Preparation of PANI/PPy/MnO_2/CC Hydrogel Electrode and its Performance Study in Microbial Fuel Cell

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Abstract

In order to further improve the power density and output voltage of the battery, this paper introduces capacitive anode material to improve the output power of MFC. The most common pseudocapacitive materials mainly include metal oxides and conductive polymers. A rapid and reversible electrode reaction occurs in metal oxides, which usually occurs inside the electrode. Therefore, the theoretical capacitance of metal oxides is relatively large, which is several times that of carbon materials. Manganese dioxide (MnO_2) has been widely used as electrode materials for different batteries due to its high capacitance, good reversibility and low cost. Among conductive polymers, polyaniline (PANI) and polypyrrole (PPy) also have good conductivity, low price and high chemical stability. Therefore, based on the characteristics of polyaniline, polypyrrole and manganese dioxide, PANI/MnO_2/CC, PPy/MnO_2/CC and PANI/ PPy/ MnO_2/ CC hydrogel electrodes were prepared on CC substrates by in-situ polymerization method, and applied as anodes to microbial fuel cell devices to study the power generation performance of the cells.

Keywords

Microbial Fuel Cell; MnO_2; Polyaniline; Polypyrrole; Electricity Production Performance.

1. Introduction

The rapid development of economy has led to the increasingly prominent contradiction between energy supply and demand. The development of sustainable and pollution-free energy has become the only way for human beings to seek long-term development [1]. The total amount of sewage discharge in China is huge every year. Sewage contains abundant organic matter and is a considerable potential energy source. In order to solve the two serious problems of energy shortage and water pollution, wastewater resource utilization has become a sustainable development direction of wastewater treatment in the future [2].

Microbial fuel cell (MFC) converts chemical energy into electrical energy by oxidizing and decomposing organic matter in ship domestic sewage by electrogenic bacteria [3]. As a new type of energy conversion device with both sewage purification and energy recovery capabilities, it has great application prospects in the fields of environment, new energy, medicine, ocean and even aerospace. MFC anode is the carrier for the growth and reproduction of electrogenic bacteria. The properties of anode materials not only determine the number of attached electrogenic bacteria, but also affect the electron transfer efficiency between cells and anode surface. Therefore, the anode has become the key to improve the power generation performance of MFC [4]. From the perspective of device power generation, anode materials with application value are selected for research, and the anode surface is

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modified by surface modification technology to improve the electron transfer rate of the electrode, which is conducive to breaking through the bottleneck of low output power of MFC [5].

2. Preparation of Different Hydrogel Electrodes

2.1 Preparation of PANI/MnO_2/CC Hydrogel Electrode

(1) Configuration solution A: 0.921 mL phytic acid was added to the reagent bottle as a crosslinking agent, and then 0.458 mL aniline monomer was poured into the above reagent bottle. After stirring, 1.904 g MnO_2 was added, and magnetic stirring was performed again for 15 min.

(2) Configuration solution B: 1 mL of deionized water was added to the reagent bottle, 0.286 g of ammonium persulfate was added, and the magnetic stirring was stirred for 5 min to make it mixed evenly;

(3) Put the treated carbon cloth in the stirred solution A, and mix A and B in the new reagent bottle;

(4) PANI/MnO_2/CC hydrogel electrode was obtained by keeping the mixed solution below 2°C for one week.

2.2 Preparation of PPy/MnO_2/CC Hydrogel Electrode

(1) Configuration solution A: 0.672 mL pyrrole monomer was added to the reagent bottle, and 1.231 mL phytic acid was added as a cross-linking agent. The reagent bottle was placed in an ice water bath and stirred magnetically for 15 min to mix the solution evenly. 4 mL isopropanol and 1.904 g MnO_2 were slowly added into the mixed solution, and the magnetic stirring was performed for 15 min；

(2) Configuration solution B: 4 mL deionized water was added to the reagent bottle, 1.092 g ammonium persulfate was added to it, and magnetic stirring was performed for 15 min to make it dissolve evenly；

(3) The treated carbon cloth was placed in solution A, mixed with A and B two groups of solutions, placed in an ice water bath for 15 min, and maintained at 2°C for one week ;

(4) After the electrode reacted into gel, it was taken out and immersed in ultrapure water for one day to remove impurities. After the removal of impurities, the PPy/MnO_2/CC hydrogel electrode was obtained.

2.3 Preparation of PANI/PPy/MnO_2/CC Hydrogel Electrode

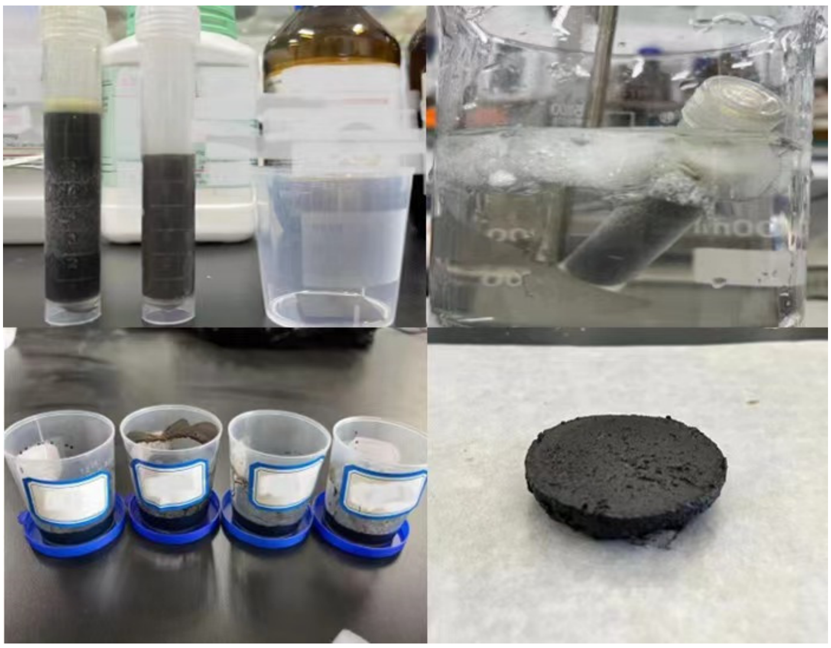

Figure 1. Preparation of different hydrogel electrodes

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(1) Configuration solution A: 0.137 mL aniline monomer was first added to the reagent bottle, and then 0.672 mL pyrrole monomer was added to it for mixing. The mixed solution was stirred in an ice water bath for 5 min. In addition, 1.472 mL of phytic acid was used as crosslinking agent and 1.904 g MnO_2 was added to the mixed solution, and magnetic stirring was performed for 15 min;

(2) Configuration solution B: 4 mL deionized water was added to the reagent bottle, 1.852 g ammonium persulfate was dissolved in it, and stirred in an ice water bath for 5 min;

(3) Put the treated carbon cloth in solution A, and quickly add solution B to solution A, so that solution A and solution B can fully react;

(4) The PANI/PPy/MnO_2/CC hydrogel electrode was taken out and immersed in deionized water to remove impurities until colorless.

The preparation of different electrodes is shown in Fig 1.

3. Characterization of Hydrogel Electrode

3.1 Scanning Electron Microscope Analysis

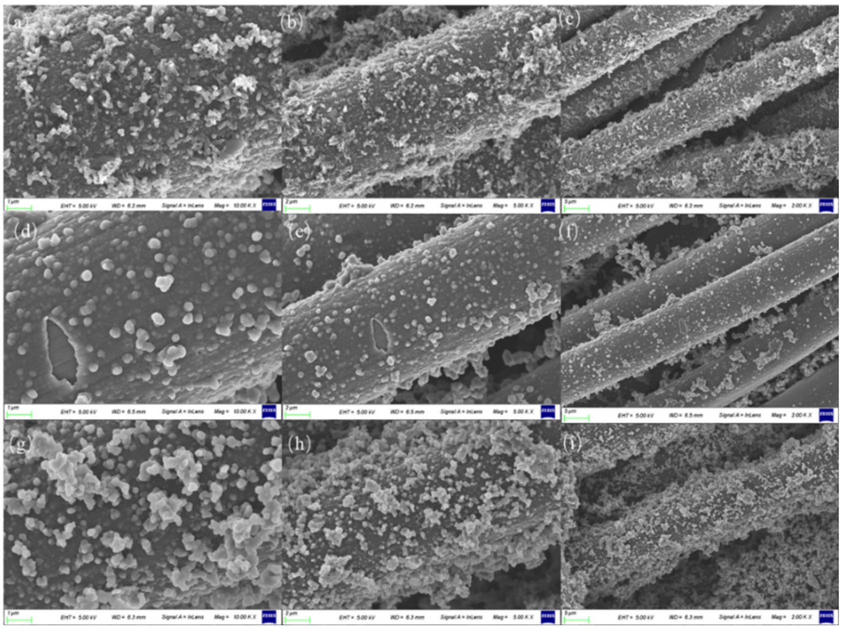

Figure 2. SEM images of different hydrogel electrodes

Fig 2 is the SEM images of PANI/MnO_2/CC, PPy/MnO_2/CC and PANI/PPy/MnO_2/CC hydrogel electrodes at 2000 times, 5000 times and 10000 times. Among them, figure (a) (b) (c) is the SEM diagram of PANI/MnO_2/CC hydrogel electrode. Figure (d) (e) (f) is the SEM image of PPy/MnO_2/CC hydrogel electrode. Fig (g) (h) (i) shows the SEM images of PANI/PPy/MnO_2/CC hydrogel electrode. It can be seen from the SEM image of PANI/MnO_2/CC hydrogel electrode that PANI/MnO_2 presents a non-uniform film-like structure on the surface of carbon cloth fiber and is accompanied by different degrees of protrusions. This is because aniline is oxidized. In the early stage, a thin film-like structure appears on the electrode [6]. As the reaction continues, the contact surface between polyaniline and the electrode is limited, which eventually causes irregular protrusions. It can be seen from the PPy/MnO_2/CC diagram that pyrrole polymerizes under the action of oxidant to form many polypyrroles similar to the shape of mushroom cloud, and the connection between polypyrrole particles is very close. Under the oxidation of ammonium persulfate, pyrrole monomer can be uniformly wrapped on the surface of manganese dioxide by hydrogen bond or π-π bond, and finally form nano-scale polypyrrole bulge on CC [7]. The needle-like structure of manganese dioxide cannot be clearly seen on the figure, because the length of manganese dioxide is about 0.2 μm, and the polypyrrole nanospheres are stacked to form an interconnected structure with a diameter of about 2

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μm ~ 5 μm. From the SEM image of PANI/PPy/MnO_2/CC hydrogel electrode, it can be seen that PPy is transformed from granular structure to coral-like structure and compounded with the lamellar structure of PANI on the surface of carbon cloth fiber, and the two polymer materials have a certain agglomeration phenomenon.The needle-like structure of the manganese dioxide material appears inside PANI.Three different materials work together to construct a network support structure on the electrode surface. This network support structure can provide more contact points for the electron transfer of microorganisms [8].

3.2 Infrared Absorption Spectroscopy Analysis

Fig 3 (a), (b) and (c) are the infrared spectra of PANI/MnO_2, PPy/MnO_2 and PANI/PPy/MnO_2 electrodes, respectively. The characteristic peaks of polyaniline and polypyrrole can be clearly seen in the figure. 1590 cm^{-1} corresponds to the C=C stretching vibration in the quinone structure. 1492 cm^{-1} corresponding to the C=C stretching vibration on the benzene ring. The absorption peak corresponding to 1302 cm^{-1} is C-N stretching vibration. These are characteristic peaks of polyaniline. 1164 cm^{-1} corresponding to the stretching vibration peak of N-H on the pyrrole ring. 1046 cm^{-1} corresponding to the stretching vibration peak of =C-H on the pyrrole ring. The characteristic peaks of the functional groups contained in polyaniline and polypyrrole are located at the corresponding positions, while the addition of MnO_2 increases the steric hindrance, broadens the characteristic peaks, and produces a shift [9].

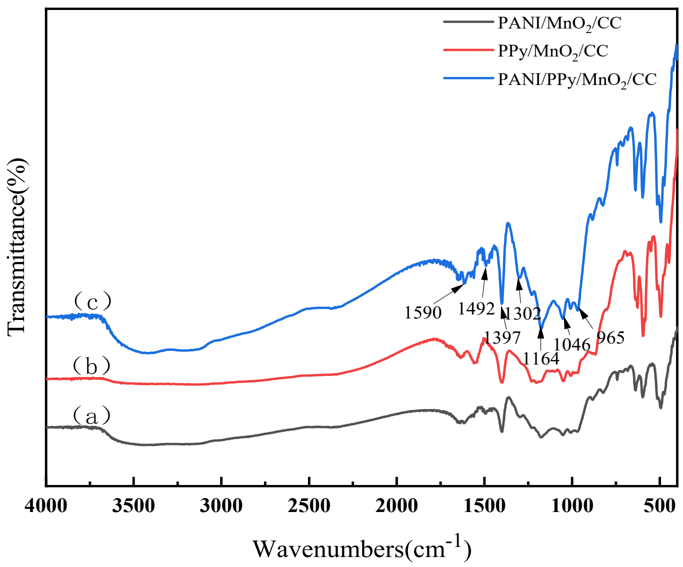

Figure 3. FTIR spectra of different electrode materials

3.3 Performance Testing of Hydrogel Electrodes

3.3.1 Contact Angle Test

The electrochemical reaction rate of the electrode is closely related to its hydrophilicity. The hydrophilicity of the electrode can be judged by the contact angle experiment [10]. The contact angle of electrode materials has a great relationship with their own structure, physical and chemical properties [11]. Three groups of hydrogel electrodes were tested by a contact angle tester, and the results are shown in Fig 4. The contact angles of PANI/MnO_2/CC, PPy/MnO_2/CC and PANI/ PPy/ MnO_2/ CC are 18.887 °, 20.222 ° and 14.482 °, respectively. The test results show that the surface contact angle of PANI/PPy/MnO_2/CC hydrogel electrode decreases obviously, and the hydrophilicity of the hydrogel electrode is the best. The surface of the electrode is easier to adhere to the electrogenic microorganisms, and it can also reduce the electron transfer resistance. According to the literature, the improvement of the hydrophilicity of the electrode makes the microbial attachment on the

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electrode surface increase significantly, which will also be beneficial to improve the power generation performance of MFC [12].

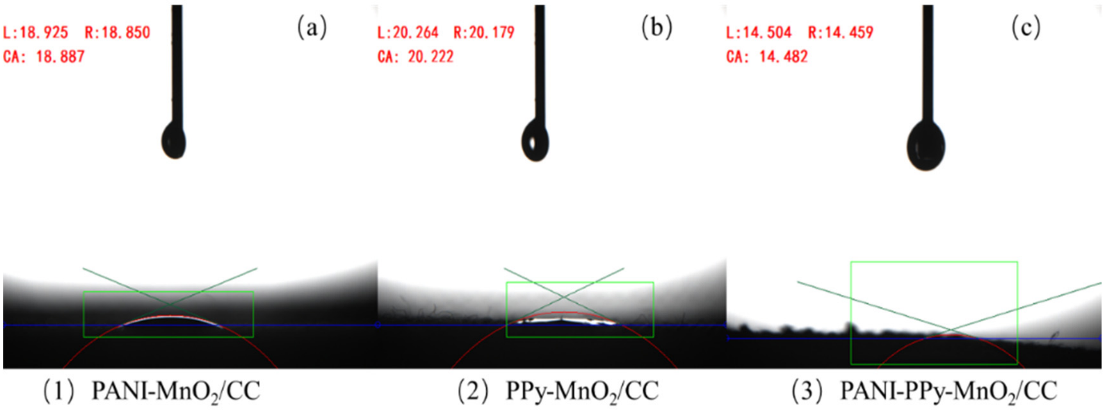

Figure 4. Contact angle of different anode surfaces

3.3.2 Swelling Performance Test

Fig 5 is the swelling performance curve of PANI/MnO_2/CC, PPy/MnO_2/CC, PANI/PPy/MnO_2 /CC hydrogel electrodes. The water absorption rate of three different hydrogels increased with the extension of soaking time. When the soaking time was about 1000 min, all hydrogel electrodes reached swelling equilibrium. It can be seen from the figure that the swelling performance of PANI/PPy/MnO_2/CC hydrogel is the best, and the swelling rate reaches 9.2 g/g. The maximum swelling ratio of PANI/MnO_2/CC hydrogel electrode was 8.6 g/g. The maximum swelling ratio of PPy/MnO_2/CC hydrogel electrode was 8.9 g/g. It is worth noting that the swelling rate of PANI/PPy/MnO_2/CC hydrogel is almost the highest when the immersion time reaches 600 min. Compared with the other two groups of electrodes, the immersion time at the maximum swelling rate is the shortest. This is because the PANI/PPy/MnO_2/CC hydrogel forms a semi-interpenetrating structure through the composite modification of PANI/PPy, and the porosity of this semi-interpenetrating structure hydrogel increases, which increases the water absorption of the hydrogel [13].

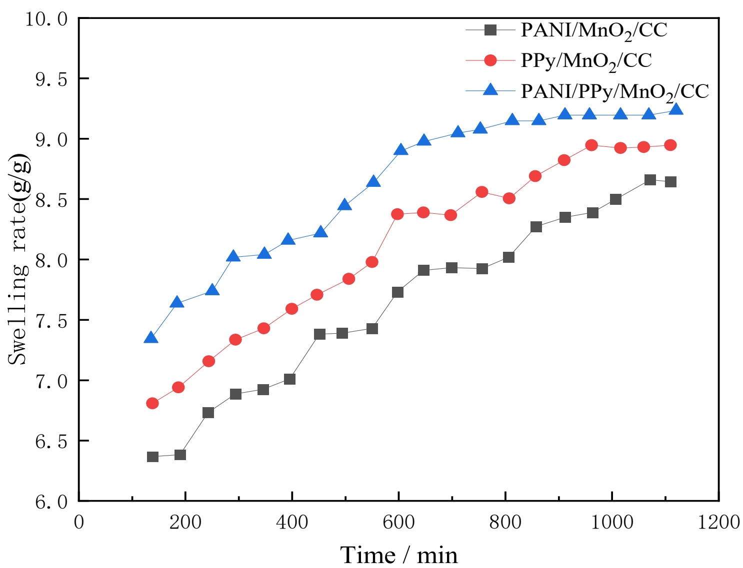

Figure 5. Water absorption multiplicity of three groups of hydrogels at different soaking times

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3.4 Performance Test of Different Electrodes in MFC

3.4.1 Cyclic Voltammetry Test

Fig 6 is the cyclic voltammetry test diagram of different hydrogel electrodes, and the test scan rate is 10 mV/s. From the test diagram, it can be seen that the area enclosed by the cyclic voltammetry curve of PANI/MnO_2/CC hydrogel electrode and PPy/MnO_2/CC hydrogel electrode is smaller. The area surrounded by PANI/PPy/MnO_2/CC hydrogel electrode is larger. According to the area of the CV curve formed by different hydrogel electrodes, the capacitance performance of PANI/ PPy/ MnO_2/ CC hydrogel electrode is significantly higher than that of PANI/MnO_2/CC and PPy/MnO_2/CC hydrogel electrodes. This is because the modified PANI/PPy/MnO_2/CC hydrogel electrode has good bioelectrocatalytic activity, which is suitable for the growth of microorganisms and accelerates the transfer rate of electrons produced by different strains to the electrode surface [14].

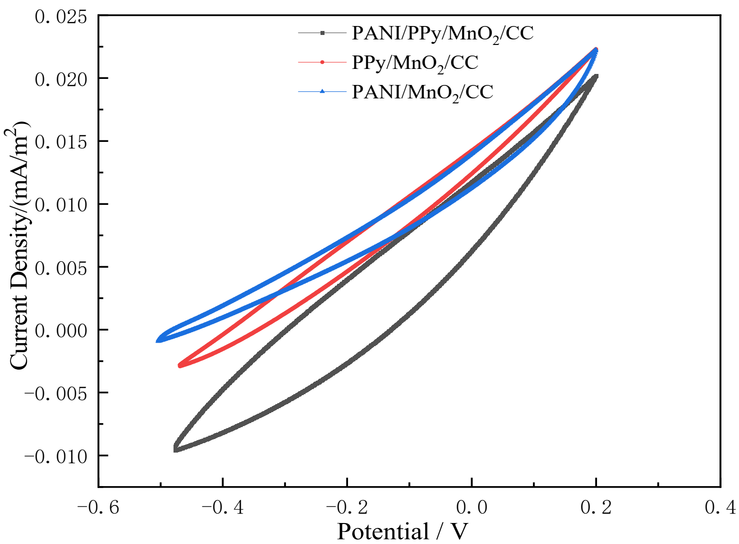

Figure 6. Cyclic voltammetric curves of different hydrogel electrodes

3.4.2 AC Impedance Test

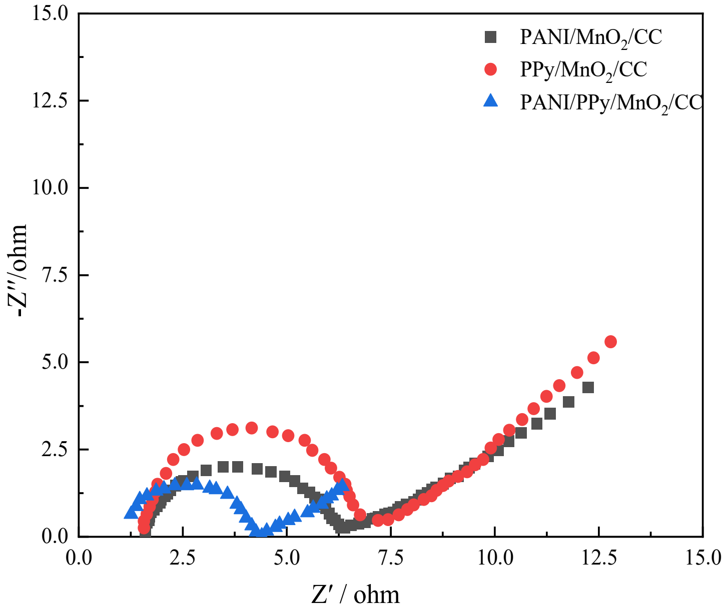

Figure 7. EIS diagram for different electrodes

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Fig 7 is the AC impedance spectra of PANI/MnO_2/CC, PPy/MnO_2/CC and PANI/PPy/MnO_2/CC hydrogel electrodes. The solution impedance R_1 and electron transfer impedance Rct of different electrodes can be obtained by curve fitting method, as shown in Table 1. The electron transfer impedance Rct of the three groups of hydrogel electrodes is in the order of PPy/MnO_2/CC > PANI/MnO_2/CC > PANI/PPy/MnO_2/CC. This is because the conductive polymer PANI and PPy composite modified electrode has higher conductivity. In addition, MnO_2 can also endow hydrogels with numerous pore structures. At this time, the microorganism hardly contacts with CC directly, but chooses to conduct electrons with the hydrogel on the outer surface, which indirectly promotes the transmission of electrons and reduces the electron transfer impedance of the hydrogel electrode.

Table 1. Fitting data for different electrode impedances

Electrode	PANI/MnO_2/CC	PPy/MnO_2/CC	PANI/PPy/MnO_2/CC
R_1 (Ω)	1.586	1.392	1.035
Rct (Ω)	4.724	5.508	3.174

3.4.3 Polarization Curve Test

Fig 8 is the polarization curves of PANI/MnO_2/CC, PPy/MnO_2/CC and PANI/PPy/MnO_2/CC hydrogel electrodes. It can be seen from the figure that as the current density increases, the battery voltage decreases to varying degrees. The polarization degree of different hydrogel electrodes is also different. The slope of PANI/PPy/MnO_2/CC hydrogel electrode is the smallest by the tilt degree of polarization curve. Compared with the other two groups of electrodes, the polarization degree is the smallest and the anti-polarization performance is the strongest. It can be seen from the figure that the voltage value of PANI/MnO_2/CC hydrogel electrode began to drop sharply from 760 mV at the initial current density. This shows that there is polarization at this time. The polarization curve of PANI/PPy/MnO_2/CC hydrogel electrode has been in a steady decline state, mainly due to the specific surface area of the hydrogel electrode modified by the two conductive polymers has been improved to a certain extent. The addition of manganese dioxide improves the electron transport ability of PANI/PPy/MnO_2/CC hydrogel electrode. The electrons produced by the electricity-producing microorganisms are more easily transmitted to the external circuit after being transmitted to the electrode, which reduces the resistance of the extracellular electron transfer of the electricity-producing microorganisms, reduces the internal resistance and polarization overpotential of the MFC, thus reducing the polarization degree of the battery.

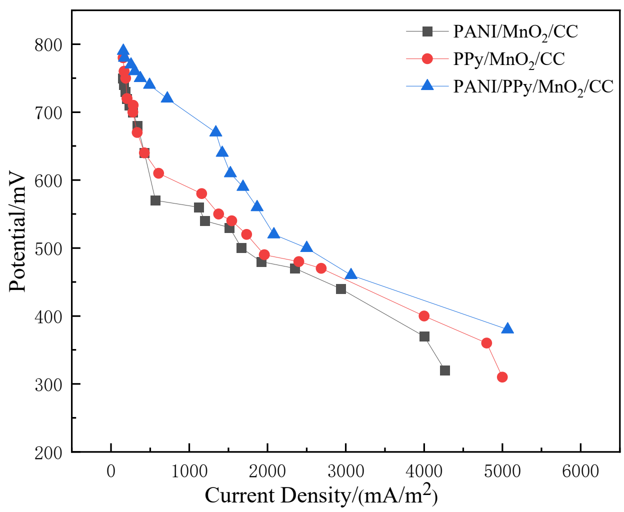

Figure 8. Polarization curves of MFC at different electrodes

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3.4.4 MFC Cycle Voltage Test

Fig 9 is the MFC output voltage curve of different electrodes with time. It can be seen from the figure that the MFC output voltage of PANI/PPy/MnO_2/CC anode is relatively high, which can reach about 671 mV. The output voltages of PANI/MnO_2/CC and PPy/MnO_2/CC are 652 mV and 645 mV, respectively. This shows that the charge transfer rate of the hydrogel electrode modified by polyaniline and polypyrrole is increased, and the output voltage of MFC is increased. At the same time, needle-like manganese dioxide can provide more surface active sites, which is conducive to the adhesion of electrogenic microorganisms and the improvement of MFC output voltage [15]. Compared with the MFC built by the other two groups of electrodes, the MFC output voltage where the PANI/PPy/MnO_2/CC hydrogel electrode is located has the fastest rate of increase over time and reaches the peak first. This may be due to polyaniline and polypyrrole modified anode to improve the electron transfer rate, voltage peak time shortened. The single cycle of MFC with PANI/PPy/MnO_2/CC anode reached about 250 h, which was slightly longer than that of the other two groups. The results showed that when PANI/PPy/MnO_2/CC hydrogel electrode was used as MFC anode, the output capacity of external load was improved.

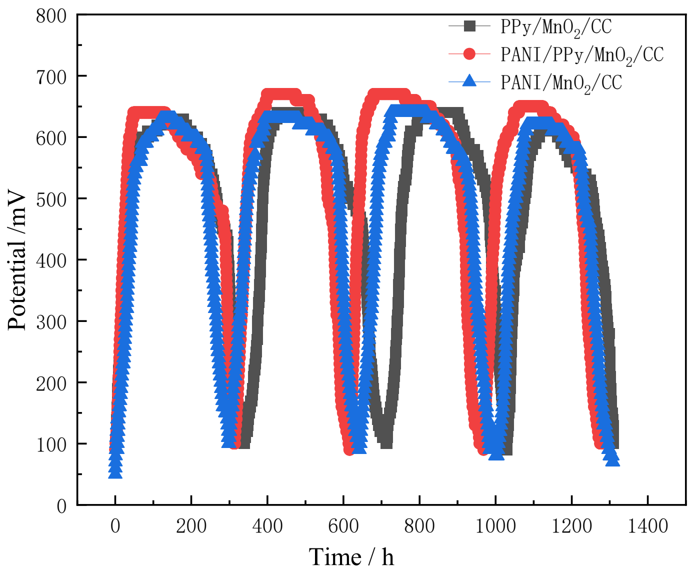

Figure 9. Output voltage curve of MFC with different electrodes

3.4.5 MFC Power Density Curve Test

Fig 10 is the power density curve of PANI/MnO_2/CC hydrogel electrode, PPy/MnO_2/CC hydrogel electrode and PANI/PPy/MnO_2/CC hydrogel electrode in MFC. The maximum power density of PANI/MnO_2/CC hydrogel electrode is 1600.28 mW/m^2. The maximum power density of PPy/ MnO_2/CC hydrogel electrode is 2106.64 mW/m^2. The maximum power density of PANI/ PPy/ MnO_2/ CC hydrogel electrode was 2406.22 mW/m^2. Compared with PANI/MnO_2/CC hydrogel electrode increased by 50.36%, compared with PPy/MnO_2/CC hydrogel electrode increased by 14.22%. It can be seen that the use of manganese dioxide to modify the surface of the hydrogel electrode can significantly reduce the internal resistance of MFC, thereby reducing the loss of MFC in the working process. In addition, compared with the single conductive polymer modification, there is a strong synergistic effect between the two different conductive polymers and manganese dioxide, thereby improving the power density of the electrode [16].

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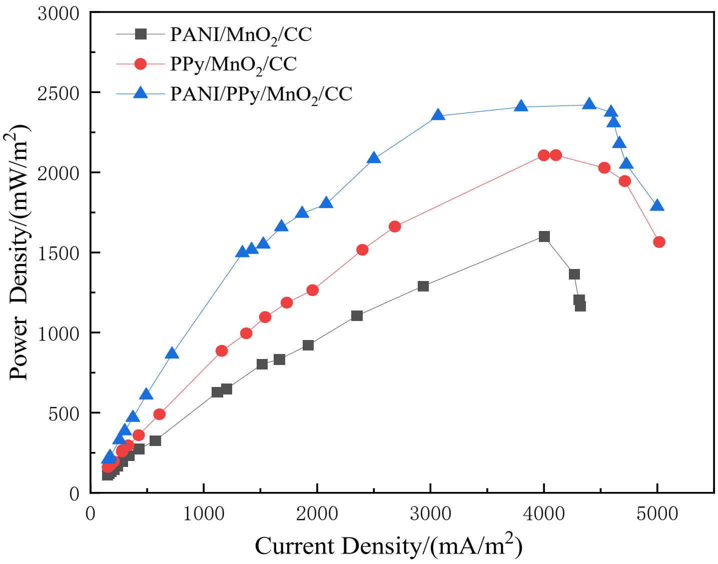

Figure 10. Power density curves of MFC with different electrodes

3.4.6 The Effect of External Resistance on the Electrical Performance of MFC Production

Fig 11 shows the output voltage of MFC in one cycle under different external resistances. The prepared PANI/PPy/MnO_2/CC hydrogel electrode was used as MFC anode and the external resistance was set to 500 Ω. At this time, the output voltage of the battery can reach 472.64 mV. In order to explore the change of output voltage with external resistance, the external resistance is further increased to 1000 Ω. At this time, the maximum output voltage of the battery rises to 670.58 mV, indicating that increasing the external resistance is beneficial to improve the output voltage of the battery. When the external resistance is reduced to 200 Ω, the output voltage of the battery is also reduced to 375.22 mV. The results show that the output voltage decreases with the decrease of the external resistance. Under three different external resistance conditions, the output voltage of the battery can remain stable for a certain period of time, indicating that the electrode has good stability and can provide lasting power [17].

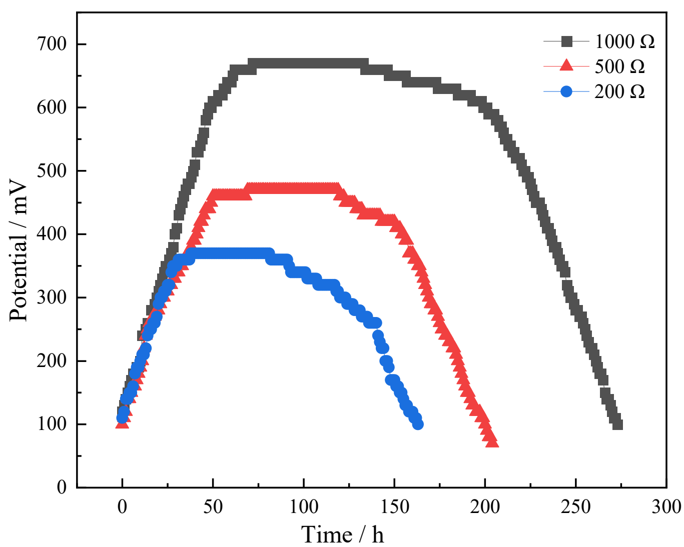

Figure 11. MFC output voltage curve under different external resistance

4. Conclusion

(1) In order to improve the electricity generation performance of MFC, three groups of hydrogel electrodes, PANI/MnO_2/CC, PPy/MnO_2/CC and PANI/PPy/MnO_2/CC, were prepared by introducing MnO_2. The results showed that the surface contact angles of the three groups of hydrogel electrodes were 18.887 °, 20.222 ° and 14.482 °, respectively. The electron transfer impedance is 4.724 Ω, 5.508 Ω and 3.174 Ω, respectively. The hydrophilicity of PANI/PPy/MnO_2/CC composite hydrogel electrode was improved, and the electron transfer impedance was reduced. The surface of the hydrogel electrode had a porous structure, which facilitated the attachment and shuttle of electrogenic bacteria, reduced the internal resistance of MFC, and made the electrode have better electron transfer ability.

(2) The results showed that the maximum output power density of PANI/MnO_2/CC hydrogel electrode was 1600.28 mW/m^2 and the output voltage was 652 mV. The maximum output power density of the PPy/MnO_2/CC hydrogel electrode is 2106.64 mW/m^2, and the output voltage is 645 mV. The maximum output power density of PANI / PPy / MnO_2 / CC hydrogel electrode is 2406.22 mW/m^2, and the output voltage is 671 mV. The maximum MFC output power of PANI/PPy/MnO_2/CC hydrogel electrode was 1.15 times that of PPy/MnO_2/CC electrode and 1.5 times that of PANI/MnO_2/CC electrode. These results indicate that the PANI/PPy/MnO_2/CC hydrogel electrode has high power generation performance and good biocompatibility.

Acknowledgments

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