall voltage output.

2. Material and methods

2.1. Consortia and inoculation

The bacterial strain came from the activated sludge in the wastewater treatment plant of a winery manufacturing plant and was acclimated in the laboratory for more than six months by using molasses wastewater as the sole carbon source to cultivate a mixed strain capable of degrading molasses wastewater. During the acclimation process, a bottle of 5 L serum solution (containing 3 L of inorganic salt solution as the buffer) was employed to provide the micronutrients necessary for the bacteria growth [27]. Additionally, molasses wastewater (COD = 1500 mg/L) was added to the bacteria once per seven days to provide the carbon source.

2.2. Preparation of air-cathode membrane electrode assembly

The PVA-HE was used to construct two types of membrane electrode, namely the plan form PVA-HEM (P-PVA-HEM) and rod form PVA-HEM (R-PVA-HEM). Regarding the preparation of P-PVA-HEM, a circular plastic container (diameter = 3 in) with six holes (diameter = 0.8 cm) on the bottom was used. The holes were sealed, and then a 13% PVA hydrogel (PVA-H) solution was loaded into the container to a height of 2 cm. After the bubbles were removed, the solution was frozen for six h at −30 °C and then recovered to room temperature. Next, a piece of carbon cloth was adhered to the PVA-H, which had yet to fully transform to the PVA-HE, and press-formed [28,29]. The resulting membrane was subjected to another freeze–thaw process to create a P-PVA-HEM (Fig. 1(a)). To prepare an R-PVA-HEM, the 13% PVA-H solution was loaded into an acrylic column (diameter = 10 cm; height = 10 cm) until a height of 2 cm. The remaining procedure was identical to that for preparing a P-PVA-HEM (Fig. 1(b)).

2.3. Construction of HEM-MFC

The main structure of the proposed HEM-MFC reactor involved an acrylic container divided into four chambers of 9.7 cm × 10 cm × 18.5 cm. The top and bottom vertical baffles were 17.5 and 15.5 cm high, respectively, and were placed 1 cm apart from each other (Fig. 2). Furthermore, each chamber employed a 4 cm × 3 cm carbon felt as the anode. After the PVA-HEM was inserted into each chamber, a titanium wire was used to connect the cathode and anode. Finally, a 1 kΩ external resistance was applied to facilitate the movement of electrons from the anode to the cathode, thus enabling an oxygen reduction reaction to generate an electrical current.

2.4. Operation of HEM-MFC

The tilt angle of the HEM-MFC was adjusted to differ the water displacement height and evaluate how it affects the molasses wastewater removal efficiency (COD = 1500 mg/L) and power generation capability of the MFCs. During the experiment, sponge pads were used to tilt the reactor, creating a tilt angle of 0°, 15°, 20°, or 25°; this adjusted the differences between the water placement height of the chambers to be 0, 2.5, 3, and 4.3 cm, respectively. Moreover, a peristaltic pump MU-D01 (Major Science, Taiwan) was adopted to control the inflow rate of wastewater and configure the HRT of the MFCs to be 5, 10, and 15 h, thus assessing the effect of HRT on the COD removal efficiency and power generation of the MFCs.

2.5. Analytical methods

The water quality analyzer DR980 (HACH Co., USA) was used to measure the COD levels of the inflow and outflow wastewater samples. The UV/VIS spectrometer Opron-3000 (Hanson Technology, Korea) was employed to determine the chroma levels of the water samples. Before a sample was analyzed, it was centrifuged at 6000 rpm for 5 min to deposit the sludge and impurities. The supernatant was acquired and analyzed at 450 nm [30]. The pH value was measured with the pH meter PH500 (Clean Instrument Co. Taiwan).

Voltage measurements of the MFC were performed with a digital electronic multimeter (digital multimeter/data acquisition system, Model 7700/2200, Keithley, USA). The output voltage was recorded every 10 min, and the Ohm's Law (V = IR) was used to determine the current value when the external resistance was set to 1 kΩ. Next, the current value was multiplied by the voltage to determine the power, which was then divided by the anode surface area (A) to acquire the power density (PD = P/A). Similarly, the current density (CD = I/A) was calculated by dividing the current by the anode area. The power density-polarization curves were plotted using the current density, PD, and output voltages under various conditions. The slope of a polarization curve can be used to determine the internal resistance of an MFC.

3. Results and discussion

3.1. Effect of membrane electrode on COD removal efficiency, decolorization, and power generation

In this study, two types of air-cathode membrane electrode (P-PVA-HEM and R-PVA-HEM) were used to assemble two types