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Microbial fuel cell as a biocapacitor by using pseudo-capacitive anode materials

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HIGHLIGHTS

- Microbial fuel cell as a biocapacitor by using pseudo-capacitive anode materials.
- The biocapacitor stores electrons generated from microbial oxidation of substrate.
- The average power significantly increases if the stored energy is shortly dissipated.
- The charge/energy stored and released is highly dependent on the anode capacitance.

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ABSTRACT

Here, we report that the microbial fuel cell (MFC) containing pseudo-capacitive anode materials such as polypyrrole (PPy)/9,10-anthraquinone-2-sulfonic acid sodium salt (AQS) composite films and RuO_2 nanoparticles can function as a biocapacitor, able to store bioelectrons generated from microbial oxidation of substrate and release the accumulated charge upon requirement. Influences of the specific capacitance of the PPy/AQS- and RuO_2-modified carbon felt anodes on the extent of accumulated charge are examined. Results show that increasing anode capacitance is responsible for the increases in the amount of electrons stored and released, and thereby leading to more energy stored and average power dissipated. The long-term charging–discharging tests indicate that the RuO_2-modified biocapacitor with a specific capacitance of 3.74 F cm^{-2} exhibits 6% loss in the amount of released charge over 10 cycles for one-month operation, and 40% loss over 60 cycles for six-month operation. Our findings suggest that the MFC anode incorporating pseudo-capacitive materials shows potential for storing energy from waste organic matter and releasing in a short time of high power to the electronic device.

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1. Introduction

Microbial fuel cells (MFCs) are attractive for environmental-friendly sustainable power applications, because they are able to produce electricity by oxidizing a variety of organic substrates available in wastewaters with microorganisms. Due to the sluggish anode kinetics and the limited organic concentrations in wastewaters, the power generated from MFCs is orders of magnitude smaller than that associated with hydrogen-based fuel cells [1,2]. Many of the studies to date have concentrated on improving anode performance by the search of effective anode materials [3–8] and microorganisms [9,10] in order to boost the power. However, the resulting power was still limited with respect to the power demand of most electronic devices [11,12]. To fill the gap, some researchers [11,13,14] have proposed an alternative solution by which the energy generated in MFCs are first accumulated in an external capacitor and then dissipated in a short time of high power to the electronic device. Their results have shown that the use of capacitor allowed more energy harvested when MFCs were operated intermittently rather than continuously.

Instead of the use of the external capacitor, a recent study [15] has developed an integrated anode system that incorporated capacitive material into the bioanode, leading to the conclusion that the capacitive anode performed better than the noncapacitive

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counterpart in terms of more charge stored during the charge–discharge experiment. This novel concept makes the anode function as a biocapacitor which shares the same electrolyte, membrane and counter electrode (cathode) with the MFC. Thus, the MFC under such a case can not only generate bioelectricity, but also store and release energy. Fig. 1 illustrates the concept of using an MFC as the biocapacitor in which the anode accumulates electrons from bacteria for charging when switch is open and releases electrons to the external circuit for discharging when switch is closed.

Deeke and his coworkers [15] prepared the capacitive anode by adding activated carbon powder and polymers including NMP (*N*-methyl-2-pyrrolidone) and PVDF (poly(vinylidene fluoride)) to the current collector. However, these may not be the optimal anode materials due to their relatively low capacitance and low electrical conductivity. Indeed, pseudo-capacitive materials including transition metal oxides like RuO_2 and conductive polymers like polypyrrole (PPy) are more appropriate for being used in the anode owing to their fast and reversible redox behavior, large surface area, and favorable metallic conductivity [16,17]. It is well recognized that their capacitance is typically 2–3 times larger than that of the carbon based materials [18]. More importantly, our previous studies [19,20] have demonstrated that they can interact well with the microorganisms and promote electron transfer from the microorganisms to the anode. In particular, it is of great concern about how the anode capacitance affects the amounts of charge stored and how the long-term operation affects performance of the capacitive materials, because these are important considerations for selection of anode materials and optimization of experimental design.

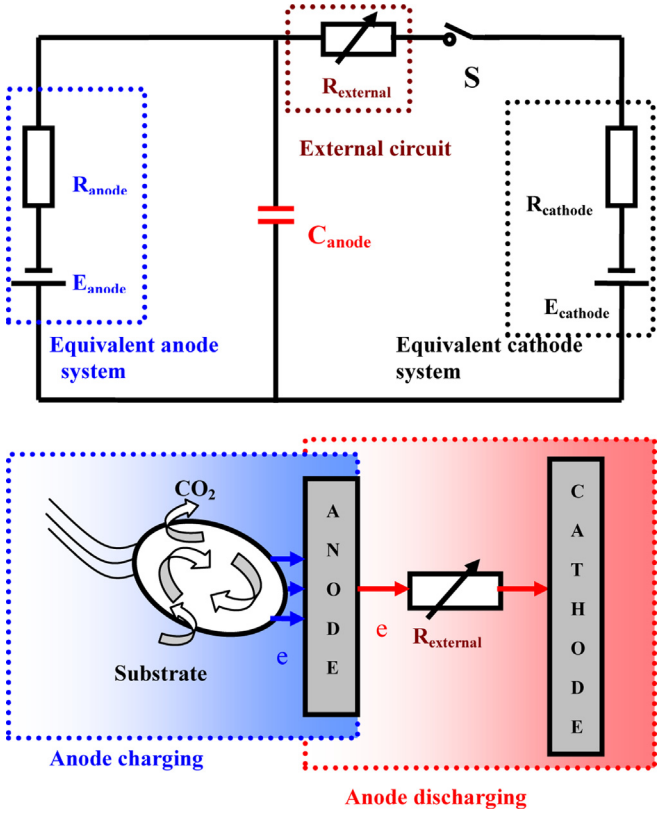

Fig. 1. Equivalent circuit of an MFC containing a capacitive anode. It consists of an equivalent anode system, an equivalent cathode system and an external circuit. R_{anode} and $R_{cathode}$ represent the sum of charge-transfer resistance, diffusion resistance and ohmic resistance at the anode and cathode, respectively. The capacitive anode stores electrons from bacteria when switch S is open and releases electrons to the external circuit when switch S is closed.

The present study thus aimed to verify the ability of the anode containing pseudo-capacitive materials to function as a biocapacitor, which can store bioelectrons generated from microbial oxidation of substrate and release the accumulated charge during the discharge experiment. To meet this goal, a series of polypyrrole (PPy)/9,10-anthraquinone-2-sulfonic acid sodium salt (AQS)- and RuO_2-modified carbon felt anodes were prepared and the influences of their specific capacitance on the extent of accumulated charge were evaluated. The energy stored in the biocapacitor was calculated and further calculation of the average power density was performed given that the energy was dissipated in a short time. In addition, attention was devoted to investigate the cycling durability of the biocapacitor by running the long-term tests with repeatable charging and discharging.

2. Experimental

2.1. Preparation of pseudo-capacitive anode materials

RuO_2 and conducting polymer are widely used in supercapacitor applications due to their electrical conductivity and reversible redox reactions. Here, PPy-modified and RuO_2-modified carbon felt electrodes as the anodes were prepared according to the procedures described in our previous work [19,20]. The starting anode substrate was a piece of carbon felt (3.0 cm × 2.0 cm × 0.5 cm) that was cleaned in a hot H_2O_2 (10%, 90 °C) solution for 3 h, followed by thorough rinse with deionized water and dry at 60 °C. A Ti wire (1 mm in diameter) was inserted inside the carbon felt to allow the external circuit connection. The room-temperature electrodeposition was performed in a three-electrode electrochemical cell including a working electrode (carbon felt), a reference electrode (saturated calomel electrode, SCE) and a counter electrode (Pt mesh). It should be noted that all the potentials reported throughout this paper were referred to SCE, if not otherwise stated. To deposit AQS doped-PPy films on the carbon felt, a constant potential of 0.8 V was applied for the anodic electropolymerization in a non-stirred N_2-saturated solution that contained 0.1 M pyrrole monomer and 5 mM AQS. RuO_2-modified carbon felt samples were prepared from a $RuCl_3$ (0.05 M) solution under the galvanostatic condition by applying a constant current density of 5 mA cm^{-2} (normalized to the projected area of electrode) which was controlled by a potentiostat (CHI 660C, Shanghai CH Instrument Company, China). The difference in capacitance among these modifying materials was controlled by varying the total charge passed to the working electrode. The freshly prepared RuO_2-modified and PPy/AQS-modified carbon felt anodes were then thoroughly rinsed with distilled water and air-dried at room temperature.

The capacitance of the synthesized materials was examined through the galvanostatic charge–discharge tests that were performed at a current load of 0.5 mA cm^{-2} within the potential window from −0.6 to 0.3 V. The EIS measurements on resistance of the anode materials were performed by recording the impedance spectra at the open circuit potential (OCP) and in the frequency range from 10,000 to 0.01 Hz with a sinusoidal excitation signal of 10 mV. All these electrochemical tests were carried out in 0.1 M Na_2SO_4 solution using the same three-electrode electrochemical cell as described above.

2.2. MFC construction, operation and tests

The two-chamber MFC made of Perspex flames was fabricated as described previously [19–21], consisting of the modified carbon felt anode and a bare carbon felt cathode (3.0 cm × 2.0 cm × 0.5 cm) in each chamber. The two chambers were separated by a cation exchange membrane (Zhejiang Qianqiu Group Co., Ltd. China) with

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