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Coating-type three-dimensional acetate-driven microbial fuel cells

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This study uses sodium acetate as fuel to construct bioelectricity in coating-type three-dimensional microbial fuel cells anode. The coating-type three-dimensional anode was constructed using iron net as structural support, adhering a layer of carbon felt as primary coating and using carbon powder and 30% PTFE solution mixture as coating. The efficiency of electricity production and wastewater treatment were analyzed for the three-dimensional acetate-fed ($C_2H_3NaO_2$) microbial fuel cells with the various ratio of the coating mixture. The results showed that the efficiency of electricity production was significantly improved when using the homemade coating-type microbial fuel cells anode compared with the one without coating on the iron net, which the apparent internal resistance was decreased by 59.4% and the maximum power density was increased by 1.5 times. It was found the electricity production was greatly influenced by the ratio of the carbon powder and PTFE in the coating. The electricity production was the highest with apparent internal resistance of 190 Ω, and maximum power density of 5189.4 mW m^{-3} when 750 mg of carbon powder and 10 ml of PTFE (i.e., ratio 75:1) was used in the coating. With the efficiency of electricity production, wide distribution and low cost of the raw materials, the homemade acetate-fed microbial fuel cells provides a valuable reference to the development of the composition microbial fuel cell anode production.

[**Key words:** Microbial fuel cells; Three-dimensional anode; Coating anode; Electricity production; Wastewater; Carbon felt]

Since the beginning of the 21st Century, energy resources exploitations and environment pollution have become the two major factors that restrain the global economic development. Because of the ability of treating wastewater and providing bioelectricity, microbial fuel cell has been increasingly concerned and been vastly developed (1,2). Comparing to the traditional energy technologies, this type of clean energy developing technology is not only used to produce electricity, it simultaneously clean wastewater. Microbial fuel cells (MFCs) have the features of mild operation condition requirement, little waste generation and low operation cost (3,4).

In the MFCs structure, the anode is functioned as microbial settlement and electron delivery, which is a critical factor for electric generation. MFC anode research plays a significant important role. A new type of anode, which is three-dimensional MFC anode (filled MFC) is using either activated carbon particles or other porous particles to fill the whole anode chamber to maximize surface area of the anode for improving microbial settlement, and battery power (5,6). Composite anode is the combination of more than two types of electrode materials. It has been proved that it can decrease the internal resistance and increase MFCs power density. Composite anode MFC made from either polypyrrole or carbon black-coated carbon paper can improve MFCs performance (7). However, combination of both materials can improve its performance more effectively and the final power density had reached 452 mW m^{-2}. Comparing with the flat-plate MFC, the sintered-type composite anode combined the sintered carbon felt with carbon paper can increase the maximum current density of MFC from 3000 mA to 8000 mA, and increase the maximum power density from 1100 mW m^{-2}to 2426 mW m^{-2} (8). Although it has proved that both three-dimensional anode and composite anode can improve electric production effectively, both have some defects. For example, development of three-dimensional anode is stagnated at where only single type material is used for filling- three-dimensional composite anode still cannot be produced. The technology for composite anode production is too complicated.

In this research, the three-dimensional anode and composite anode were combined to improve the performance of microbial fuel cells with low cost and broad source anode materials.

MATERIALS AND METHODS

Coating-type three-dimensional composite anode The process to make the coating-type three-dimensional composite anode is shown in Fig. 1. The size of iron net is 40 × 40 × 20 mm, and the grid is 2 × 2 mm. Two layers of iron net are covered by a single layered carbon felt with the thickness of 1 mm. Iron net and carbon felt were separately soaked in 1 mol * L^{-1}HCl solution for 24 h to remove the impurity ion prior to assembly, rinsed with the distill water and dried. Certain amount of carbon powder and 30% PTFE solution were mixed and settled aside for later use for coating. After adhering the carbon felt onto iron net, the coating was brushed onto the carbon felt with the brush and dried at the room temperature. The coating mixture was stirred at the same time. When white color started to appear on the carbon felt, the second layer was added. This step was repeated until all the coating mixture was applied onto the carbon felt. The coated carbon felt was dried for 24 h in air- and water-free environment. The coating-type three-dimensional composite anode is a three-dimensional hollow structure. A stainless steel was used for ion collection and external electrode circuit connection. The surface of the anode is a hard gray-white solid with visible carbon felt fiber lines.

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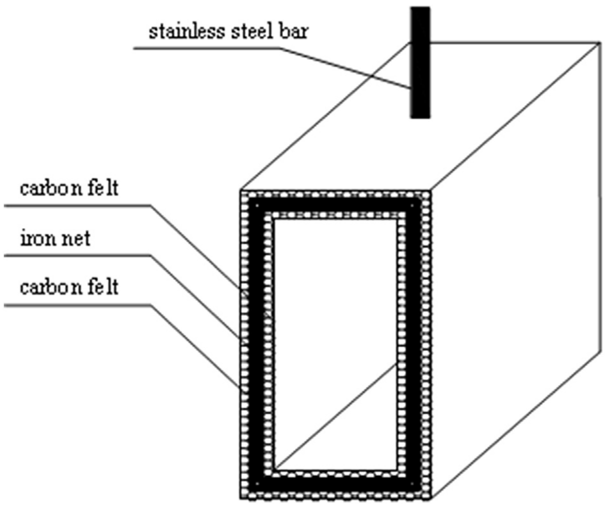

FIG. 1. Schematic diagram of the homemade three-dimensional composite anode.

Testing apparatus MFC apparatus was single-chamber air-cathode (9) microbial fuel cell (ACMFC) with the organic glass material. Its dimension was 5 cm × 3 cm × 5 cm (length × width × height). Total volume was 75 ml, and effective volume was 50 ml. The homemade ferrous carbon cloth as cathode was used. Iron catalyst capacity was 0.87 mg cm^{-2}. The distance between the cathode and the anode was 1.5 cm, which was connected by copper wire. Both sampling port and reference electrode reserve port were homemade on the glass container.

Microbial inoculation and operation The bacteria used in the experiment were obtained from anaerobic sludge in digesters of the sewage plant in Shenyang. The sludge was cultured anaerobically in acetic acid water for 30 days, and 25 ml of sludge culture stock was inoculated to the container. In the anode chamber, acetic acid water was added as fuel and other chemicals were added as the culture media: CH_3COONa 1640, NH_4Cl 500, KH_2PO_4 300. $MgCl_2 \cdot 6H_2O$ 100, $CaCl_2 \cdot 2H_2O$ 100 and KCl 100 (in unit of mg L^{-1}). The pH of the final mixture was adjusted to 7.5. The substrate was added into anode chamber using syringe when the open circuit voltage was decreased to 50 mV below. The operation temperature is kept at about 22°C and external connection capacity was 1000 Ω.

Analysis and calculation method The apparent internal resistance of MFC was determined by the steady-state discharge (10). The voltage was measured using UT71A intelligent digital multimeter. The range of load resistance is 90,000–10 Ω. The power density P (mW m^{-3}) was calculated by the equation of $P = U^2/RV$, where U is voltage (V), R is resistance (Ω), and V is effective volume (m^3). The chemical oxygen demand (CODcr) was measured using fast confined catalytic digestion-spectrophotometer (11).

RESULTS AND DISCUSSION

Electric production of three-dimension anode made with iron net The iron net is a widely available, and low cost conductive material. To compare electricity generation performance of three-dimensional composite anode, three-dimensional anode with iron net itself was used to analyze the MFCs polarization curve and power density–current curve (Fig. 2).

When using iron net alone as three-dimensional anode in the MFC, both the polarization curve and power density–current curve changes gently. The apparent internal resistance of MFC is 469 Ω identified from the fitting linear portion of the polarization curve, and the calculated maximum power density was 1888.1 mW m^{-3}. The maximum open circuit voltage reached 618 mV, which is significantly higher than using other materials as anode (carbon felt anode was 522 mV, stainless steel 591 mV and carbon rod was 307 mV). However, due to its higher apparent internal resistance, the battery with iron net has lower output power. Battery apparent internal resistance is composed of activation resistance, Ohmic resistance and the mass transfer resistance. The activation resistance is generated by the substrates decomposed by the microbial at the anode. The Ohmic resistance is generated by the battery structure itself. The mass transfer resistance is generated by

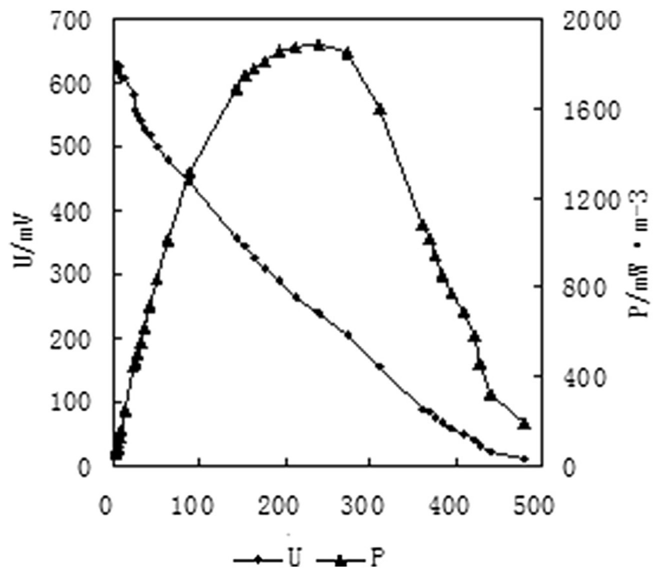

FIG. 2. Polarization curve and power density–current curve in the MFC.

traveling of proton produced by substrate, from the anode chamber to cathode. Because of the large hole in the iron net, and its smooth surface, few microbes can be adhere to the iron net. Thus, this assembled battery activation resistance should be comparably larger than others. Iron net is metal anode, which has a great electro-conductivity and low internal resistance. It results in lower Ohmic resistance compared with other batteries. The holes in the iron net are big with little effects on delivery of proton. It results in lower mass transfer resistance. The experiment found that small amount of rust appeared on the iron net after running the battery for a while. Therefore, to increase the surface roughness of the iron net and to apply with coating to prevent the iron from contacting with anode liquid are applicable methods to decrease the resistance of the iron net.

The battery made with iron net has high open circuit voltage. However, due to its high apparent internal resistance, it lowers electricity generation performance. Using carbon felt as anode material can give rough surface, large surface area, and low resistant, which has a great environment for microbial growth at the anode. Combination of iron net with carbon felt, using iron net as the structure or the three-dimensional anode can lower the Ohmic resistance and maintain the three-dimensional structure. Adhere carbon felt on the iron net can increase the amount of the microbes adhering to the abode with increased the porosity. Carbon felt was adhered with the iron net by mixing the carbon powder and 30% PTFE solution together. PTFE itself has no conductivity and it is only served as the function of adherence. Mixing with carbon felt can not only increase the anode surface roughness, but also increase the coating conductivity allowing free delivery of electron on the electrode. The best ratio content of the coating materials was investigated in this study in order to increase the MFCs electricity generation performance.

Effect of the coating component ratio on performance of the three-dimensional anode MFC: electricity generation performance and efficiency of the wastewater treatment To study the effect of the coating component ratio on performance of the three-dimensional anode MFC, the ratio of the carbon powder versus 30% PTFE solution was set to 50:1. Four groups of coating materials with different content were chosen: group A (carbon powder: 100 mg and 30% PTFE solution: 2 ml), group B (carbon powder: 500 mg and 30% PTFE solution: 10 ml), group C (carbon