



Power output of microbial fuel cell emphasizing interaction of anodic binder with bacteria

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HIGHLIGHTS

- PBE is a promising anodic binder for the power output improvement of MFCs.
- This improvement is emphasized by the strong interaction of PBE with bacteria.
- This interaction is more crucial for forming an EAB than hydrophilicity.

ARTICLE INFO

Keywords:

Poly(bisphenol A-co-epichlorohydrin)
Anodic binder
Microbial fuel cell
Interaction with bacteria

ABSTRACT

Electrochemically active biofilm is necessary for the electron transfer between bacteria and anodic electrode in microbial fuel cells and selecting the type of anodic electrode material that favours formation of electrochemically active biofilm is crucial for the microbial fuel cell operation. We report a new finding that the interaction of anodic binder with bacteria plays more important role than its hydrophilicity for forming an electrochemically active biofilm, which is emphasized by applying poly(bisphenol A-co-epichlorohydrin) as an anodic binder of the microbial fuel cell based on carbon nanotubes as anodic electrode and *Escherichia coli* as bacterium. The physical characterizations and electrochemical measurements demonstrate that poly(bisphenol A-co-epichlorohydrin) exhibits a strong interaction with bacteria and thus provides the microbial fuel cell with excellent power density output. The MFC using poly(bisphenol A-co-epichlorohydrin) reaches a maximum power density output of 3.8 W m^{-2} . This value is larger than that of the MFCs using polytetrafluoroethylene that has poorer hydrophilicity, or polyvinyl alcohol that has better hydrophilicity but exhibits weaker interaction with bacteria than poly(bisphenol A-co-epichlorohydrin).

1. Introduction

Fossil fuel depletion and environmental pollution increase the urgency for new reliable energy sources to attain sustainable development [1,2]. Microbial fuel cells (MFCs) are ideal energy converters because they can convert organic matters into electrical energy [3–7]. However, the lower power of MFCs restricts their practical applications unless they are connected as stacks [8,9].

MFC is composed of anodic and cathodic electrodes and an electrolyte (membrane). Electrons are generated at the anodic electrode through the anaerobic oxidation of substrates in electrolyte, and then flow through an external circuit to the cathodic electrode where they

combine with protons and oxygen to form water [10]. Many factors affect the power density output of the MFCs, one of which is the electron abstraction by microbes to anodic electrode [11–14]. There are different pathways to electron abstraction by microbes, including mediator or extracellular electron transfer (EET) [9]. EET mechanism is usually the limited process for power density output of MFC and enhancing its kinetics is necessary to improve the power density output of the MFCs.

Various nanomaterials including graphene [15,16], carbon nanotubes [17,18], MWCNTs/SnO₂ [19], ZnO/Au [20], Mo₂C composites [21,22], and conductive polymers such as polyaniline [23–25], polypyrrole [26–28] and polydopamine [29] have been developed to

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accelerate the anodic kinetics. For example, the application of carbon nanotubes generated an improvement of ~ 6 fold power density compared to the graphite electrode [17]; polypyrrole-modified MnO_2 electrodes exhibited excellent power density and durability [28]. These efforts mainly emphasized on the electrode materials, but less attention was paid to the contribution of the anodic binder.

Whatever anodic material is used, an electrochemically active bio-film (EAB) is required for the EET mechanism to take place. Since the electrolyte solution for MFCs is aqueous, the hydrophilicity of the anodic electrode should be beneficial for the EAB formation. Poly(tetrafluoroethylene) (PTFE) was usually used in MFCs as an anodic binder, but its hydrophobicity is not beneficial for the EAB formation on anodic electrode [30,31]. In our previous report, we found that the power density output of a MFC could be enhanced by substituting polyvinyl alcohol (PVA) for PTFE, which was mainly attributed to the improved hydrophilicity of PVA compared to PTFE [32].

In this work, we reported a new finding that the interaction of an anodic binder with bacteria played a more important role than its hydrophilicity for the EAB formation on anodic electrode. Poly(bisphenol A-co-epichlorohydrin) (PBE), an inexpensive and commercially available polymer that possessed good adhesive properties and had been successfully used as a cathodic binder [33], was considered as an anodic binder. Through electrochemical measurements, spectroscopic analyses and theoretical calculations, it was demonstrated that PBE was less hydrophilic but provided MFC with higher power density output due to its stronger interaction with bacteria than PVA as the anodic binder. In the evaluation of MFC performances, carbon nanotubes were used as anodic electrocatalyst and *Escherichia coli* (*E. coli*) as bacterium. *E. coli* was considered because it is a representative bacterium mainly based on EET mechanism [9].

2. Experimental

2.1. Anodic electrode preparation

Poly(bisphenol A-co-epichlorohydrin) (PBE, Aladdin Industrial Corporation) was dispersed in deionized water under sonication. Carbon nanotubes (2 mg cm^{-2} , CNTs, Chengdu Organic Chem. Co., Ltd., China) were added into the solution with various contents of PBE, and then dispersed by sonication. The resulting slurry was coated on carbon felt (Beijing Carbon Sci. Tech. Co., Ltd.) to prepare anodic electrodes ($3.0 \text{ cm} \times 3.0 \text{ cm}$) [34]. An anodic electrode with 5% PTFE (optimal loading content as reported in the literature [30]) as the binder was also prepared for a comparison. Before use, CNTs were purified with a solution of concentrated HNO_3 and HCl (1:3 in volume) at 140°C for 14 h, while the carbon felts were treated with 1 M HCl and then with 1 M NaOH , followed by washing with deionized water and drying in vacuum at 60°C for 10 h.

2.2. Bacteria cultivation

E. Coli DH5 α (Biovector NTCC Inc., China) was cultivated in a standard Luria-Bertani medium, consisting of 10 g peptone, 5 g yeast extract and 10 g sodium chloride in 1 L deionized water, at 38°C in HPX-9052 incubator (Boxun, China). After cultivation for 16 h, *E. coli* was kept at 4°C in the fridge space for further use.

2.3. MFC set-up

MFCs were constructed using poly (methyl methacrylate) chambers (Fig. S1, $5.0 \times 4.0 \times 5.0 \text{ cm}$) and a membrane cathode used as counter electrode for various electrochemical measurements. The membrane cathode was assembled by loading 0.2 mg cm^{-2} Pt/C (20% Pt, Inno Chem. Sci. Tech. Co., Ltd., China) as the electrocatalyst on carbon paper (Hesen, China). The electrocatalyst was mixed with polyvinylidene fluoride (PVDF) (65:15, in weight) in N-methyl-2-pyrrolidone (NMP).

The slurry was pasted on carbon paper, and hot-pressed on the cation exchange membrane (Iontech Environ. Tech. Co., Ltd., China) [35].

Anolyte was phosphate-buffered basal medium (PBBM), 0.1 g KCl , 5.8 g NaCl , 0.25 g NH_4Cl , 10 mL trace mineral solution, and 10 mL vitamin solution, in 1 L of phosphate buffer (50 mM, $\text{pH} = 7.0$). Glucose (2 g L^{-1}) was used as an electron donor. All the materials before use were sterilized in autoclaves (DSX-280B, China) at 120°C for 30 min and the chamber of the MFC was inoculated with 10 mL cell suspension and PBBM at 30°C in the incubator (HPG-280H, China) [36].

2.4. Characterizations and calculations

All the electrochemical measurements were carried out on Solartron 1480 (England) in the MFCs containing anodic electrode, air cathode and saturated calomel electrode (SCE). To ensure the accuracy of the experimental results, all the electrochemical measurements were performed with five MFCs for every sample and the reported results in this work were the mean values of five MFCs. Power outputs of the MFCs were obtained by linear sweep voltammetry with the anodic electrode as working electrode, SCE as reference electrode, and the cathode as counter electrode, from open circuit potential to 0 V (vs. SCE) at 1 mV s^{-1} . Before measurements, the MFCs were inoculated in PBBM containing glucose (2 g L^{-1}) and *E. coli* for three days at room temperature. The power density, P (W m^{-2} , Watts per footprint area of anodic electrode), was obtained according to $P_A = IU/S$, where I (A) stands for the current, U (V) for voltage between the working electrode and air cathode, and S (m^2) for the projected area of the working electrode [37]. Electrochemical impedance spectroscopy was carried out at open circuit potential with a sine signal of 5 mV from 10^5 to 0.01 Hz on the MFCs after inoculation in PBBM containing glucose (2 g L^{-1}) and *E. coli* for three days. Chronoamperometry was performed under a constant potential of 0.1 V (vs. SCE) from the inoculation beginning of the MFCs in PBBM containing glucose (2 g L^{-1}) and *E. coli*. The operation was in batch with three days per batch and 80% of anolyte was replaced by the PBBM containing only glucose for a new batch. Chronopotentiometry was performed under a constant current of 0.1 μA after the MFCs were inoculated in PBBM containing glucose (2 g L^{-1}) and *E. coli* for three days at room temperature.

Scanning electron microscopy (SEM, JSM-6510, Japan) was used to observe the surface morphologies of the materials. The hydrophilicity of the materials was determined by measuring contact angle on JC2000C (China). In the measurement of contact angle, deionized water (10 μL) was dropped on the films that were prepared by coating the binder mixture on a glass and then drying at 50°C . Molecular structure was identified by Fourier transform infrared spectroscopy (FTIR, BRUKER TENSOR 27, Germany) within from 4000 to 400 cm^{-1} . Thermal stability was determined by a thermogravimetric analyser (TGA, Perkin-Elmer TGA7, America).

All the calculations were carried out using the Gaussian 09 package [38]. To optimize the equilibrium structures of molecules, M06-2X level in conjunction with the 6-311 + G(d) level basis set was adopted [39]. The effect of bulk solvent (dielectric constant: 78.5) was investigated by polarized continuum models (PCM).

The interaction energy (E_i) was obtained based on Eq. (1).

$$E_i = E[\text{C}_2\text{F}_4 \cdots \text{C}_6\text{H}_{12}\text{O}_6] - E(\text{C}_6\text{H}_{12}\text{O}_6) - E(\text{CF}_4) \quad (1)$$

Where $E(\text{C}_6\text{H}_{12}\text{O}_6)$, $E(\text{CF}_4)$, and $E[\text{CF}_4 \cdots \text{C}_6\text{H}_{12}\text{O}_6]$, were the total energy with the counterpoise correction for the isolated $\text{C}_6\text{H}_{12}\text{O}_6$ molecule, the CF_4 , and the corresponding $\text{CF}_4 \cdots \text{C}_6\text{H}_{12}\text{O}_6$ [40]. The charges on oxygen atom were obtained by fitting the electrostatic potential of molecule (CHELPG method) [41].

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