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Original Research

Enhance Soil Remediation and Power Generation Capabilities of Plant Microbial Fuel Cells through PANI-CeO_2 Modified Anode

Liping Fan[1*], **Naijia Wang**[1, 2]

[1]Key Laboratory of Collaborative Control and Optimization Technology of Industrial Environment and Resource of Liaoning province, Shenyang University of Chemical Technology, Shenyang, 110142, China
[2]College of Environment and Safety Engineering, Shenyang University of Chemical Technology, Shenyang, 110142, China

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Abstract

Heavy metal pollution in soil has become a serious problem affecting crop cultivation and human health. Plant microbial fuel cells (PMFCs) have great prospects in soil remediation and regenerative power generation. In order to improve the ability of PMFCs to decontaminate heavy metal contaminated soil and generate electricity simultaneously, a plant microbial fuel cell (PMFC) experimental system was constructed by using polyaniline and nanocerium oxide modified electrodes (PANI-CeO_2/CF) as anodes, cadmium contaminated soil as substrate, and acclimated sludge as electrogenesis bacteria, and a flowerpot with epipremnum aureum as the reactor. Scanning electron microscopy (SEM), electrochemical impedance spectroscopy (EIS), cyclic voltammetry (CV), and Cd removal rate were used to characterize the electricity generation and soil remediation properties. The experimental results show that the removal rates of Cd in soil by PMFC using the PANI-CeO_2/CF anode were 93.91%. Compared with the PMFC with the conventional CF anode, the output voltage of the PMFC with the PANI-CeO_2 modified anode was increased by 307.2%, the equivalent internal resistance was reduced by 38.4%, and the Cd removal rate was increased by 53.8%. The anode modification based PANI-CeO_2 significantly improved the power generation capacity and soil remediation effect of PMFC.

Keywords: Plant microbial fuel cell, soil remediation, power generation, anode modification, cadmium contaminated soil

Introduction

With the development of human society and economic prosperity, demand for energy is also constantly increasing. Traditional energy sources mainly rely on coal, oil, and natural gas. However, these fossil fuels not only cause serious environmental pollution, but also have limited resources. With excessive exploitation and utilization in recent years, fossil fuels are gradually becoming depleted. In order to realize the sustainable development of human society, vigorously developing renewable energy technology is the key solution [1-3].

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*e-mail: fanliping@syuct.edu.cn;
Tel.: +86-24-89386011;
Fax: +86-24-89386011

On the other hand, heavy metal pollution in soil has become an increasingly important environmental issue in the world today [4-6]. Due to the inability of heavy metals to be biodegradable, the pollution they cause is usually more persistent than organic pollutants. Especially cadmium (Cd), due to its characteristics of stability, enrichment, easy absorption by plants, strong migration, and difficulty in degradation, has become the first polluting element in soil in certain regions [7-9]. After continuous accumulation in the soil, Cd is easy to enter the food chain through enrichment, which not only poses a threat to plants and animal systems, but also causes great harm to human health. The environmental risks caused by Cd to food crops have been reported for many times [10-12]. In order to realize the sustainable development of agricultural production and the healthy survival of human beings, it is necessary to suppress cadmium pollution in soil. However, traditional technologies such as physical remediation, chemical remediation, and bioremediation are costly and prone to secondary pollution [13-15].

Plant microbial fuel cell (PMFC) is an emerging biotechnology device that combines plants and microorganisms. PMFC uses the photosynthesis of plants to convert carbon in the atmosphere into energy-rich organic matter in the roots and release oxygen, and the energy-rich organic matter is then oxidized by rhizosphere bacteria to produce electricity. This makes PMFC a generator that meets the requirements of carbon neutrality [16, 17]. The PMFC forms a soil–plant-microbe complex zone that can promote microbial activity in the rhizosphere, thereby enhancing water regulation, nutrient retention, organic/inorganic ion transport, and heavy metal fixation [18]. Based on their sustainable microbial metabolism, PMFCs have potential soil remediation functions [19, 20] and have become a research hotspot in recent years. However, under the existing research conditions, PMFCs still have the problem of low working efficiency, and further research is needed for practical application [21].

The quality and cost of anode materials are two of the most important factors limiting the large-scale application of MFC. Therefore, improving the overall performance of MFC by anodic modification has become a common and most effective method [22-24]. Because of its low cost and good biocompatibility, carbon felt has become the most widely used anode material in MFC [25]. However, the density of the biofilm formed on the surface of carbon felt is low, resulting in a low electron uptake rate and extracellular electron transfer efficiency of the electrode [26, 27], which affects the electricity generation rate of MFC. Therefore, by modifying carbon felt electrodes to change their surface roughness, hydrophilicity, and biological affinity, the formation of surface biofilms can be accelerated, the density of the biofilm on the electrode surface can be increased, and thus the MFC performance can be improved. Nanocerium oxide (CeO_2) is a low-cost lanthanide-based nanorare earth oxide material. Due to its excellent redox ability, nano-CeO_2 can promote electron transfer on the electrode and exhibits excellent photochemical properties and stability, making it more suitable as an anode catalyst [28]. The use of nano-CeO_2 to modify carbon felt anodes in MFC can significantly increase the power generation of MFC [29]. Polyaniline (PANI) is a kind of conductive polymer with strong conductivity, good biocompatibility, and high stability. When it is used to modify the anode, a large number of microorganisms can be attached to the anode and rapidly propagated, thus improving the anode's performance [30, 31]. In view of these, PANI and CeO_2 are combined to modify the anode to further improve the soil remediation performance and power generation performance of PMFC.

Materials and Methods

Construction of an Experimental System

The experimental system was mainly composed of a single-chamber PMFC and an external load and data acquisition system, as shown in Fig. 1. Among them, the single-chamber PMFC was composed of a potted Scindapsus Aureus, a cathode, and an anode. The plant pot, with a diameter of 16 cm and a height of 10 cm, actually acted as the PMFC reactor. Bacteria-rich activated sludge mixed with cadmium contaminated soil was used as the planting soil. The bottom of the flowerpot was covered with 2~3 cm thick planting soil, and a square of carbon felt of 5 cm×5 cm was placed above it as the anode. Scindapsus Aureus was planted in the pot, and another part of the planting soil was used to fill the roots. The soil layer height was about 8 cm to ensure that the plant roots were located near the anode. The cathode is a 5 cm×5 cm square of carbon felt and was placed on the surface of the pot soil to make it fully in contact with oxygen in the air. The cathode and anode were connected through copper wires and externally connected with a 1000 Ω resistor. Organic matter in soil is degraded by electrogenic microorganisms to produce electrons. The output voltage generated by PMFC was collected in real-time through a data acquisition card (MPS-010602; Beijing Qichuang Mofei Electronic Technology Co., Ltd., China) and transmitted to a computer through a USB interface for storage, processing, and display.

Microbial Culture and Planting Soil Preparation

Electrogenic bacteria are indispensable for soil remediation and power generation processes in PMFC. Soil is rich in a variety of microbial species and is a common source of inoculants used in biological treatment systems, which can directly supply some of the microorganisms needed for PMFC. In order to further enhance the number and activity of exoelectrogens

Fig. 1. Experimental system of PMFC.

in the pot soil, some soil was acclimated to obtain microbial strains and added to the pot soil. The soil used for microbial cultivation was taken from the roots of a certain tree on campus. 0.01 g $Cd(NO_3)_2$, 100 g soil, 400 mL deionized water, and 3 mL trace element solution (SL-6, Shandong Tuopu Biol-engineering CO., LTD., China) were added to the culture bottle, which was kept in an anaerobic state. The culture bottle was then placed in a biochemical incubator and cultured at 30ºC for about three days. When there are obvious suspended flocculents and gas expansion in the mixed solution in the cultivation bottle, it can be considered that microbial cultivation has been successful.

Cadmium contaminated soil was obtained through manual addition. The soil collected from the campus was naturally air dried, then ground and passed through a stainless steel sieve with evenly distributed 2 mm holes to obtain impure and pollution-free loess. 0.1 g of $Cd(NO_3)_2$ was mixed with 1000 g and left at room temperature for one day, then used as pot soil.

Preparation of Electrodes

Firstly, the carbon felt of 5 cm×5 cm×0.8 cm was soaked in acetone for 2 h, then washed repeatedly with deionized water until neutral, and then dried at 60ºC in a blast drying oven for 3 h to obtain the unmodified conventional carbon felt (CF) electrode.

The CeO_2 solution was obtained by dissolving 0.5 g nanoscale cerium oxide powder in 200 mL deionized water and stirring for 2 h. Then, the carbon felt electrode prepared in the previous step was immersed in CeO_2 solution for 2 h, and then dried in an oven at 60ºC for 3 h to obtain the nanoscale CeO_2 modified carbon felt anode (CeO_2/CF).

Dissolve 2.28 g ammonium persulfate in 100 mL deionized water, 50 mL of which was polymerized with 4 mL aniline monomer to form polyaniline, then dissolved in 200 ml deionized water and stirred for 2 h to obtain a PANI solution. The prepared conventional carbon felt anode was soaked in PANI solution for 2 h, and then dried in a 60ºC oven for 3 h to obtain a polyaniline modified carbon felt anode (PANI/CF).

54 mL PANI solution and 0.5 g CeO_2 were dissolved in 200 mL deionized water and stirred for 2 h to obtain PANI-CeO_2 solution. The conventional CF electrode was soaked in PANI- CeO_2 solution for 2 h, and then dried in an oven at 60ºC for 3 h to obtain a polyaniline and cerium oxide modified carbon felt anode (PANI-CeO_2/CF).

Main Analysis Method

The voltage and current density are used to reflect the electrical performance of the PMFC. The output voltage of PMFC is the potential difference between the anode and cathode. In the experiments, the voltage values were collected in real time by a data acquisition card. The current density was calculated based on the measured voltage, effective anode surface area, and load resistance, namely:

$$I_A = \frac{U}{AR} \tag{1}$$

where I_A is the current density, U is the measured voltage, A is the surface area of the anode, and R is the load resistance. In this study, the surface area of the anode was 2.5×10^{-3} m^2, and the value of load resistance was set to 1000 Ω.

In order to better understand the surface structure changes of the modified carbon felt, the surface morphology of anode samples before and after modification was characterized by scanning electron microscopy (SEM, JSM-6360LV, Tokyo JEOL Co., LTD., Japan).

In order to further analyze the conductivity of the electrodes, the cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) tests were performed on a chemical workstation (CHI660E, Shanghai Chenhua Instrument Co., LTD., China), with an MFC anode as the working electrode, a platinum electrode as the auxiliary counter electrode, and a saturated calomel electrode (SCE) as the reference electrode. In the tests, the potential range is -0.6 V~0.6 V, and the scanning speed is 0.01 V/s.

In order to understand the polarization and power generation characteristics and analyze the effect of the anode modification method on the performance of PMFC, the polarization curve of PMFC was measured by the variable resistance method. Adjust the load resistance of the PMFC in the range of 9000~100 Ω (9000-7000-5000-3000-2000-1000-500-200-100 Ω) through the resistance box, record the steady-state output voltage under different load conditions, and calculate the corresponding current density, thus obtaining the polarization curve of the PMFC.

The Cd removal rate is an important indicator to characterize the effectiveness of soil remediation. The total concentration of Cd in the soil was determined by graphite furnace atomic absorption spectrophotometry (AA-6880) after digestion with hydrochloric acid-nitric acid-hydrofluoric acid-perchloric acid. By comparing the initial and final Cd concentrations in soil, the removal rate of Cd in soil by PMFC was calculated, and the remediation effect of different configurations of PMFC on Cd contaminated soil was analyzed. The formula for calculating the removal rate of Cd is as follows:

$$C_{\mathrm{cdrr}}(\%)=\frac{C_{\mathrm{cd0}}-C_{\mathrm{cdend}}}{C_{\mathrm{cd0}}}\times 100\% \tag{2}$$

where C_{cd0} and C_{cdend} represent the initial and final values of cadmium in the soil corresponding to the beginning and end of the experiment, respectively, and C_{cdrr} denotes the removal rate of cadmium by PMFC at the end of the operation.

Results and Discussion

Electricity Generation Performance

The output voltage curves of PMFCs with CF, CeO_2/CF, PANI/CF, and PANI-CeO_2/CF anodes are shown in Fig. 2. Due to the influence of microbial activity and other factors, the output voltage decreased significantly after running for 200 h, so the operating cycle was set to 200 h. It can be seen that the maximum output voltage values of PMFCs loaded with CF, CeO_2/CF, PANI/CF, and PANI-CeO_2/CF anodes were 2.46, 4.64, 6.42, and 10.01 mV, respectively. Compared with CF, CeO_2/CF and PANI/CF, the output voltage of PMFC using the PANI-CeO_2/CF anode increased by 307.2%, 115.8%, and 55.9%, respectively, indicating that the anodic carbon felt modification with PANI or CeO_2 has a certain effect on improving the generation capacity of PMFC. PANI and CeO_2 are effective electrode modification materials, especially when using a blend of PANI and CeO_2 for anode modification. The electricity generation capacity of PMFC was improved most significantly.

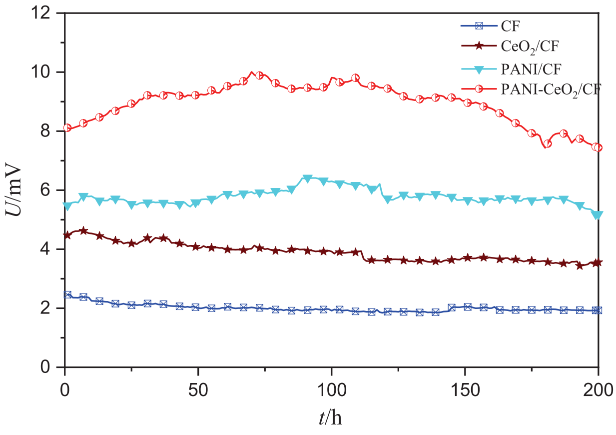

Fig. 2. the output voltage of PMFCs.

Further observation of Fig. 2 reveals voltage fluctuations within the operating cycle. This is mainly due to the complex biochemical reactions between microorganisms, soil, and plants. Random factors such as temperature, microbial activity, and light intensity can have impacts on the reaction process, leading to aperiodic fluctuations in the generating voltage.

Polarization Characteristics

Fig. 3 shows the polarization curves of four sets of PMFCs with different anodes. The polarization curve can directly reflect the total voltage loss of a fuel cell. The "higher" the polarization curve, the smaller the total voltage loss of the fuel cell. It can be seen from the polarization curves shown in Fig. 3 that, under the same current generation, the output voltage of PMFC using the PANI-CeO_2/CF anode was always higher than that of PMFCs using other three anodes, while the polarization curve of PMFC using the conventional unmodified anode was the lowest. These show that the voltage losses of PMFCs with three modified anodes were reduced to different degrees compared with PMFCs with an unmodified anode, and the voltage loss of PMFC with PANI-CeO_2/CF was the least, which presented a better effect on improving the power supply efficiency of PMFC.

CV Characteristics

The cyclic voltammetry (CV) curves of four different anodes are shown in Fig. 4. From the results in the figure, it can be seen that among the four types of anodes, the PANI-CeO_2/CF anode has the largest integrated area and the best symmetry in the CV closure curve. The integral area of the CV curve represents the discharge capacity of a fuel cell. The larger the area, the closer the mutual contact between the electrode and electrolyte, and the greater the charge storage capacity and charge transfer amount, resulting in a larger battery capacity [32, 33]. The better the symmetry of the CV curve, the better the reversibility of the electrochemical

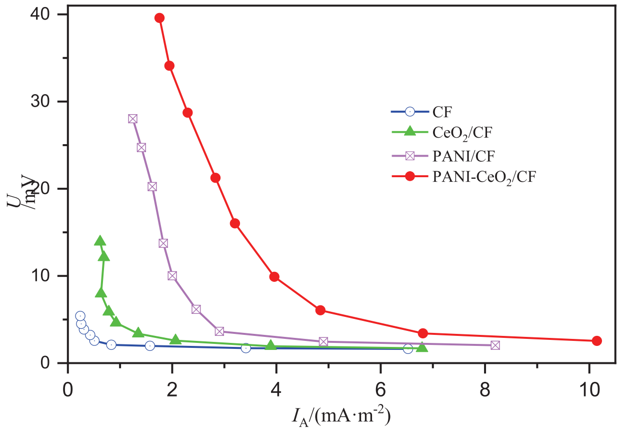

Fig. 3. Polarization curves.

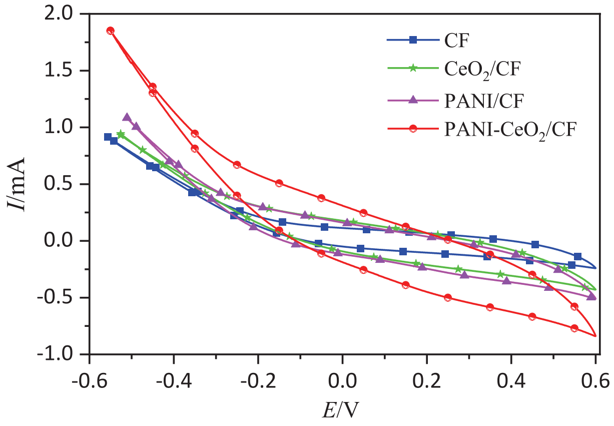

Fig. 4. CV curves.

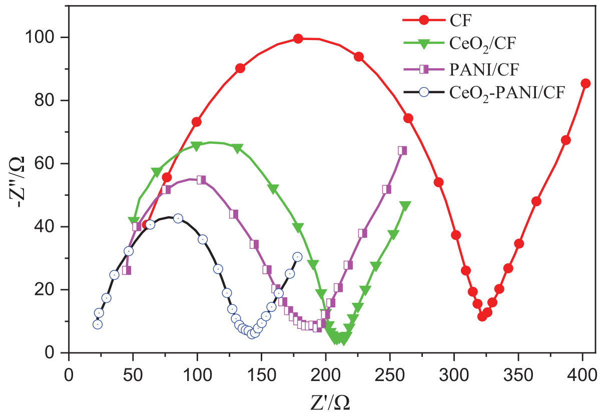

Fig. 6. EIS graph.

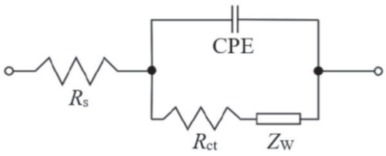

Fig. 7. Randles equivalent circuit.

reaction of the electrode. Therefore, the CV test results fully demonstrate that the synergistic effect of PANI and CeO_2 can improve the electrochemical activity of the anode surface, thereby promoting the rapid formation of biofilm on the anode surface, accelerating the electron transfer rate, and intensifying the reaction process of organic matter in soil being oxidized by microbial metabolism in the anode and oxides being reduced in the cathode, thus significantly improving the abilities of PMFC in power generation and soil purification.

SEM Images

The SEM images of the four different anodes measured before the experiments are shown in Fig. 5. It can be seen that the surface of a conventional CF anode without modification was smooth, so the specific surface area was small, and the number of microorganisms that attached to and grew on its surface was also small. The surface of the anode modified with nano-CeO_2 or PANI was loaded with a small amount of CeO_2 or PANI particles, but no obvious biofilm has been formed, which to some extent increased the specific surface area of the anode. Compared with the first three types of anodes, the carbon felt electrode co-modified by CeO_2 and PANI formed a more obvious biofilm on the surface, attached more conductive particles on the surface, further improved the surface roughness, and significantly increased the specific surface area and pore volume, which was more conducive to the adhesion and growth of microorganisms on the electrode surface and greatly improved the electron transfer rate, and thus, significantly improving the electricity generation performance and soil treatment ability of PMFC.

EIS Tests

Internal resistance is an important factor affecting the efficiency of the power supply. The internal resistance of PMFC can be measured by EIS characteristics. The EIS characteristics were tested using an electrochemical workstation at a polarization potential of -0.1 V with a scanning frequency range of 1~1.0 × 10^6 Hz. The Nyquist curves of the EIS of PMFCs with the four different anodes are shown in Fig. 6, and the Randles equivalent circuit is shown in Fig. 7, where R_s represents the solution internal resistance, R_{CT} represents the charge transfer resistance, CPE represents the constant phase angle element, and Z_W is the Warburg impedance.

In the EIS spectrum, the left intersection point between the semicircle and the horizontal axis of the EIS curve corresponds to the solution internal resistance R_s, and the diameter of the semicircle corresponds to the charge transfer impedance R_{CT} [34]. The impedance values of the four anodes obtained from the EIS test

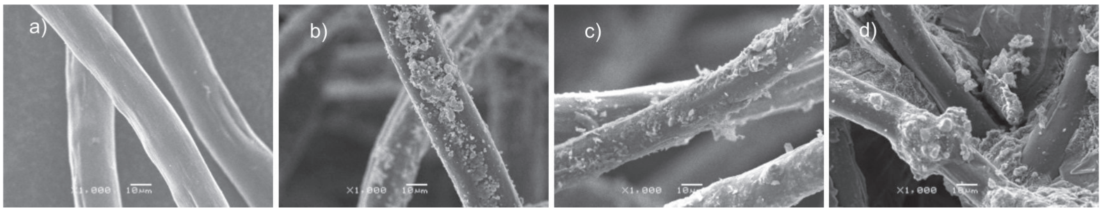

Fig. 5. SEM images of anodes.

Table 1. Equivalent resistance of four different anode PMFCs.

	CF	CeO_2/CF	PANI/CF	PANI-CeO_2/CF
R_s/Ω	39.05	20.77	21.79	17.97
R_{ct}/Ω	285.1	185.6	159.8	120.2
R_{eq}/Ω	224.15	206.37	181.59	138.17
Reduction of R_{eq}/%	0	7.9	18.9	38.4

Table 2. Cd removal rate of PMFC with four different anodes.

	CF	CeO_2/CF	PANI /CF	PANI- CeO_2/CF
C_{cd0}/(mg/kg)	100	100	100	100
C_{cdend}/(mg/kg)	38.93	8.53	7.25	6.09
C_{cdrr}/%	61.07%	91.48%	92.75%	93.91%
Increase of C_{cdrr}/%	0	49.8%	51.9	53.4

results are shown in Table 1, where the equivalent internal resistance of PMFC was approximately calculated by $R_{eq} \approx R_s + R_{CT}$.

According to the above EIS test results, it can be seen that the use of PANI-CeO_2 modified anodes effectively reduced the internal resistance of PMFCs, which indicates that this anode modification method is very effective for improving the electric generation performance and power supply efficiency of PMFCs.

Cd Removal Rate

The removal rate of heavy metals reflects the remediation ability of PMFC on contaminated soil. The test data for soil cadmium content and cadmium removal rate before and after the experiment are shown in Table 2.

From the experimental data in Table 2, it can be seen that the removal rates of Cd in soil by PMFCs using CF, CeO_2/CF, PANI/CF, and PANI-CeO_2/CF anodes were 61.07%, 91.48%, 92.75%, and 93.91%, respectively. PMFC using PANI CeO_2/CF modified carbon felt had the highest Cd removal rate, which was 53.77%, 2.66%, and 1.25% higher than that using CF, CeO_2/CF, and PANI/CF modified carbon felt anodes, respectively. This indicates that PMFC not only has the ability to use plants and soil to generate electricity, but also has a good role in solving the problem of heavy metal pollution in soil. The anodic modification of PMFC with CeO_2 and PANI significantly increased the removal rate of heavy metals in soil, which is very effective in improving the soil remediation ability of PMFC.

Conclusion

CeO_2, PANI, and PANI-CeO_2 modified carbon felt anodes were prepared using the solution immersion method and loaded onto PMFCs for the treatment of Cd contaminated soil. Nanoscale CeO_2 has a good catalytic effect, which is conducive to the transfer of electrons between electrodes. PANI has the advantages of strong conductivity, good biocompatibility, and high stability. During the composite process of the two, the nano CeO_2 particles are coated with polyaniline, forming a core-shell structure, further improving the conductivity and thermal stability of the electrode. The electricity generation and soil remediation capacity of PMFCs with PANI, CeO_2, and PANI-CeO_2 modified anodes have been improved compared to those using conventional CF anodes. Among them, the electricity generation capacity and heavy metal removal rate of PMFC using a PANI-CeO_2 composite modified anode have been significantly improved. Compared with PMFC with a conventional CF anode, PMFC using PANI-CeO_2/CF anode has a 307.2% increase in electricity generation voltage and a 53.8% increase in cadmium removal rate. The synergistic effect of PANI and CeO_2 improves the biocompatibility of the anode surface, promotes the formation of biofilm on the anode surface, accelerates the electron transfer rate, and thus improves the performance of PMFC.

Acknowledgments

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Conflict of Interest

The authors declare no conflict of interest.

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