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Modifying proton exchange membrane in a microbial fuel cell by adding clay mineral to improve electricity generation without reducing removal of toluene

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ABSTRACT

This work develops a tubular air-cathode microbial fuel cell (T-AC-MFC) by modifying the proton exchange membrane (PEM) using a combination of polyvinyl alcohol hydrogel (PVA-H) and light expanded clay aggregate (clay). The modified PEM was then incorporated into a cathode to form a membrane electrode assembly (MEA) to promote electricity generation. The electrochemical performance of PEM_{PVA-H-clay} was the best among three types of tested PEMs. The proton conductivity of PEM_{PVA-H-clay} was 2.87-fold that of PEM_{PVA-H}, indicating that adding clay mineral is an important modification of the PEM. PEM_{PVA-H-clay} exhibited a high proton conductivity of 3.786×10^{-5} M/d at a PVA/clay powder ratio and clay powder size of 10:1 and 150 μm , respectively. The current density that was generated by our MFC using PEM_{PVA-H-clay} was 35 times those found in the literature. Fifty mg/L of toluene was completely degraded in T-AC-MFC that was continuously operated for six days, and a maximum closed-circuit voltage of 285.63 mV as well as a power density of 25.14 mW/m² were obtained. Our T-AC-MFC exhibited the highest efficiency of removal of toluene (>99%) and a power density that was 1.16–9.67-fold those of other MFCs in the literature.

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1. Introduction

A proton exchange membrane (PEM) functions as a separator between anode and cathode in microbial fuel cells (MFCs) and is often used to transport protons. An effective PEM has a high ionic conductivity and high mechanical strength; supports stable electrochemical ionic transport, and is compatible with both the cathode and the anode. A PEM comprises a dry solid polymer, hydrogel, a membrane polymer, or a composite polymer [1,2]. Of these materials, hydrogel (such as Nafion) reportedly has the best ionic conductivity and mechanic strength [3], and thus has a potentially positive effect on voltage output. To maintain electrochemical ionic transport stability, composite materials (such as ZrO₂, TiO₂ and SiO₂) are added to the PEM, reportedly improving ionic conductivity and compatibility with the electrodes [4–7].

Polyvinyl alcohol hydrogel (PVA-H) as a proton conductor for use in a PEM in an MFC may be cost-effective alternative to Nafion, and has been widely used in recent years [8–10]. PVA-H can be obtained by a physical crosslinking reaction, which is induced by freezing and thawing [11–14]. However, insufficient water uptake and retention by PVA-H result in a low proton conductivity [15–18]. Vermiculite is the common and more readily available soil amendments for improving water retention; hydroxy-interlayered vermiculite reportedly has potential to selectively retain K⁺ and NH₄⁺ [19]. Light expanded clay aggregate (simply called “clay”) consists of small, lightweight, bloated particles of burnt clay, and is characterized by its high porosity, high water absorbency and retentive capacity [20]. Thus, clay is used in many applications, such as a proper construction material for flooring and roofing, a filter media for growth of microbes on wastewater treatment, and agriculture [21]. Vermiculite and clay reportedly have a high water uptake and retention [22,23], and so are added to PVA-H to improve its water uptake and retention.

The modified PEM from PVA-H was integrated herein with a cathode to form a membrane electrode assembly (MEA) in an MFC to improve the generation of electricity. The goal of this inves-

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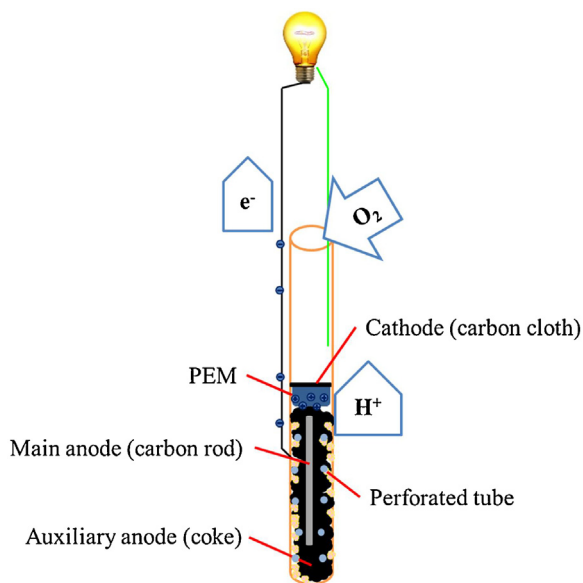


Fig. 1. Schematic diagram of T-AC-MFC.

tigation is to develop a tubular air-cathode microbial fuel cell (T-AC-MFC) by modifying a proton exchange membrane (PEM) using polyvinyl alcohol hydrogel (PVA-H) and clay mineral. The innovative T-AC-MFC, with high proton conductivity, is used to study the enhancement in treating toluene-contained water by the modification of the generation of electricity and toluene removal efficiency of the MFC. A new T-AC-MFC is developed future use in the remediation of contaminated ground water.

2. Material and methods

2.1. Configuration of microbial fuel cell

The developed T-AC-MFC comprised a PVC pipe (diameter: 4 cm; length: 14 cm), as depicted in Fig. 1. The anode chamber (length: 6 cm; volume: 75 cm³), the cathode, and the PEM all had a diameter of 4 cm. Conductive coke grains with a size of 270 μm (60 g, act an auxiliary electrode material), and 10% PVA-H (25 g as adhesive) were blended into a PVA-coke slurry mixture. The mixture was placed in the anode chamber, and a carbon rod (the main electrode) was inserted into the center of the chamber in full contact with the coke to receive electrons. The chamber, which contained the PVA-coke slurry mixture, was heated at 50 °C to cause the coke to bond to the PVA to form a stable, solid structure. The chamber was drilled with holes to facilitate the attachment of microorganisms in water to the anode.

PEMs were fabricated by mixing PVA (molecular weight = 105,600 g/mol) with deionized water; allowing the mixture to form a 10% PVA gel in an autoclave; cooling to 25 °C; refrigerating it at −30 °C for one day, and allowing it to thaw at room temperature. Then, the PVA underwent cross-linking reactions with water molecules to form a three-dimensional crystal structure (PEM_{PVA-H}) [24,25]. PEM_{PVA-M} was prepared by heating PVA-M (polyvinyl alcohol membrane) in an oven at 50 °C for one day, removing the membrane and leaving it at room temperature to yield a 1 cm-thick PVA sample that did not undergo any cross-linking reaction. PEM_{PVA-M} was used as a control to test proton conductivity for different types of PEMs by adding clay or vermiculite. The MEA in the MFC had a carbon cloth as the cathode, which was pressed into PEMs to form a unified whole. Therefore, the MEA received protons from the anode via PEMs and used oxygen in the

atmosphere to induce reduction reactions in the cathode to produce H₂O, causing the MFC to generate current.

2.2. Bacterial inoculum and nutrient media

A bacterial consortium was obtained from a petrochemical wastewater treatment plant in Central Taiwan and acclimated in a 1.5 L bioreactor that was fed with toluene as the sole carbon source for six months. The mixed culture was then transferred to an anode chamber, in which solution was sparged with nitrogen to maintain the dissolved oxygen level below 1 mg/L. Each liter of the mineral salt solution in the anode chamber was prepared as follow: K₂HPO₄ 1750 mg, KH₂PO₄ 2145 mg, MgCl₂·6H₂O 100 mg, FeCl₃·6H₂O 1 mg, CaCl₂ 45 mg, NH₄Cl 10 mg, CuCl₂·2H₂O 0.25 mg, CoCl₂·6H₂O 0.25 mg, ZnCl₂ 1 mg, MnCl₂·4H₂O 1 mg, Na₂MoO₄·2H₂O 0.1 mg, and NiCl₂·6H₂O 0.02 mg.

2.3. Chemical analysis

Gaseous samples were periodically taken from the headspace of the anode chamber of MFC. The remaining toluene was analyzed using a GC apparatus that was equipped with a flame ionization detector (GC-FID, Shimadzu, GC-2014, Japan) and a Stabilwax column (30 m × 0.53 mm id × 1 μm film thickness, Restek, USA). The temperature of GC oven was set to 105 °C. The injector and detector temperatures were set to 200 °C and 250 °C, respectively. A set of toluene (99.9%, Fisher Scientific, USA) of analytic grade standard solutions with concentrations from 20 to 70 mg/L were used to plot a calibration curve. This curve enabled the integrals of the sample peaks on the chromatograms to be converted to concentrations.

2.4. Fabrication of PEM, water absorbency and proton transfer of PEM

The clay used was a commercial product, containing 60% SiO₂, 20% Al₂O₃, 8% Fe₂O₃, and ~12% CaO & MgO (Jongkind Grond B.V. Co, Netherlands). The vermiculite employed was a commercial product, containing 43% SiO₂, 19% Al₂O₃, 14% Fe₂O₃, 13% MgO, and ~11% CaO & K₂O (Nan Hai Co., Taiwan). Vermiculite can selectively absorb anions and can absorb large amounts of water [23]. Clay is porous, highly permeable, and absorbent, but cannot absorb ions [20]. In this investigation, vermiculite and clay were mixed with PVA-H to form PEM_{PVA-H-vermiculite} and PEM_{PVA-H-clay}, respectively. Both were used as PEMs with improved water retention to transport protons through water. Various ratios of PVA-H to clay or vermiculite (10:1, 15:1, and 20:1) were prepared to yield PEMs. The proton transfer and water absorbency of each membrane were analyzed.

An experiment on proton transfer through the PEM was performed using a two-chamber MFC that was constructed in the traditional “H” shape, comprising two bottles (diameter: 4 cm) that were joined by a glass tube that contained a PEM. 500 mL of 0.01 M hydrochloric acid (HCl) was added to the first bottle and 500 mL of DI water (pH=6.9) was added to the second. From the bottle that contained HCl were transported protons to the bottle that contained DI water, which had a lower proton concentration gradient. The pH of DI water was measured as a function of time using a pH meter (SX751, Major science, USA).

The water content of PVA directly influences its ability to transfer protons; therefore, the water absorbency of PVA was measured. The water absorbency values of PEMs that had been placed in 100 mL of DI water for 14 days were obtained [26].

2.5. Electrochemical analysis

The voltage of MFC was measured every 10 min using a multi-digital meter (CHY-48R, CHY, Taiwan). The polarization curves and

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