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DIRECT METHANOL FUEL CELL MEMBRANE ELECTRODE FOR IMPROVING CATALYST UTILIZATION AND PREPARATION METHOD THEREOF

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ABSTRACT

The present invention discloses a direct methanol fuel cell (DMFC) membrane electrode capable of improving catalyst utilization and a preparation method thereof, and relates to the technical field of fuel cells. In the fuel cell membrane electrode prepared by the method of the present invention, the microporous layer has a bilayer structure, including an outer microporous layer with hydrophobic polymers and an inner microporous layer with proton-conductive polymers. The microporous bilayer structure, with extremely-large three-phase reaction interface and electrochemical reaction area, can effectively reduce the material transport resistance inside the electrode and remarkably improves the catalyst utilization, thereby enhancing the electrode discharge performance and extending the battery life. As determined by a single-cell performance test, the membrane electrode prepared by the preparation method of the present invention has significantly-improved monomer performance compared to a membrane electrode prepared by a conventional method.

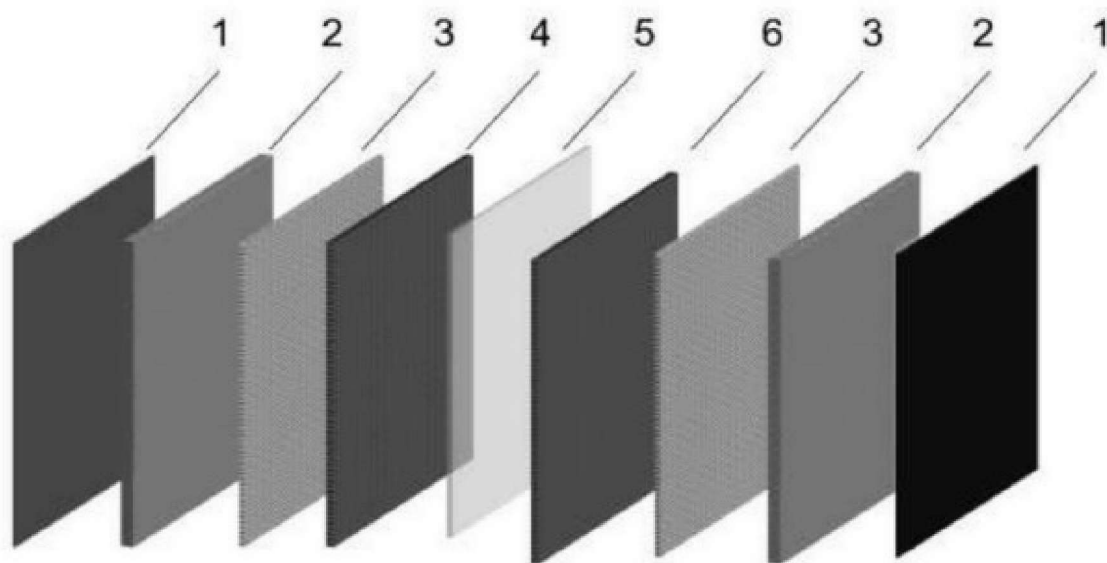


FIG. 1

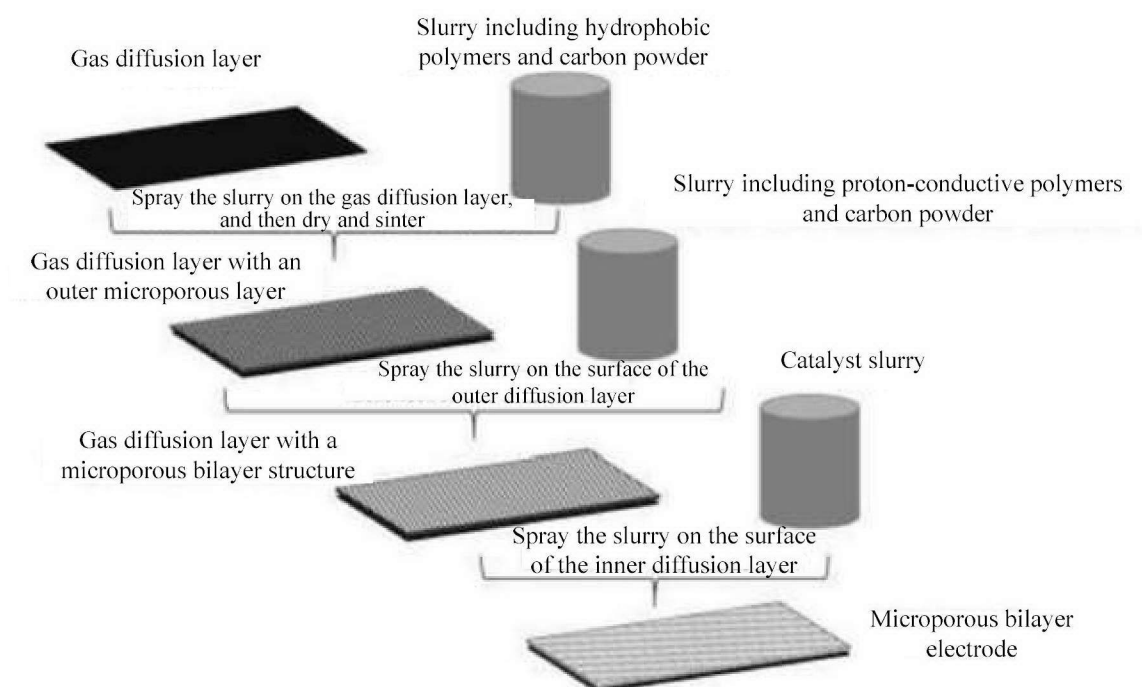


FIG. 2

DIRECT METHANOL FUEL CELL MEMBRANE ELECTRODE FOR IMPROVING CATALYST UTILIZATION AND PREPARATION METHOD THEREOF

TECHNICAL FIELD

The present invention relates to the technical field of fuel cells, and in particular, to a direct methanol fuel cell (DMFC) membrane electrode capable of improving catalyst utilization and a preparation method thereof.

BACKGROUND

Direct methanol fuel cell (DMFC) is a power generation device that can continuously convert the chemical energy in fuel and oxidant into electrical energy, and has attracted extensive attention worldwide due to its environment protection and high efficiency properties. Methanol fuel cells directly use methanol or a methanol aqueous solution as anode fuel, and oxygen or air as oxidant. Owing to advantages such as wide sources of methanol, portability, convenient storage and replenishment, high volumetric and gravimetric energy density, simple structure and no need for external reforming devices, DMFC has promising application prospects in portable power supply, small commercial power supply, vehicle power supply and the like.

Membrane electrode, as a core component of DMFC, directly determines the performance of the cell. However, at present, DMFC has a problem that the methanol anode has a low electrocatalytic activity. As methanol oxidation mechanism is very complicated, some unstable and insoluble intermediate products will be produced during the oxidation process. Certain intermediate products will be adsorbed on the surface of the catalyst, which inhibits the activity of the catalyst, thereby causing catalyst poisoning and low catalyst utilization. Therefore, optimizing the membrane electrode structure, improving catalyst utilization and increasing electrochemical reaction area are hot topics for DMFC research.

At present, most of the effort is focusing on the modification of the catalyst carrier or catalyst layer structure, but it is still necessary to solve the problem of battery performance degradation caused by the catalyst sinking due to assembly pressure, methanol flow and gas flow. After the battery has been running for a period of time, part of the catalyst cannot be in contact with ionic polymers, so that protons cannot be transferred and the catalyst cannot work. Therefore, the electrode has a large resistance to the electrochemical reaction, resulting in degradation in battery performance. In view of this, a novel microporous bilayer membrane electrode is constructed in the present invention. The microporous bilayer

structure is composed of an inner microporous layer (added with Nafion polymers) and an outer microporous layer (added with polytetrafluoroethylene (PTFE) polymers). The presence of the inner microporous layer can expand the three-phase interface area for reaction, and thus improve the catalyst utilization.

SUMMARY

The present invention is intended to provide a DMFC membrane electrode for improving catalyst utilization, thereby achieving the purpose to increase the three-phase reaction interface and electrochemically active area, improve the catalyst utilization, reduce the electrochemical reaction resistance and mass transfer resistance for an electrode, and enhance the performance of a single cell.

The present invention has the following technical solutions.

A DMFC membrane electrode capable of improving catalyst utilization is provided, including a DMFC membrane electrode having a microporous layer prepared thereon, and the microporous layer has a bilayer structure, including an outer microporous layer with hydrophobic polymers and an inner microporous layer with proton-conductive polymers.

A method for preparing a microporous bilayer membrane electrode is provided, including the following steps:

step 1: treatment of a gas diffusion layer

soaking a gas diffusion layer in a solution of hydrophobic polymers with a certain concentration for 30 s to 90 s; drying the gas diffusion layer in an oven; and then subjecting the dried gas diffusion layer to sintering in a muffle furnace to obtain a gas diffusion layer with a hydrophobic surface, where the hydrophobic polymer has a mass fraction of 20 wt.% to 40 wt.% in the gas diffusion layer;

step 2: preparation of an outer microporous layer

uniformly spraying a slurry including carbon powder and hydrophobic polymers on the gas diffusion layer with a hydrophobic surface obtained in step 1; drying the carbon paper sprayed with the slurry in an oven; and then subjecting the dried carbon paper to sintering in a muffle furnace to obtain an outer microporous layer, where PTFE has a mass fraction of 15 wt.% to 30 wt.% in the outer microporous layer;

step 3: preparation of an inner microporous layer

uniformly spraying a slurry including carbon powder and proton-conductive polymers on the surface of the outer microporous layer obtained in step 2, and drying the microporous layer sprayed with the slurry in an oven to obtain a microporous layer having a bilayer structure, where the proton-conductive polymer has a mass fraction of 20 wt.% to 40 wt.% in the inner microporous layer;

step 4: preparation of a catalyst layer

evenly spraying a catalyst slurry including proton conductors on the microporous layer having a bilayer structure obtained in step 3, and drying the resulting microporous layer in an oven for a period of time to obtain a dried DMFC electrode for improving catalyst utilization, where the catalyst is Pt/C or Pt Ru/C with 40 wt.% to 70 wt.% of metal, in percentage by weight; and

step 5: assembly of a membrane electrode

pressing the DMFC electrode for improving catalyst utilization obtained in step 4 and a proton exchange membrane with ion conductivity together at room temperature under a pressure of 6.0 N·m to 8.0 N·m, without heat pressing, to obtain a DMFC membrane electrode for improving catalyst utilization.

Further, in step 1, the gas diffusion layer is soaked in hydrophobic polymers for 30 s to 90 s; and the sintering in a muffle furnace is conducted at 350°C to 400°C for 40 min to 60 min.

Further, the gas diffusion layer in step 1 is carbon paper, carbon cloth, nickel foam or other conductive materials that have surface inclusions removed.

Further, the hydrophobic polymer in step 1 is PTFE, polyvinyl alcohol or polyvinylidene fluoride, and has a mass fraction of 20 wt.% to 40 wt.% in the gas diffusion layer.

Further, the slurry in step 2 is a mixed solution of carbon powder, hydrophobic polymers and a dispersing solvent, and the hydrophobic polymer has a mass fraction of 15 wt.% to 30 wt.% in the outer microporous layer.

Further, the slurry in step 3 is a mixed solution of carbon powder, proton-conductive polymers and a dispersing solvent, and the dispersing solvent has a volume of 5 ml to 10 ml.

Further, the proton-conductive polymer in step 3 is Nafion; the dispersing solvent is isopropyl alcohol, ethanol or acetone; and Nafion has a mass fraction of 20 wt.% to 40 wt.%

in the inner microporous layer.

Further, in step 4, the anode catalyst is Pt/C or Pt Ru/C with 40 wt.% to 70 wt.% of metal, in percentage by weight, and the anode catalyst layer has a metal loading of 2 mg cm⁻² to 5 mg cm⁻²; and the cathode catalyst is Pt/C or Pt Ru/C with 40 wt.% to 70 wt.% of metal, in percentage by weight, and the cathode catalyst layer has a metal loading of 1.5 mg cm⁻² to 3 mg cm⁻².

Further, in step 4, the proton conductor in the catalyst layer has a mass fraction of 25 wt.% to 40 wt.%.

Further, the proton exchange membrane in step 5 is a perfluorosulfonic acid membrane, and the DMFC electrode and the proton exchange membrane are pressed together at room temperature under a pressure of 6.0 N m to 8.0 N m.

(1) Treatment of a gas diffusion layer

A gas diffusion layer is soaked in a solution of PTFE with a certain concentration for 30 s; the carbon paper is dried in an oven; and then the dried carbon paper is subjected to sintering in a muffle furnace to obtain a carbon paper with a hydrophobic surface, where PTFE has a mass fraction of 20 wt.% to 30 wt.% in the gas diffusion layer.

(2) Preparation of an outer microporous layer

A slurry including carbon powder and PTFE is uniformly sprayed on the carbon paper with a hydrophobic surface obtained in step (1); the carbon paper sprayed with the slurry is dried in an oven; and then the dried carbon paper is subjected to sintering in a muffle furnace to obtain an outer microporous layer, where PTFE has a mass fraction of 15 wt.% to 20 wt.% in the outer microporous layer.

(3) Preparation of an inner microporous layer

A slurry including carbon powder and Nafion is uniformly sprayed on the surface of the outer microporous layer obtained in step (2); and the microporous layer sprayed with the slurry is dried in an oven to obtain a microporous layer having a bilayer structure, where Nafion has a mass fraction of 25 wt.% to 35 wt.% in the inner microporous layer.

(4) Preparation of a catalyst layer

A catalyst slurry including proton conductors is evenly sprayed on the microporous layer having a bilayer structure obtained in step (3), and the resulting microporous layer is dried in

an oven for a period of time to obtain a dried DMFC electrode for improving catalyst utilization, where the catalyst is Pt/C or Pt Ru/C with 40 wt.% to 70 wt.% of metal, in percentage by weight.

(5) Assembly of a membrane electrode

The DMFC electrode for improving catalyst utilization obtained in step 4 and a proton exchange membrane with ion conductivity are pressed together at room temperature under a pressure of 7.5 N·m, without heat pressing, to obtain a DMFC membrane electrode for improving catalyst utilization.

Compared with a traditional microporous monolayer membrane electrode, the microporous bilayer membrane electrode provided in the present invention has the following advantages:

(1) Larger electrochemically active area

After the battery has been running for a period of time, part of the catalyst will sink into the microporous layer due to assembly pressure, methanol feeding and gas flow. As there are no proton conductors in a conventional microporous monolayer electrode structure, this part of the catalyst cannot be utilized, and thus the three-phase reaction interface is reduced. In the microporous bilayer electrode provided in the present invention, owing to proton conductors added in the inner microporous layer, this part of the catalyst can be utilized, and thus the electrochemically active area is increased.

(2) Higher catalyst utilization

The inner microporous layer described in the present invention allows the part of the catalyst leaked into the microporous layer to be utilized, which improves the catalyst utilization, and thus greatly reduces the preparation cost for an electrode.

(3) Lower mass transfer resistance

The microporous monolayer of a conventional electrode is usually a hydrophobic structure, which makes it difficult to feed methanol at a low concentration, increases the mass transfer resistance in the electrode, and thus compromises the performance of a single cell. The microporous bilayer of the present invention is added with Nafion having high hydrophilicity to strengthen the methanol transport and reduce the mass transfer resistance for the electrode.

BRIEF DESCRIPTION OF DRAWINGS

FIG. 1 is a schematic diagram of the structure of the DMFC membrane electrode for improving catalyst utilization according to the present invention;

FIG. 2 is a flow chart of the preparation process of the DMFC electrode for improving catalyst utilization according to the present invention;

FIG. 3 is a flow chart of assembly process of the DMFC membrane electrode for improving catalyst utilization according to the present invention;

FIG. 4 is a discharge performance curve for the fuel cell of Example 1;

FIG. 5 is a discharge performance curve for the fuel cell of Example 2;

FIG. 6 is a discharge performance curve for the fuel cell of Example 3;

FIG. 7 is a discharge performance curve for the fuel cell of Example 4;

FIG. 8 is a discharge performance curve for the fuel cell of Comparative Example 1;

FIG. 9 is a discharge performance curve for the fuel cell of Comparative Example 2;

FIG. 10 is a discharge performance curve for the fuel cell of Comparative Example 3;

FIG. 11 is a discharge performance curve for the fuel cell of Comparative Example 4.

Reference numerals in the drawings are as follows:

1: gas diffusion layer; 2: microporous layer including PTFE; 3: microporous layer including Nafion; 4: anode catalyst layer; 5: proton exchange membrane; 6: cathode catalyst layer.

DETAILED DESCRIPTION

Example 1

DMFC electrode and membrane electrode for improving catalyst utilization were prepared according to the process shown in FIG. 2, and a discharge test was performed. The main steps were as follows:

(1) Preparation of an electrode

A carbon paper including PTFE was adopted as a diffusion layer, and a microporous layer 2 including PTFE was coated on the hydrophobic diffusion layer, where PTFE had a

content of 15 wt.%. The outer microporous layer was coated with a microporous layer 3 including Nafion, where Nafion had a content of 30 wt.%. A catalyst slurry was prepared at an appropriate ratio, and isopropyl alcohol was used as a solvent. Pt Ru/C was used as catalyst on the anode side, and Pt/C was used as catalyst on the cathode side. A catalyst slurry was sprayed on the microporous layer 3 including Nafion to form a catalyst layer.

(2) Treatment of a proton exchange membrane

The membrane was boiled in a 5 wt.% H_2O_2 solution for 1 h, then washed in deionized water, then boiled in a 0.5 M sulfuric acid solution for 1 h, and finally boiled in deionized water for 1 h. The pretreated membrane was stored in deionized water before membrane electrode assembly (MEA) was prepared by pressing.

(3) Assembly of a membrane electrode

Two electrodes having a microporous bilayer structure and Nafion 212 membrane were pressed together at room temperature under a pressure of 7.5 N·m, without heat pressing, to obtain a DMFC membrane electrode for improving catalyst utilization described in the present invention.

(4) Discharge performance test

The obtained MEA and a sealed air cushion were assembled in a single cell for testing. The test was performed under the following conditions: battery operating temperature: 60°C; atmospheric pressure; anode fuel: 0.5 M methanol (at a flow rate of 3 ml min⁻¹); and cathode intake air: dry oxygen (at a flow rate of 199 ml min⁻¹). The limit current density could reach 120.17 mA cm⁻² and the maximum power density could reach 22.23 mW cm⁻². Compared with Comparative Example 1, Example 1 had a maximum power density increased by 56.53%.

Example 2

The DMFC membrane electrode for improving catalyst utilization described in the present invention was tested under conditions of high-concentration methanol and dry oxygen. First, a microporous bilayer electrode was prepared according to the same procedure as in Example 1.

The obtained MEA and a sealed air cushion were assembled in a single cell for testing. The test was performed under the following conditions: battery operating temperature: 60°C; atmospheric pressure; anode fuel: 2 M methanol (at a flow rate of 3 ml min⁻¹); and cathode

intake air: dry oxygen (at a flow rate of 199 ml min⁻¹). The limit current density could reach 500.21 mA cm⁻² and the maximum power density could reach 76.29 mW cm⁻². Compared with Comparative Example 2, Example 2 had a maximum power density increased by 41.01%.

Example 3

The DMFC membrane electrode for improving catalyst utilization described in the present invention was tested under conditions of low-concentration methanol and humidified oxygen. First, a microporous bilayer electrode was prepared according to the same procedure as in Example 1.

The obtained MEA and a sealed air cushion were assembled in a single cell for testing. The test was performed under the following conditions: battery operating temperature: 60°C; atmospheric pressure; anode fuel: 0.5 M methanol (at a flow rate of 3 ml min⁻¹); and cathode intake air: humidified oxygen (with a relative humidity of 60% and at a flow rate of 199 ml min⁻¹). The limit current density could reach 135.05 mA cm⁻² and the maximum power density could reach 29.12 mW cm⁻². Compared with Comparative Example 3, Example 3 had a maximum power density increased by 97.38%.

Example 4

The DMFC membrane electrode for improving catalyst utilization described in the present invention was tested under conditions of high-concentration methanol and humidified oxygen. First, a microporous bilayer electrode was prepared according to the same procedure as in Example 1.

The obtained MEA and a sealed air cushion were assembled in a single cell for testing. The test was performed under the following conditions: battery operating temperature: 60°C; atmospheric pressure; anode fuel: 2 M methanol (at a flow rate of 3 ml min⁻¹); and cathode intake air: humidified oxygen (with a relative humidity of 60% and at a flow rate of 199 ml min⁻¹). The limit current density could reach 550.12 mA cm⁻² and the maximum power density could reach 81.04 mW cm⁻². Compared with Comparative Example 4, Example 4 had a maximum power density increased by 36.65%.

Comparative Example 1

The fuel cell electrode and membrane electrode having a conventional microporous layer structure were prepared for discharge performance comparison. The steps were as follows:

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(1) Preparation of a membrane electrode: A carbon paper including PTFE was adopted as an anode diffusion layer. An outer microporous layer was coated on the hydrophobic diffusion layer, where PTFE had a content of 15 wt.%. An appropriate amount of catalyst was dispersed in a mixed solution of deionized water, isopropyl alcohol and Nafion to obtain a catalyst slurry. Pt Ru/C was used as catalyst on the anode side, and Pt/C was used as catalyst on the cathode side. A catalyst slurry was coated on the microporous layer to form a catalyst layer. Nafion 212 membrane was pretreated to remove organic and inorganic contaminants. The pretreatment process was as follows: membrane was boiled in a 5 wt.% H₂O₂ solution for 1 h, then washed in deionized water, then boiled in a 0.5 M sulfuric acid solution for 1 h, and finally boiled in deionized water for 1 h. The pretreated membrane was stored in deionized water before MEA was assembled.

(2) Assembly of a membrane electrode: The electrolyte membrane was Nafion 212 membrane. Two prepared identical gas diffusion electrodes were placed on two sides of the electrolyte membrane respectively, and these components were pressed together under an assembly pressure of 7.5 N·m to obtain a three-in-one MEA.

(3) Single cell test: The obtained three-in-one MEA and a sealed air cushion were assembled in a single cell for testing, and the test was conducted under the same conditions as in Example 1. The limit current density reached 99.98 mA cm⁻² and the maximum power density reached 14.20 mW cm⁻².

Comparative Example 2

The fuel cell electrode and membrane electrode having a conventional microporous layer structure were tested under conditions of high-concentration methanol and dry oxygen for discharge performance comparison.

First, fuel cell electrode and membrane electrode having a conventional microporous layer structure were prepared and assembled according to the same procedure as in Comparative Example 1. The obtained three-in-one MEA and a sealed air cushion were assembled in a single cell for testing, and the test was conducted under the same conditions as in Example 2. The limit current density reached 350.07 mA cm⁻² and the maximum power density reached 54.11 mW cm⁻².

Comparative Example 3

The fuel cell electrode and membrane electrode having a conventional microporous layer structure were tested under conditions of low-concentration methanol and humidified oxygen

for discharge performance comparison.

First, fuel cell electrode and membrane electrode having a conventional microporous layer structure were prepared and assembled according to the same procedure as in Comparative Example 1. The obtained three-in-one MEA and a sealed air cushion were assembled in a single cell for testing, and the test was conducted under the same conditions as in Example 3. The limit current density reached 82.34 mA cm^{-2} and the maximum power density reached 14.75 mW cm^{-2} .

Comparative Example 4

The fuel cell electrode and membrane electrode having a conventional microporous layer structure were tested under conditions of low-concentration methanol and humidified oxygen for discharge performance comparison.

First, fuel cell electrode and membrane electrode having a conventional microporous layer structure were prepared and assembled according to the same procedure as in Comparative Example 1. The obtained three-in-one MEA and a sealed air cushion were assembled in a single cell for testing, and the test was conducted under the same conditions as in Example 4. The limit current density reached $375.04 \text{ mA cm}^{-2}$ and the maximum power density reached 59.30 mW cm^{-2} .

It can be seen from comparative examples that the DMFC electrode and membrane electrode for improving catalyst utilization according to the present invention have better discharge performance. At a temperature of 60°C , with either humidified or dry oxygen, compared with comparative examples, all examples have significantly-increased maximum discharge current density and maximum power density. Compared with Comparative Example 1, Example 1 has a maximum power density increased by 56.53%; compared with Comparative Example 2, Example 2 has a maximum power density increased by 41.01%; compared with Comparative Example 3, Example 3 has a maximum power density increased by 97.38%; and compared with Comparative Example 4, Example 4 has a maximum power density increased by 36.65%. It indicates that the introduction of the inner microporous layer allows the part of the catalyst that has sunk into the microporous layer due to the operation of the battery and thus cannot be utilized to be utilized, which improves the catalyst utilization. The microporous bilayer structure increases the electrochemically active area of the battery, reduces the mass transfer resistance, and promotes the electrochemical reaction efficiency and mass transfer, thereby effectively improving the discharge performance of the battery.

It should be noted that, according to the examples of the present invention, those skilled in the art can completely implement the full scope of the independent claims and dependent claims of the present invention, and the implementation process and method are the same as those of the above examples. The part of the present invention that is not elaborated belongs to the technology well known in the art.

The above descriptions are merely some specific implementations of the present invention, but the protection scope of the present invention is not limited thereto. Any modification or replacement easily conceived by those skilled in the art within the technical scope of the present invention should fall within the protection scope of the present invention.

What is claimed is:

1. A direct methanol fuel cell (DMFC) membrane electrode for improving catalyst utilization, comprising a gas diffusion layer, a microporous layer, a catalyst layer and a proton exchange membrane, wherein the microporous layer in the DMFC membrane electrode has a bilayer structure, comprising an outer microporous layer with hydrophobic polymers and an inner microporous layer with proton-conductive polymers.

2. A preparation method of the DMFC membrane electrode for improving catalyst utilization according to claim 1, comprising the following steps:

step 1: treatment of a gas diffusion layer: soaking a gas diffusion layer in a solution of hydrophobic polymers with a certain concentration; after a certain period of time, drying the gas diffusion layer in an oven; and then subjecting the dried gas diffusion layer to sintering in a muffle furnace to obtain a gas diffusion layer with a hydrophobic surface;

step 2: preparation of an outer microporous layer: uniformly spraying a slurry comprising carbon powder and hydrophobic polymers on the gas diffusion layer with a hydrophobic surface obtained in step 1; drying the gas diffusion layer sprayed with the slurry in an oven; and then subjecting the dried gas diffusion layer to sintering in a muffle furnace to obtain an outer microporous layer;

step 3: preparation of an inner microporous layer: uniformly spraying a slurry comprising carbon powder and proton-conductive polymers on the surface of the outer microporous layer obtained in step 2; and drying the microporous layer sprayed with the slurry in an oven to obtain a microporous layer having a bilayer structure;

step 4: preparation of a catalyst layer: evenly spraying a catalyst slurry comprising proton conductors on the microporous layer having a bilayer structure obtained in step 3, and drying the resulting microporous layer in an oven for a period of time to obtain a dried DMFC electrode for improving catalyst utilization; and

step 5: assembly of a membrane electrode: pressing the DMFC electrode for improving catalyst utilization obtained in step 4 and a proton exchange membrane with ion conductivity together to obtain a DMFC membrane electrode for improving catalyst utilization.

3. The preparation method of the DMFC membrane electrode for improving catalyst utilization according to claim 2, wherein, in step 1, the gas diffusion layer is soaked in hydrophobic polymers for 30 s to 90 s; and the sintering in a muffle furnace is conducted at

350°C to 400°C for 40 min to 60 min.

4. The preparation method of the DMFC membrane electrode for improving catalyst utilization according to claim 2, wherein the gas diffusion layer in step 1 is carbon paper, carbon cloth, nickel foam or other conductive materials that have surface inclusions removed;

wherein the hydrophobic polymer in step 1 is polytetrafluoroethylene (PTFE), polyvinyl alcohol or polyvinylidene fluoride, and has a mass fraction of 20 wt.% to 40 wt.% in the gas diffusion layer;

wherein the slurry in step 2 is a mixed solution of carbon powder, hydrophobic polymers and a dispersing solvent, and the hydrophobic polymer has a mass fraction of 15 wt.% to 30 wt.% in the outer microporous layer;

wherein the slurry in step 3 is a mixed solution of carbon powder, proton-conductive polymers and a dispersing solvent, and the dispersing solvent has a volume of 5 ml to 10 ml.

5. The preparation method of the DMFC membrane electrode for improving catalyst utilization according to claim 2, wherein, in step 4, the anode catalyst is Pt/C or Pt Ru/C with 40 wt.% to 70 wt.% of metal, in percentage by weight, and the anode catalyst layer has a metal loading of 2 mg cm⁻² to 5 mg cm⁻²; and the cathode catalyst is Pt/C or Pt Ru/C with 40 wt.% to 70 wt.% of metal, in percentage by weight, and the cathode catalyst layer has a metal loading of 1.5 mg cm⁻² to 3 mg cm⁻²;

wherein, in step 4, the proton conductor in the catalyst layer has a mass fraction of 25 wt.% to 40 wt.%;

wherein, the proton exchange membrane in step 5 is a perfluorosulfonic acid membrane, and the DMFC electrode and the proton exchange membrane are pressed together at room temperature under a pressure of 6.0 N m to 8.0 N m.

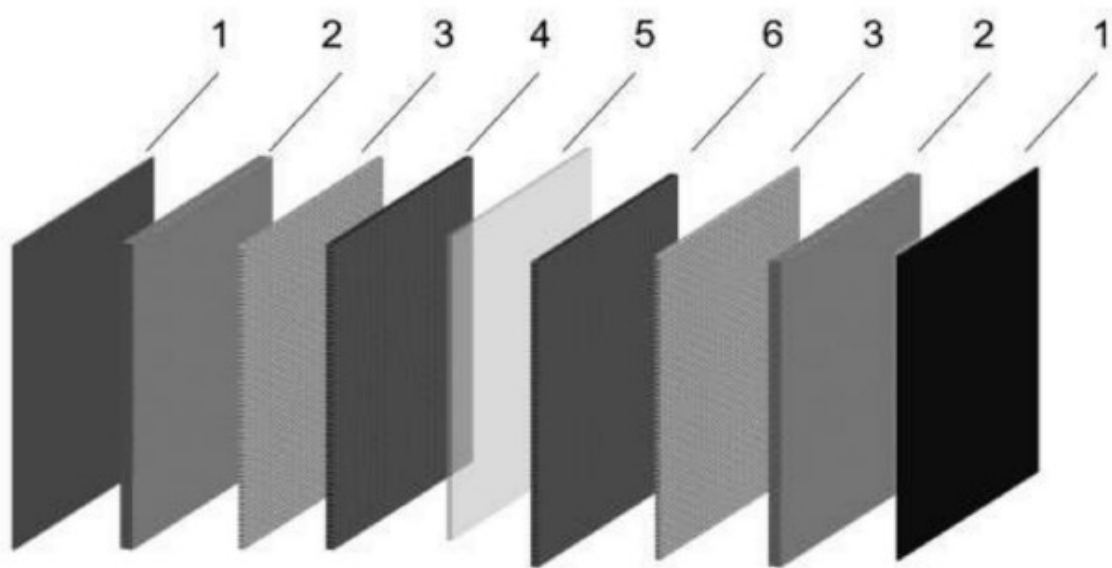


FIG. 1

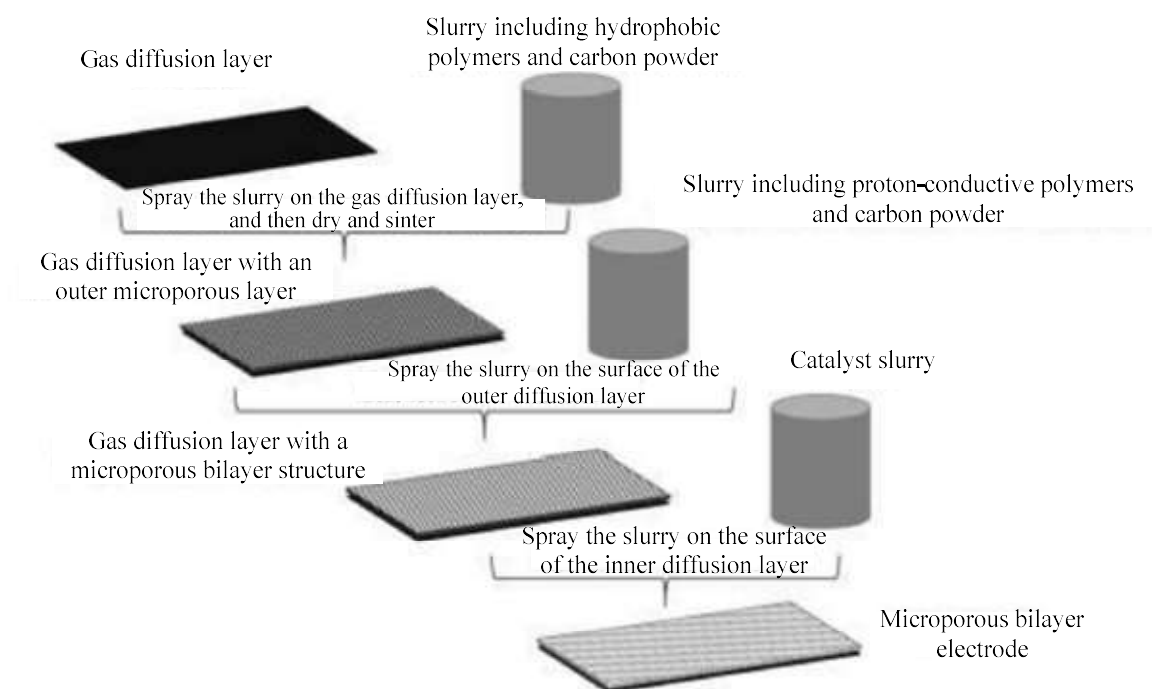


FIG. 2

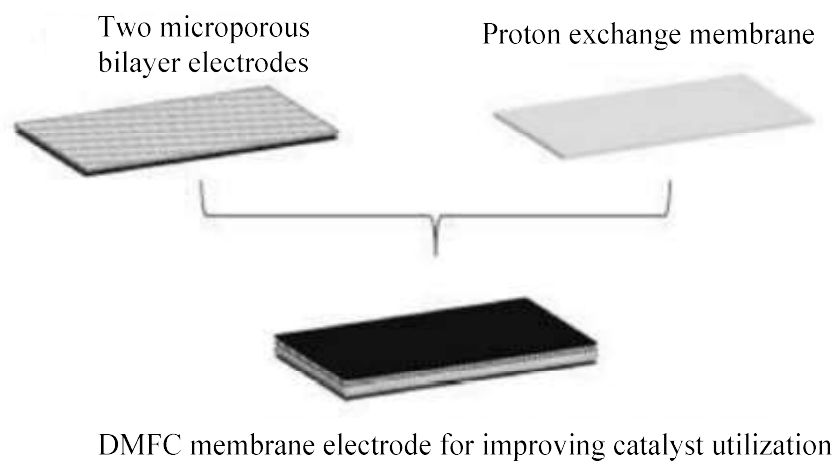


FIG. 3

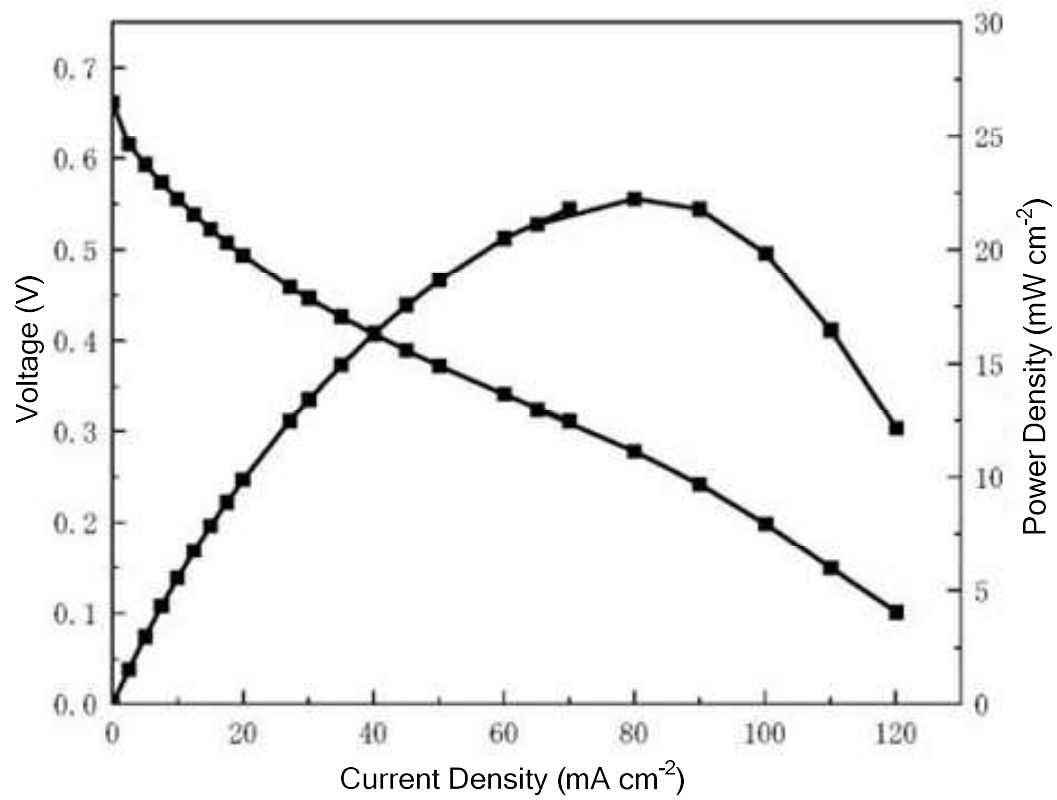


FIG. 4

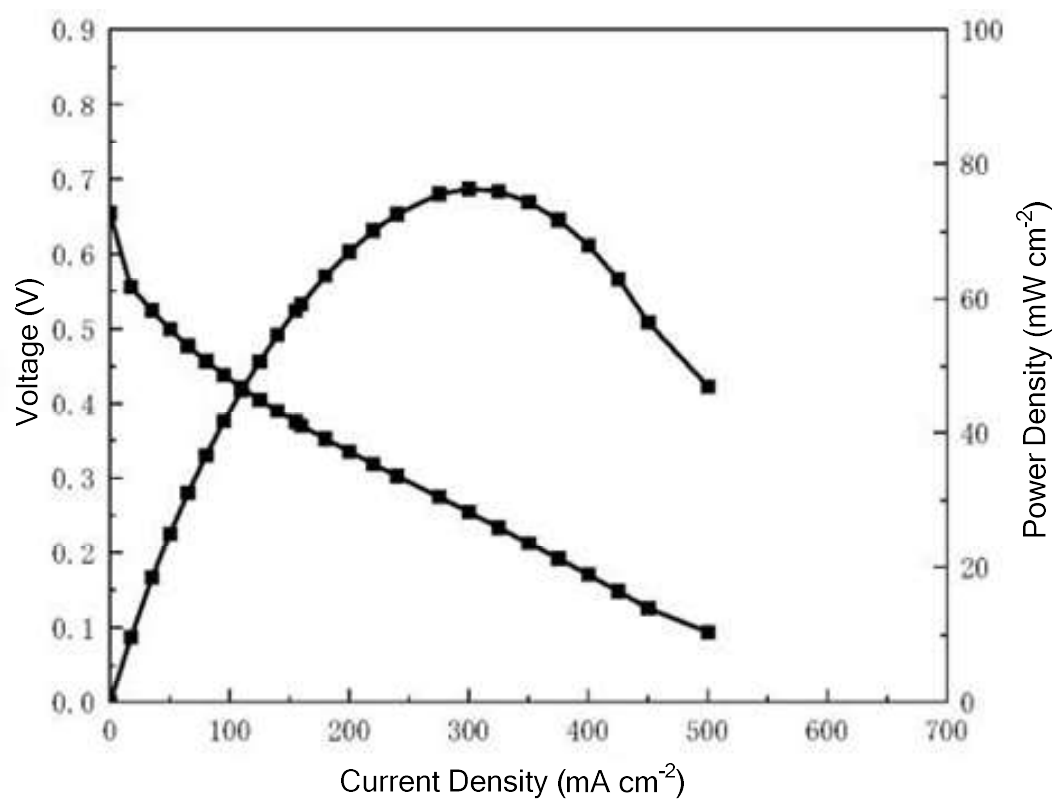


FIG. 5

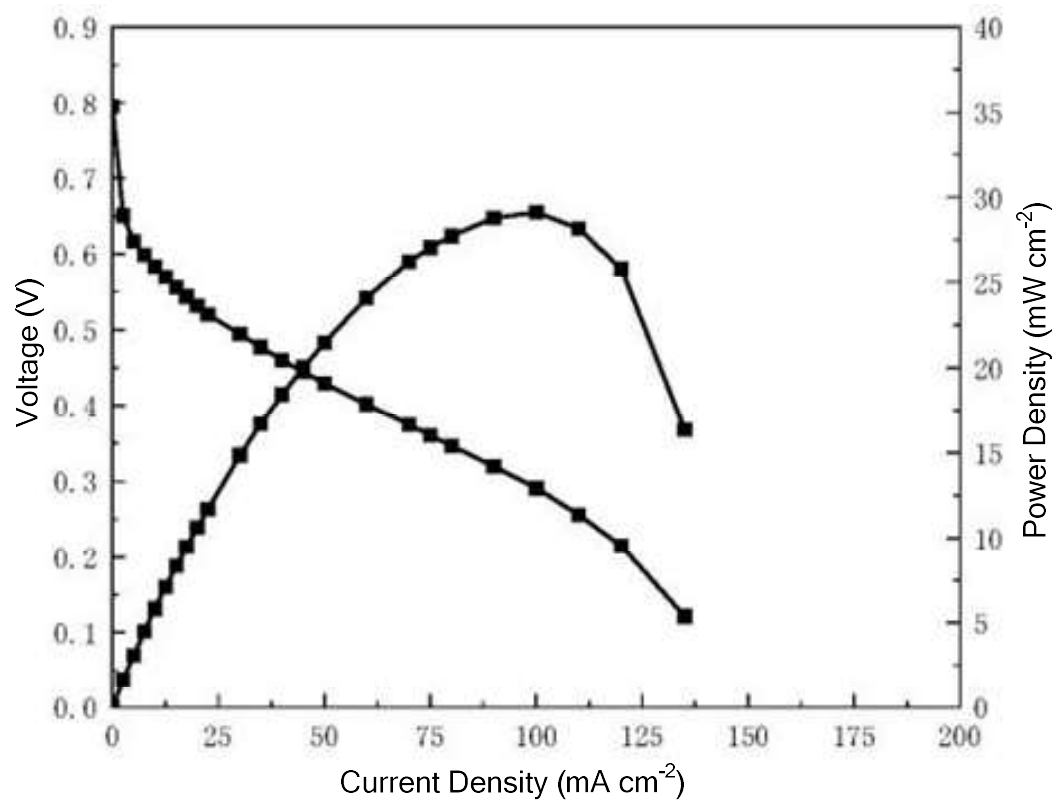


FIG. 6

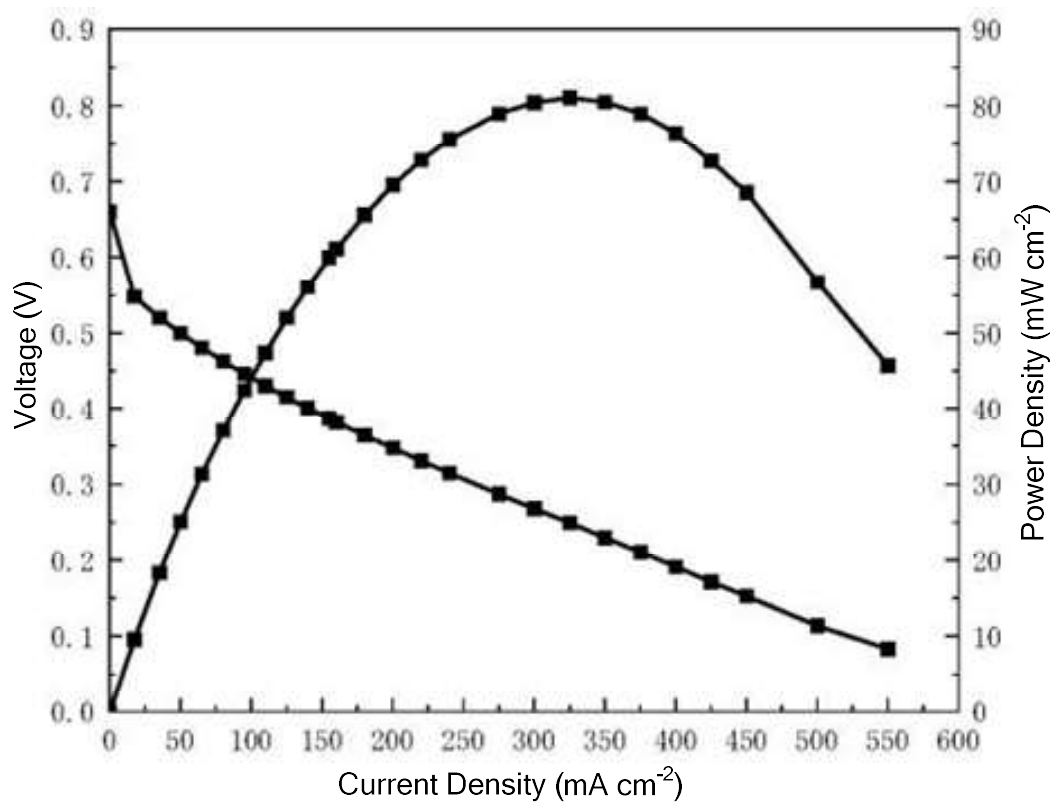


FIG. 7

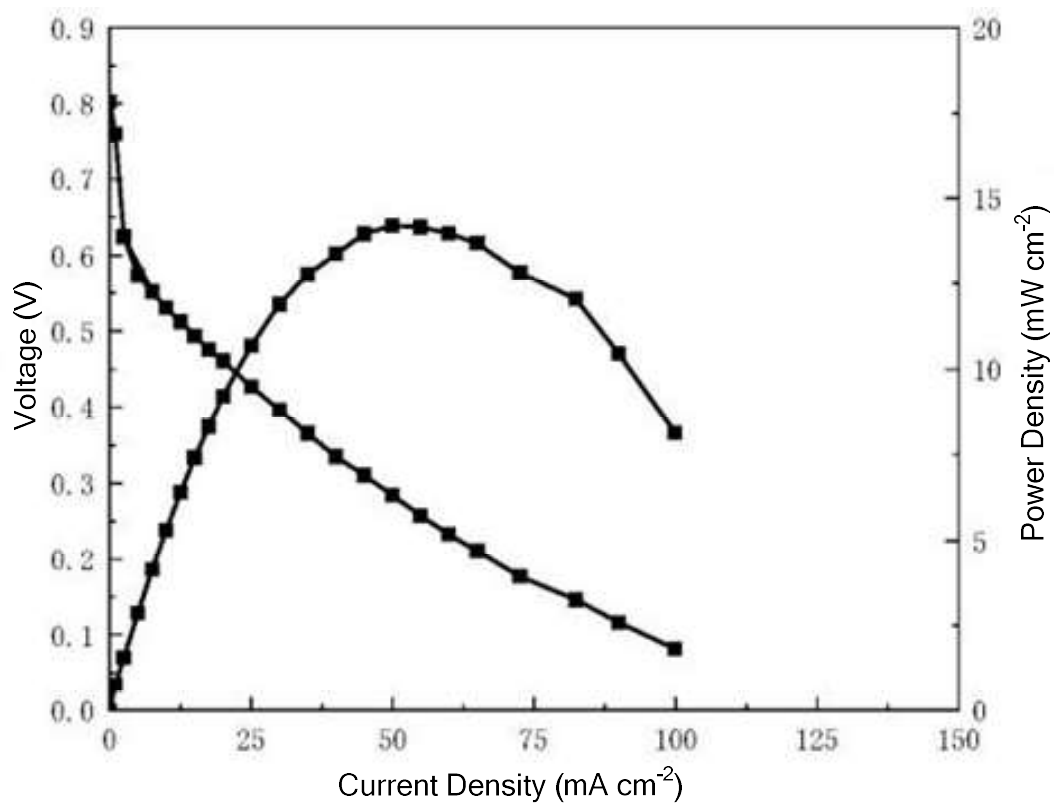


FIG. 8

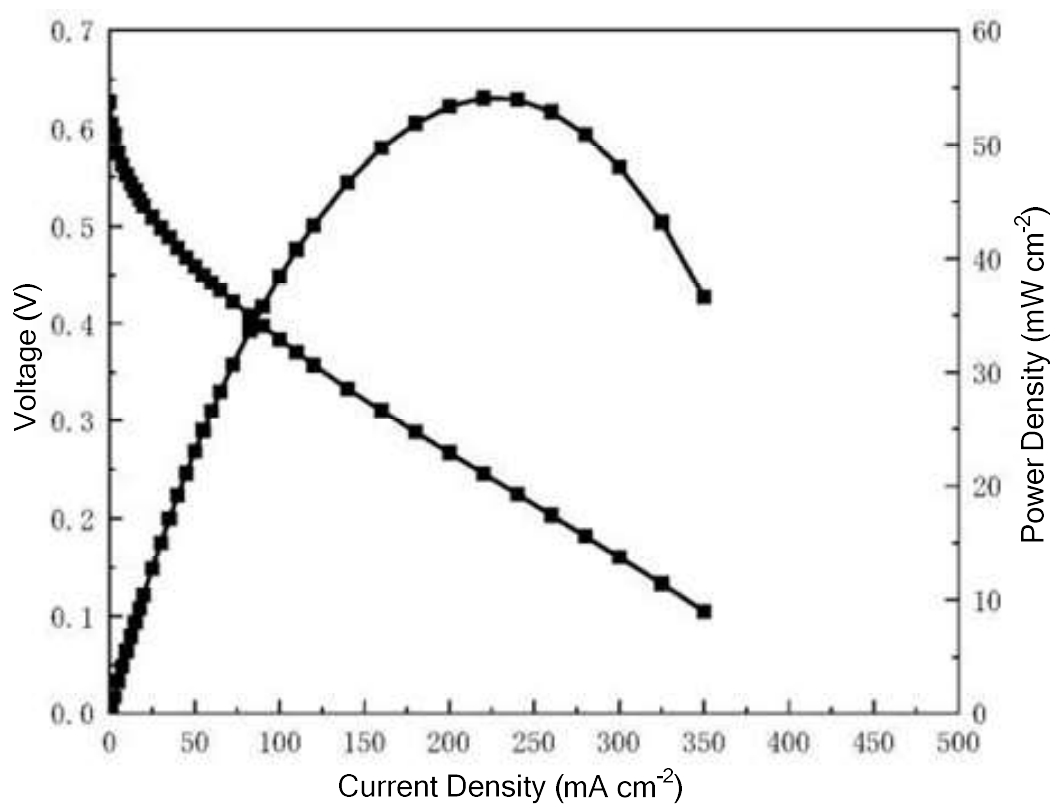


FIG. 9

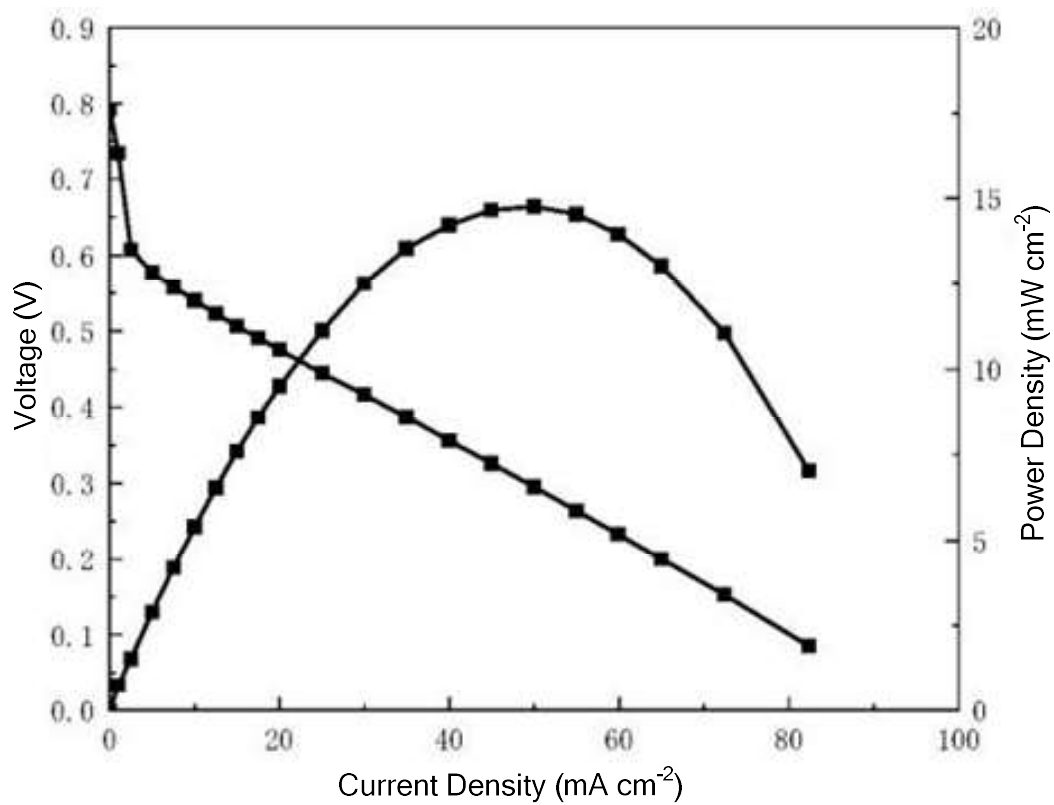


FIG. 10

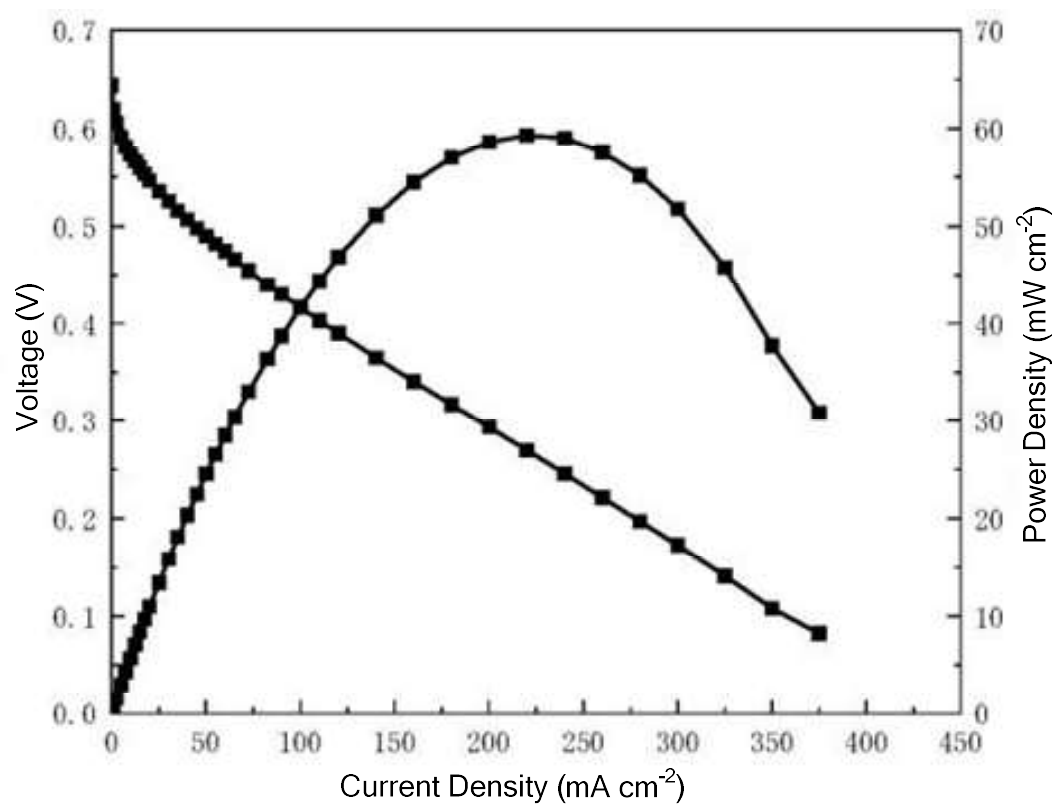


FIG. 11