

PREPARATION METHODS FOR CATALYST LAYERS AND MEMBRANE ELECTRODE ASSEMBLIES FOR FUEL CELLS

CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] This application claims priority to the Chinese Patent Application No. 2024115375649, filed on October 31, 2024, the contents of which are hereby incorporated by reference.

TECHNICAL FIELD

[0002] The present disclosure generally relates to a field of fuel cell technology, and in particular to a preparation method for a catalyst layer and a membrane electrode assembly for fuel cells.

BACKGROUND

[0003] Fuel cells, as efficient and clean energy conversion devices, have attracted extensive attention and have been widely researched in recent years. The fuel cells directly generate electrical energy through the chemical reaction of hydrogen and oxygen, offering advantages such as high energy conversion efficiency and minimal environmental pollution. In the fuel cells, a catalyst layer is one of the key components, directly affecting the performance and efficiency of the fuel cells. The main function of the catalyst layer is to promote the electrochemical reaction of fuel and oxidant on the electrode surface, thereby improving the output power and stability of the fuel cells.

[0004] Water management in the fuel cells is particularly challenging under low humidity or non-humidified conditions. During operation, the moisture in a proton exchange membrane is lost significantly, which can cause deformation of the catalyst layer and affect proton transport efficiency. Simultaneously, the water management materials currently used have poor electrical conductivity, affecting the establishment of an electron transport network within the catalyst layer and consequently impacting fuel cells performance.

[0005] In view of this, it is necessary to provide the preparation method for the catalyst layer and the membrane electrode assembly for the fuel cells, which may solve technical problems such as poor water management effect of the catalyst layer for the fuel cells and poor conductivity of the water management material used.

SUMMARY

[0006] One or more embodiments of the present disclosure provide a membrane

electrode with a catalyst layer for fuel cells, wherein the catalyst layer includes an anode catalyst and a cathode catalyst, and the anode catalyst layer includes a Pt-C catalyst, a high-dielectric small-molecule alcohol, sulfonated lignin-doped conductive polymer nanoparticles, a hydrophilic binder, an ionomer solution, and deionized water; the cathode catalyst layer includes a catalyst, the ionomer solution, the high-dielectric small-molecule alcohol, polytetrafluoroethylene, the deionized water; in the anode catalyst layer, within the Pt-C catalyst, a loading amount of Pt is 0.03-0.3 mg/cm², mass of an effective component in the ionomer solution is 20-80% of mass of C in the Pt-C catalyst, mass of the Pt-C catalyst is 5-20% of mass of the deionized water, a mass ratio of the deionized water to the high-dielectric small-molecule alcohol is 1:(1-4), mass of the sulfonated lignin-doped conductive polymer nanoparticles is 0.5-5% of the mass of the Pt-C catalyst, and mass of the hydrophilic binder is 2-5% of the mass of the deionized water; in the cathode catalyst layer, the catalyst of the cathode catalyst layer is at least one of a Pt-C catalyst and a Ni-Fe catalyst; in the Pt-C catalyst, a loading amount of Pt is 0.3-0.7 mg/cm², mass of an effective component in the ionomer solution is 30-80% of mass of C in the Pt-C catalyst, mass of the Pt-C catalyst is 5-20% of mass of the deionized water, a mass ratio of the deionized water to the high-dielectric small-molecule alcohol is 1:(1-4), and mass of the polytetrafluoroethylene is 1-5% of the mass of the deionized water; a preparation process of the membrane electrode includes: a preparation of an anode-side adhesive layer: dispensing a conductive polymer-sulfonated lignin hydrogel onto a peripheral area of a surface of the anode catalyst layer, thereby forming the anode-side adhesive layer configured for bonding the anode catalyst layer; a preparation of the cathode-side adhesive layer: dispensing the ionomer solution onto a peripheral area of a surface of the cathode catalyst layer, thereby forming the cathode-side adhesive layer configured for bonding a cathode diffusion layer, wherein the ionomer solution is a Nafion-PEDOT conductive polymer, and the membrane electrode assembly employs a frame-like encapsulation; wherein the high-dielectric small-molecule alcohol includes at least one of ethanol, ethylene glycol, n-propanol, and isopropanol; the ionomer solution includes at least one of a sulfonated poly(ether ether ketone) solution, a sulfonated polystyrene solution, a sulfonated polysulfone solution, a sulfonated polyethylene solution, and a perfluorosulfonic acid resin solution; and the hydrophilic binder includes glycerol and polyethylene glycol (PEG).

BRIEF DESCRIPTION OF THE DRAWINGS

[0007] The present disclosure will be further illustrated by way of exemplary embodiments, which will be described in detail with reference to the accompanying drawings. These embodiments are not limiting, and in these embodiments, the same reference numerals denote the same structures, wherein:

[0008] FIG. 1 is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to some embodiments of the present disclosure;

[0009] FIG. 1a is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to Embodiment 1 of the present disclosure;

[0010] FIG. 1b is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to Embodiment 3 of the present disclosure;

[0011] FIG. 1c is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to Embodiment 2 and Comparative Embodiment 1 of the present disclosure;

[0012] FIG. 1d is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to Embodiment 2 and Comparative Embodiment 2 of the present disclosure;

[0013] FIG. 2 is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to some embodiments of the present disclosure;

[0014] FIG. 2a is a schematic diagram illustrating a variation of a power of a fuel cell with time according to Embodiment 1 of the present disclosure;

[0015] FIG. 2b is a schematic diagram illustrating a variation of a power of a fuel cell with time according to Embodiment 3 of the present disclosure;

[0016] FIG. 2c is a schematic diagram illustrating a variation of a power of a fuel cell with time according to Embodiment 2 and Comparative Embodiment 1 of the present disclosure;

[0017] FIG. 2d is a schematic diagram illustrating a variation of a power of a fuel cell with time according to Embodiment 2 and Comparative Embodiment 2 of the present disclosure.

DETAILED DESCRIPTION

[0018] To more clearly illustrate the technical solutions of the embodiments of the present disclosure, the accompanying drawings required for describing the embodiments are briefly introduced below. Obviously, the accompanying drawings in the following

description are merely some examples or embodiments of the present disclosure. For those skilled in the art, the present disclosure may be applied to other similar scenarios based on these accompanying drawings without creative effort. Unless obviously obtained from the context or the context illustrates otherwise, the same numeral in the drawings refers to the same structure or operation.

[0019] A fuel cell is a device that directly generates electrical energy through an electrochemical reaction between fuel (e.g., hydrogen, methanol, methane, etc.) and oxidizer (e.g., oxygen, air, etc.). Traditional power generation devices require a plurality of conversions of energy from chemical to thermal to electrical. In contrast, the fuel cells directly convert the chemical energy to electrical energy in a single step, resulting in higher energy conversion efficiency, more complete reactions, and emissions typically limited to water and carbon dioxide, making it a greener and cleaner technology. Therefore, the fuel cells are widely applicable to scenarios such as distributed energy and new energy vehicles.

[0020] In some embodiments, the present disclosure provides a membrane electrode for a catalyst layer of the fuel cells. The catalyst layer includes an anode catalyst and a cathode catalyst.

[0021] The catalyst refers to a substance having electrocatalytic activity. In some embodiments, the anode catalyst and the cathode catalyst may exist in the catalyst layer in various forms. For example, the anode catalyst and the cathode catalyst may be anode catalyst particles and cathode catalyst particles. As another example, the anode catalyst and the cathode catalyst may be an anode catalyst film and a cathode catalyst film.

[0022] In some embodiments, the anode catalyst and the cathode catalyst may be an anode catalyst layer and a cathode catalyst layer. That is, the catalyst layer includes the anode catalyst layer and the cathode catalyst layer. The anode catalyst layer and the cathode catalyst layer are located on two sides of a proton exchange membrane, respectively.

[0023] The membrane electrode is a core component of the fuel cell. The membrane electrode implements an electrochemical reaction through the synergistic effect of the anode catalyst layer and the cathode catalyst layer.

[0024] In some embodiments, when an organic compound (e.g., methanol, methane, etc.) is configured as the fuel, the anode catalyst layer of the fuel cell is configured to consume the fuel and convert the fuel and water into protons, carbon dioxide, and

electrons. The generated electrons are transmitted to an external circuit to supply power. The generated protons are conducted to the cathode catalyst layer side through the proton exchange membrane. The cathode catalyst layer of the fuel cell is configured to consume oxygen. The cathode catalyst layer receives electrons from the external circuit and converts oxygen, protons, and electrons into water.

[0025] Merely by way of example, when the methane and air are configured as the reaction system, the reaction in the anode catalyst layer is: $\text{CH}_4 + 2\text{H}_2\text{O} \rightarrow 2\text{CO}_2 + 8\text{H}^+ + 8\text{e}^-$. The reaction in the cathode catalyst layer is: $2\text{O}_2 + 8\text{H}^+ + 8\text{e}^- \rightarrow 4\text{H}_2\text{O}$. The overall reaction of the fuel cell is: $\text{CH}_4 + 2\text{O}_2 \rightarrow \text{CO}_2 + 2\text{H}_2\text{O}$. It can be seen that the anode catalyst layer consumes a large amount of water. Hydrophilic raw materials are used to avoid drying out of the anode catalyst layer. The cathode catalyst layer requires oxygen to be introduced and simultaneously produces a large amount of water. Hydrophobic raw materials are used to transport the produced water to the anode, to avoid the cathode catalyst layer being flooded with water, and to maintain good gas transport properties.

[0026] In some embodiments, the anode catalyst layer includes a Pt-C catalyst, a high-dielectric small-molecule alcohol, sulfonated lignin-doped conductive polymer nanoparticles, a hydrophilic binder, an ionomer solution, and deionized water; the cathode catalyst layer includes a catalyst, the ionomer solution, the high-dielectric small-molecule alcohol, polytetrafluoroethylene, the deionized water.

[0027] The catalyst is a main active ingredient in the membrane electrode. The anode catalyst layer and the cathode catalyst layer may use the same or different types of catalysts based on actual needs.

[0028] In some embodiments, the anode catalyst layer uses the Pt-C catalyst, and the cathode catalyst layer uses at least one of the Pt-C catalyst and a Ni-Fe catalyst. The Pt-C catalyst is a catalyst in which a platinum-based alloy is dispersed and supported on a carbon carrier (e.g., porous graphite, etc.). The Ni-Fe catalyst is a catalyst in which a composite oxide or alloy of metallic nickel and iron is dispersed and supported on a carrier (e.g., porous graphite, porous alumina, etc.).

[0029] The sulfonated lignin-doped conductive polymer nanoparticles are hydrophilic conductive polymers, potentially enhancing both the conductivity and hydrophilicity of the anode catalyst layer.

[0030] In some embodiments, the conductive polymer includes, but is not limited to,

poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS), polyaniline (PAn), polypyrrole (PPy), polythiophene derivatives (e.g., PTAA-PAH), or the like.

[0031] In some embodiments, the sulfonated lignin-doped conductive polymer nanoparticles may be made into a conductive polymer-sulfonated lignin hydrogel, which is dispensed on a surface edge of the anode catalyst layer to form an anode-side adhesive layer for adhering the anode catalyst layer. More descriptions may be found in S3 and the related descriptions below.

[0032] The present disclosure adds hydrophilic substances to raw materials of the anode catalyst. Even under low humidity conditions or non-humidified conditions, efficient water management under high current density may still be supported. The sulfonated lignin-doped conductive polymer nanoparticles are added to the anode catalyst. The sulfonated lignin has good hydrophilicity. Furthermore, due to its highly branched three-dimensional structure, it can form a stable conductive network in cooperation with the nano-conductive particles, thereby improving the overall conductivity of the catalyst layer and achieving a synergistic effect greater than the sum of its parts.

[0033] In some embodiments, the high-dielectric small-molecule alcohol includes at least one of ethanol, ethylene glycol, n-propanol, and isopropanol.

[0034] In some embodiments, the high-dielectric small-molecule alcohol can not only improve the electrical conductivity of the catalyst layer, thereby improving the electrochemical performance of the fuel cells, but also serve as a solvent or dispersant to help the catalyst disperse uniformly in the raw materials, thereby improving the uniformity and stability of the catalyst layer.

[0035] The ionomer solution refers to a homogeneous mixture formed by polymers containing a small number of ionic groups dissolved in a solvent.

[0036] In some embodiments, the ionomer solution includes at least one of a sulfonated poly(ether ether ketone) solution, a sulfonated polystyrene solution, a sulfonated polysulfone solution, a sulfonated polyethylene solution, and a perfluorosulfonic acid resin solution.

[0037] In some embodiments, the ionomer solution has dual functions of proton conduction and binder. The ionic groups (e.g., sulfonate groups, etc.) of the ionomer solution dissociate in the solvent to form a proton transport network, ensuring rapid migration of protons in the electrochemical reaction. The ionomer solution can anchor the catalyst particles on the surface of the proton exchange membrane through molecular chain entanglement, ensuring the uniform distribution of catalytic reaction sites.

[0038] The hydrophilic binder can improve the dispersibility of the catalyst particles, thereby preventing agglomeration and clogging of catalyst particles. In some embodiments, the hydrophilic binder includes at least one of glycerol and polyethylene glycol (PEG).

[0039] The hydrophilic binder can not only serve as a dispersant to help the catalyst particles disperse uniformly in the raw materials, thereby improving the uniformity and stability of the catalyst layer, but also improve the water management performance of the catalyst layer, ensuring water distribution and transport of the proton exchange membrane under low humidity or non-humidified conditions.

[0040] The polytetrafluoroethylene is an additive for enhancing the hydrophobicity of the cathode catalyst layer.

[0041] The polytetrafluoroethylene has good hydrophobic properties. When the fuel cells operate at a high current density, a large amount of water is produced on the cathode side. The polytetrafluoroethylene can ensure that the water is rapidly discharged, avoid the cathode catalyst layer being flooded with water, and thereby maintain good gas transport channels.

[0042] In some embodiments, performing hydrophilic treatment on a diffusion layer is beneficial for controlling the content of free water inside the fuel cells and avoiding high temperature and power drop caused by water blockage.

[0043] In some embodiments, the types and ratios of substances included in the anode catalyst layer and the cathode catalyst layer may be determined based on actual scenarios.

[0044] In some embodiments, for the Pt-C catalyst of the anode catalyst layer, a loading amount of Pt is 0.03-0.3 mg/cm², mass of an effective component in the ionomer solution is 20-80% of mass of C in the Pt-C catalyst, mass of the Pt-C catalyst is 5-20% of mass of the deionized water, a mass ratio of the deionized water to the high-dielectric small-molecule alcohol is 1:(1-4), mass of the sulfonated lignin-doped conductive polymer nanoparticles is 0.5-5% of the mass of the Pt-C catalyst, and mass of the hydrophilic binder is 2-5% of the mass of the deionized water

[0045] In some embodiments, for the Pt-C catalyst of the anode catalyst layer, the loading amount of Pt may also be one of 0.03 mg/cm², 0.05 mg/cm², 0.1 mg/cm², 0.15 mg/cm², 0.2 mg/cm², 0.25 mg/cm², and 0.3 mg/cm².

[0046] In some embodiments, for the Pt-C catalyst of the anode catalyst layer, the loading amount of Pt may also be one of 0.03~0.1 mg/cm², 0.05~0.2 mg/cm², 0.1~0.3

mg/cm², 0.03~0.15 mg/cm², 0.05~0.25 mg/cm², and 0.15~0.3 mg/cm².

[0047] In some embodiments, the mass of the effective component in the ionomer solution can also be one of 20%, 30%, 40%, 50%, 60%, 70%, and 80% of the mass of C in the Pt-C catalyst.

[0048] In some embodiments, the mass of the effective component in the ionomer solution may also be one of 20~40%, 30~50%, 40~60%, 50~70%, 60~80%, 20~60%, 30~70%, and 40~80% of the mass of C in the Pt-C catalyst.

[0049] In some embodiments, the mass of the Pt-C catalyst may also be one of 5%, 10%, 15%, and 20% of the mass of the deionized water.

[0050] In some embodiments, the mass of the Pt-C catalyst may also be one of 5~10%, 8~15%, 10~20%, 5~15%, and 10~18% of the mass of the deionized water.

[0051] In some embodiments, the mass ratio of the deionized water to the high-dielectric small-molecule alcohol may also be one of 1:1, 1:2, 1:3, and 1:4.

[0052] In some embodiments, the mass ratio of the deionized water to the high-dielectric small-molecule alcohol may also be one of 1:(1~2), 1:(2~3), 1:(3~4), 1:(1~3), and 1:(2~4).

[0053] In some embodiments, the sulfonated lignin-doped conductive polymer nanoparticles may also be one of 0.5%, 1%, 2%, 3%, 4%, and 5% of the mass of the Pt-C catalyst.

[0054] In some embodiments, the sulfonated lignin-doped conductive polymer nanoparticles may also be one of 0.5~2%, 1~3%, 2~4%, 3~5%, 0.5~3%, 1~4%, and 2~5% of the mass of the Pt-C catalyst.

[0055] In some embodiments, the mass of the hydrophilic binder may also be one of 2%, 3%, 4%, and 5% of the mass of the deionized water.

[0056] In some embodiments, the mass of the hydrophilic binder may also be one of 2%~3%, 3%~4%, 4%~5%, 2%~4%, and 3%~5% of the mass of the deionized water.

[0057] In some embodiments, the catalyst of the cathode catalyst layer is at least one of the Pt-C catalyst and the Ni-Fe catalyst; when the catalyst of the cathode catalyst layer is the Pt-C catalyst, a loading amount of Pt is 0.3-0.7 mg/cm², mass of an effective component in the ionomer solution is 30-80% of mass of C in the Pt-C catalyst, mass of the Pt-C catalyst is 5-20% of mass of the deionized water, a mass ratio of the deionized water to the high-dielectric small-molecule alcohol is 1:(1-4), and mass of the polytetrafluoroethylene is 1-5% of the mass of the deionized water.

[0058] In some embodiments, when the catalyst of the cathode catalyst layer is the Pt-C catalyst, the loading amount of Pt may also be one of 0.3 mg/cm², 0.4 mg/cm², 0.5

mg/cm², 0.6 mg/cm², and 0.7 mg/cm².

[0059] In some embodiments, when the catalyst of the cathode catalyst layer is the Pt-C catalyst, the loading amount of Pt may also be one of 0.3~0.5 mg/cm², 0.4~0.6 mg/cm², 0.5~0.7 mg/cm², 0.3~0.6 mg/cm², and 0.4~0.7 mg/cm².

[0060] In some embodiments, the mass of the effective component in the ionomer solution may also be one of 30%, 40%, 50%, 60%, 70%, and 80% of the mass of C in the Pt-C catalyst.

[0061] In some embodiments, the mass of the effective component in the ionomer solution may also be one of 30%~50%, 40%~60%, 50%~70%, 60%~80%, 30%~60%, 40%~70%, and 50%~80% of the mass of C in the Pt-C catalyst.

[0062] In some embodiments, the mass of the Pt-C catalyst may also be one of 5%, 10%, 15%, and 20% of the mass of the deionized water.

[0063] In some embodiments, the mass of the Pt-C catalyst may also be one of 5%~10%, 8%~15%, 10%~20%, 5%~15%, and 10%~18% of the mass of the deionized water.

[0064] In some embodiments, the mass ratio of the deionized water to the high-dielectric small-molecule alcohol may also be one of 1:1, 1:2, 1:3, and 1:4.

[0065] In some embodiments, the mass ratio of the deionized water to the high-dielectric small-molecule alcohol may also be one of 1:(1~2), 1:(2~3), 1:(3~4), 1:(1~3), and 1:(2~4).

[0066] In some embodiments, the mass of the polytetrafluoroethylene may also be one of 1%, 2%, 3%, 4%, and 5% of the mass of the deionized water.

[0067] In some embodiments, the mass of the polytetrafluoroethylene may also be one of 1%~2%, 2%~3%, 3%~4%, 4%~5%, 1%~3%, 2%~4%, and 3%~5% of the mass of the deionized water.

[0068] In some embodiments, by using the sulfonated lignin-doped conductive polymer nanoparticles within the above mass range, a hydrophilic effect and conductive effect of the sulfonated lignin-doped conductive polymer nanoparticles in the anode catalyst layer are effectively improved, and the sulfonated lignin-doped conductive polymer nanoparticles exist more stably within the anode catalyst. If the used mass is not within this range, both the conductive performance and the hydrophilic performance of the sulfonated lignin-doped conductive polymer nanoparticles decrease.

[0069] In some embodiments, by using the polytetrafluoroethylene within the above mass range, a hydrophobic effect of the polytetrafluoroethylene in the cathode catalyst layer is effectively improved, and the polytetrafluoroethylene exists more stably within the cathode catalyst. If the used mass is not within this range, the hydrophobic performance

of the polytetrafluoroethylene decreases.

[0070] In some embodiments, the present disclosure provides a preparation process of the membrane electrode for fuel cells includes: a preparation of the anode-side adhesive layer: dispensing the conductive polymer-sulfonated lignin hydrogel onto a peripheral area of a surface of the anode catalyst layer, thereby forming the anode-side adhesive layer configured for bonding the anode catalyst layer; a preparation of the cathode-side adhesive layer: dispensing the ionomer solution onto a peripheral area of a surface of the cathode catalyst layer, thereby forming the cathode-side adhesive layer configured for bonding a cathode diffusion layer, wherein the ionomer solution is a Nafion-PEDOT conductive polymer, and the membrane electrode assembly employs a frame-like encapsulation.

[0071] In some embodiments, the membrane electrode assembly employs a frame-like encapsulation. The frame-like encapsulation employs an annular closed encapsulation structure at the edge of the membrane electrode, configured for realizing edge fixation and sealing of a multi-layer structure. A planar shape of the frame-like encapsulation is similar to the Chinese character "口" (i.e., a rectangular closed frame). More descriptions regarding the frame-like encapsulation may be found in the related descriptions concerning the PEN frame and the flat-plate hot pressing in S2 and S5 below.

[0072] S1, a preparation of the anode catalyst layer: mixing raw materials contained in the anode catalyst layer, preparing the mixing raw materials contained in the anode catalyst layer on one side of a proton exchange membrane; a preparation of the cathode catalyst layer: mixing raw materials contained in the cathode catalyst layer and preparing the mixed raw materials on the other side of the proton exchange membrane.

[0073] In some embodiments, in the S1, a mixing process includes at least one of an ultrasonic mixing, a high-speed shear mixing, a ball milling mixing, and a high-pressure homogenization mixing.

[0074] In some embodiments, types and parameters (e.g., mixing time, mixing power, etc.) of the mixing process may be determined based on actual needs.

[0075] In some embodiments, the mixing of the ingredients employs at least one of the ultrasonic mixing, the high-speed shear mixing, the ball milling mixing, and the high-pressure homogenization mixing. Using the above mixing process makes structures of both the anode catalyst layer and the cathode catalyst layer more stable, avoiding material precipitation.

[0076] In some embodiments, in S1, processes such as spraying, dip coating, or roll

coating may be used to blade-coat the mixed anode catalyst layer ingredients onto the other side of the proton exchange membrane, and blade-coat the mixed cathode catalyst layer ingredients onto the other side of the proton exchange membrane.

[0077] In some embodiments, in S1, mixed anode catalyst layer ingredients are applied to one side of the proton exchange membrane using screen printing, and the mixed cathode catalyst layer ingredients are blade-coated onto the other side of the proton exchange membrane.

[0078] The screen printing is a printing process. The screen printing utilizes a squeegee to squeeze the ingredients through a stencil, printing the ingredients onto the proton exchange membrane. Parameters for the screen printing may be determined based on actual needs.

[0079] In some embodiments, the mixed anode catalyst layer ingredients are applied to one side of the proton exchange membrane using the screen printing; and the mixed cathode catalyst layer ingredients are blade-coated onto the other side of the proton exchange membrane, this process enables precise control over the catalyst layer thickness, meeting the strict requirements for thinness and flatness. Furthermore, the screen printing process is simple, and the equipment cost is low, which is beneficial for low-cost large-scale production.

[0080] In some embodiments, in S1, thicknesses of the anode catalyst layer and the cathode catalyst layer may be determined based on actual needs.

[0081] In some embodiments, the thicknesses of the anode catalyst layer and the cathode catalyst layer are 5~10 μm , respectively.

[0082] In some embodiments, the thicknesses of the anode catalyst layer and the cathode catalyst layer may also be one of 5 μm , 6 μm , 7 μm , 8 μm , 9 μm , and 10 μm , respectively.

[0083] In some embodiments, the thicknesses of the anode catalyst layer and the cathode catalyst layer may also be one of 5~6 μm , 6~7 μm , 7~8 μm , 8~9 μm , and 9~10 μm , respectively.

[0084] In some embodiments, the thicknesses of the anode catalyst layer and the cathode catalyst layer are set to be 5~10 μm , respectively, ensuring that the anode catalyst layer and the cathode catalyst layer have good catalytic efficiency while avoiding waste of the raw materials.

[0085] S2, using a Polyethylene Naphthalate (PEN) frame with a hot-melt adhesive to seal the anode catalyst layer and the cathode catalyst layer, wherein edges of the anode

catalyst layer and the cathode catalyst layer are 0.5-1 cm away from the PEN frame; adhering the PEN frame by performing an initial flat-plate hot pressing;

[0086] The PEN frame is a frame structure configured for encapsulating the catalyst layers. In some embodiments, the PEN frame is primarily composed of a PEN substrate compounded with a hot-melt adhesive. The PEN frame is sealed around the edges of the anode catalyst layer and the cathode catalyst layer, the anode catalyst layer, the proton exchange membrane, and the cathode catalyst layer are encapsulated into a three-layer composite structure.

[0087] In some embodiments, a total thickness of the PEN frame may be determined based on actual needs.

[0088] In some embodiments, the total thickness of the PEN frame is 40~80 μm .

[0089] In some embodiments, the total thickness of the PEN frame may also be one of 40 μm , 45 μm , 50 μm , 55 μm , 60 μm , 70 μm , and 80 μm .

[0090] In some embodiments, the total thickness of the PEN frame may also be one of 40~50 μm , 50~60 μm , 60~70 μm , 70~80 μm , 40~60 μm , 50~70 μm , and 60~80 μm .

[0091] In some embodiments, by setting the total thickness of the PEN frame to 40~80 μm , the thickness range of the PEN frame can improve hydrophilic and water management performance while avoiding waste of the raw materials, saving materials, and reducing costs.

[0092] The flat-plate hot pressing is an encapsulation process for bonding the PEN frame. The flat-plate hot pressing may realize the frame-like encapsulation of the membrane electrode.

[0093] In some embodiments, the flat-plate hot pressing includes fitting the PEN frame around peripheries of the anode catalyst layer, the proton exchange membrane, and the cathode catalyst layer, then placing it into a hot press machine. Pressure and heat are applied to the PEN frame via platens, causing a hot-melt adhesive within the PEN frame to fully melt and infiltrate encapsulation interfaces. After the hot pressing is completed, the PEN frame is cooled to allow for the solidification and setting of the hot-melt adhesive, ultimately achieving a tight bond between the PEN frame and the anode catalyst layer, proton exchange membrane, and cathode catalyst layer.

[0094] In some embodiments, the flat-plate hot pressing may include an initial flat-plate hot pressing and a second flat-plate hot pressing. The initial flat-plate hot pressing encapsulates the anode catalyst layer, the proton exchange membrane, and the cathode catalyst layer into a three-layer composite structure. More descriptions regarding the

second flat-plate hot pressing may be found in S5 and related descriptions below.

[0095] In some embodiments, the initial flat-plate hot pressing is conducted with a pressure of less than 0.3 MPa, a temperature below 120°C, and a duration of 30 s~1 min.

[0096] In some embodiments, the initial flat-plate hot pressing is conducted with a pressure of one of 0.1 MPa, 0.15 MPa, 0.2 MPa, and 0.25 MPa.

[0097] In some embodiments, the initial flat-plate hot pressing is conducted with a pressure of one of 0.1~0.2 MPa, 0.2~0.3 MPa, and 0.1~0.25 MPa.

[0098] In some embodiments, the initial flat-plate hot pressing is conducted with a pressure of one of 80°C, 100°C, 110°C, and 115°C.

[0099] In some embodiments, the initial flat-plate hot pressing is conducted with a pressure of one of 80~100°C, 100~110°C, 110~120°C, 80~110°C, and 100~120°C.

[0100] In some embodiments, the duration of the initial flat-plate hot pressing may also be one of 30 s, 40 s, 50 s, and 60 s.

[0101] In some embodiments, the duration of the initial flat-plate hot pressing may also be one of 30~40 s, 40~50 s, 50~60 s, 30~50 s, and 40~60 s.

[0102] S3, a preparation of the anode-side adhesive layer: dispensing the conductive polymer-sulfonated lignin hydrogel onto the peripheral area of the surface of the anode catalyst layer, thereby forming the anode-side adhesive layer configured for bonding the anode catalyst layer; a preparation of the cathode-side adhesive layer: dispensing the ionomer solution onto the peripheral area of the surface of the cathode catalyst layer, thereby forming the cathode-side adhesive layer configured for bonding the cathode diffusion layer;

[0103] The anode-side adhesive layer is a structure for bonding the anode catalyst layer and the anode diffusion layer. The cathode-side adhesive layer is a structure for bonding the cathode catalyst layer and the cathode diffusion layer.

[0104] In some embodiments, the conductive polymer-sulfonated lignin hydrogel may be dispensed on an edge of the surface of the anode catalyst layer to form the anode-side adhesive layer for adhering the anode catalyst layer.

[0105] The conductive polymer-sulfonated lignin hydrogel is a hydrophilic conductive binder, which may be prepared from the sulfonated lignin-doped conductive polymer nanoparticles.

[0106] In some embodiments, in the conductive polymer-sulfonated lignin hydrogel, the ratio of conductive polymer to sulfonated lignin may be determined based on actual needs.

[0107] In some embodiments, in the conductive polymer-sulfonated lignin hydrogel, the

solid content of the conductive polymer is 2% to 10%, and the remaining solid is sulfonated lignin. For example, the solid ratio of conductive polymers to sulfonated lignin may be one of the following: 2% : 98%, 3% : 97%, 4% : 96%, 5% : 95%, 6% : 94%, 7% : 93%, 8% : 92%, 9% : 91%, 10% : 90%.

[0108] In some embodiments, sulfonated lignin may be added to the deionized water, and the ultrasonic mixing is performed to obtain the sulfonated lignin hydrogel. The conductive polymer nanoparticles are gradually added to the sulfonated lignin hydrogel, and the ultrasonic mixing is performed again to obtain the conductive polymer-sulfonated lignin hydrogel.

[0109] In some embodiments, the ionomer solution is dispensed onto the peripheral area of the surface of the cathode catalyst layer, thereby forming the cathode-side adhesive layer configured for bonding the cathode diffusion layer.

[0110] The Nafion-PEDOT conductive polymer is a relatively hydrophobic conductive binder. The Nafion-PEDOT conductive polymer includes a Nafion resin and poly(3,4-ethylenedioxythiophene) (PEDOT). The Nafion resin is a perfluorosulfonic acid-perfluorovinyl ether copolymer resin.

[0111] In some embodiments, the PEDOT has good conductivity and hydrophobicity. The Nafion resin contains abundant sulfonic acid groups and has good proton conductivity and hydrophilicity. The Nafion resin and the PEDOT polymer can synergistically achieve co-conduction of electrons and protons, adapting to the electrochemical reaction requirements of the fuel cell catalyst layer.

[0112] In some embodiments, the hydrophobicity and conductivity of the cathode-side adhesive layer may be adjusted by adjusting the ratio of Nafion to PEDOT. The ratio of Nafion to PEDOT may be determined based on actual needs.

[0113] The preparation method of the present disclosure uses hydrophilic and relatively hydrophobic conductive binders for dispensing on the anode and cathode, respectively, to fix the diffusion layers without sacrificing conductivity. Simultaneously, the frame-like encapsulation performs hydrophilic and hydrophobic management on the two sides, respectively, which can effectively improve the performance.

[0114] S4, laminating an anode diffusion layer onto a surface of the anode-side adhesive layer, and laminating the cathode diffusion layer onto a surface of the cathode-side adhesive layer.

[0115] The anode diffusion layer is a porous material layer used for fuel transport and electron conduction. The cathode diffusion layer is a porous material layer used for

oxidant transport and electron conduction. The diffusion layer is a porous material with good air permeability and good drainage characteristics. The diffusion layer may transport fuel and oxidant to the catalyst layer, providing sufficient reactants for the catalyst layer. Simultaneously, the diffusion layer discharges products from the catalyst layer, preventing product accumulation in the catalyst layer from hindering further reaction.

[0116] In some embodiments, each of the cathode diffusion layer and the anode diffusion layer includes nano-silica and a dual-layer composite structure of carbon fiber and graphite, and the nano-silica coats the dual-layer composite structure of carbon fiber and graphite.

[0117] The dual-layer composite structure of carbon fiber and graphite is a substrate of the diffusion layer. In some embodiments, graphite may be made into graphite ingredients, coated on a carbon fiber woven fabric, and dried and sintered to obtain the dual-layer composite structure of carbon fiber and graphite.

[0118] The nano-silica is an additive for modifying the diffusion layer. The nano-silica may enhance the mass transfer capability and hydrophilicity of the diffusion layer.

[0119] In some embodiments, the nano-silica adsorbs on an outer surface of the carbon fiber and the graphite layer through physical adsorption and chemical hydrogen bonding. Furthermore, the nano-silica penetrates and deposits in pores of the carbon fiber and interlayer gaps of the graphite layer through capillary action, such that the nano-silica coats the dual-layer composite structure of carbon fiber and graphite.

[0120] In some embodiments, parameters such as the ratio of the nano-silica and the thicknesses of the carbon fiber and the graphite may be determined based on actual needs.

[0121] In some embodiments, the mass of the nano-silica is 10-40% of mass of the dual-layer composite structure of carbon fiber and graphite, a thickness of the carbon fiber is 150-310 μm , and a thickness of the graphite is 20-40 μm .

[0122] In some embodiments, the mass of the nano-silica may also be one of 10%, 15%, 20%, 25%, 30%, 35%, and 40% of the mass of the dual-layer composite structure of carbon fiber and graphite.

[0123] In some embodiments, the mass of the nano-silica may also be one of 10~15%, 15~20%, 20~25%, 25~30%, 30~35%, 35~40%, 10~20%, 20~30%, 30~40%, 10~25%, and 25~40% of the mass of the dual-layer composite structure of carbon fiber and graphite.

[0124] In some embodiments, the thickness of the carbon fiber may also be one of 150

μm, 180 μm, 210 μm, 240 μm, 270 μm, 300 μm, and 310 μm.

[0125] In some embodiments, the thickness of the carbon fiber may also be one of 150~180 μm, 180~210 μm, 210~240 μm, 240~270 μm, 270~300 μm, 300~310 μm, 150~210 μm, 210~270 μm, 270~310 μm, 150~240 μm, and 240~310 μm.

[0126] In some embodiments, the thickness of the graphite may also be one of 20 μm, 25 μm, 30 μm, 35 μm, and 40 μm.

[0127] In some embodiments, the thickness of the graphite may also be one of 20~25 μm, 25~30 μm, 30~35 μm, 35~40 μm, 20~30 μm, 30~40 μm, 20~35 μm, and 25~40 μm.

[0128] In some embodiments, setting the mass of the nano-silica to 10~40% of the mass of the dual-layer composite structure of carbon fiber and graphite, the thickness of the carbon fiber to 150~310 μm, and the thickness of the graphite to 20~40 μm can ensure that the cathode diffusion layer and the anode diffusion layer have appropriate pore density and pore size, while ensuring the conductivity of the cathode diffusion layer and the anode diffusion layer.

[0129] In some embodiments, the nano-silica may be attached to a surface of the dual-layer composite structure of carbon fiber and graphite by soaking or spraying to form a film-like coating, i.e., the anode diffusion layer or the cathode diffusion layer.

[0130] In some embodiments, the anode diffusion layer or the cathode diffusion layer is formed by immersing the dual-layer composite structure of carbon fiber and graphite in a silica suspension, removing and drying the dual-layer composite structure of carbon fiber and graphite, and repeating the immersing and drying process multiple times.

[0131] In some embodiments, because a single immersing and drying process cannot cause a sufficient amount of the nano-silica to attach to the surface of the dual-layer composite structure of carbon fiber and graphite. Therefore, the immersing and drying process may be repeated multiple times until the attachment amount of the nano-silica meets the requirement. Immersing parameters (e.g., an immersion time), drying parameters (e.g., a drying temperature and a drying duration), and a number of cycles may be determined based on actual needs. The number of cycles refers to the number of process cycles in which the dual-layer composite structure of carbon fiber and graphite is sequentially subjected to immersing in the silica suspension and removal for drying.

[0132] In some embodiments, the immersion time in the silica suspension is 10 s, the drying temperature is 140°C, the drying duration is 10 min, and the number of cycles is 4~5 times.

[0133] In some embodiments, the immersion time in the silica suspension may also be

one of 5 s, 10 s, 15 s, and 20 s.

[0134] In some embodiments, the immersion time in the silica suspension may also be one of 5~10 s, 10~15 s, 15~20 s, 5~15 s, and 10~20 s.

[0135] In some embodiments, the drying temperature in the silica suspension may also be one of 120°C, 130°C, 140°C, 150°C, and 160°C.

[0136] In some embodiments, the drying temperature in the silica suspension may also be one of 120~140°C, 140~160°C, 120~130°C, 130~150°C, 150~160°C, and 120~160°C.

[0137] In some embodiments, the drying duration in the silica suspension may also be one of 5 min, 10 min, 15 min, and 20 min.

[0138] In some embodiments, the drying duration in the silica suspension may also be one of 5-10 min, 10-15 min, 15-20 min, 5-15 min, or 10-20 min.

[0139] In some embodiments, the number of cycles of repeating the immersing and drying process may also be one of 4 or 5 times.

[0140] In some embodiments, the number of cycles of repeating the immersing and drying process may also be one of 3-4 times, 4-5 times, 5-6 times, 3-5 times, or 4-6 times.

[0141] In some embodiments, the anode diffusion layer or the cathode diffusion layer is prepared by immersing the dual-layer composite structure of carbon fiber and graphite in the silica suspension, removing and drying the dual-layer composite structure of carbon fiber and graphite, and repeating the immersing and drying multiple times. The silica suspension penetrates into the pores of the carbon fiber-graphite composite structure through capillary action. After drying, a nano-silica film is formed, and by repeating the impregnation and drying multiple times, problems such as local agglomeration or uneven coating caused by a single impregnation can be avoided.

[0142] In some embodiments, by setting the immersion time in the silica suspension to 10 s, the drying temperature to 140°C, the drying duration to 10 min, and the number of cycles to 4-5 times, the loading amount of nano-silica can be precisely controlled, ensuring uniformity of the anode diffusion layer and the cathode diffusion layer across different batches.

[0143] In some embodiments, a preparation process of the silica suspension includes: obtaining a mixture by performing an initial sonication of silica, acetylacetone, and isopropanol; adjusting pH of the mixture to less than 1 by adding concentrated sulfuric acid; and obtaining the silica suspension by performing a second sonication of the mixture.

[0144] The acetylacetone is a polarity regulator for the silica suspension. In some embodiments, because the surface of the nano-silica has a large count of hydroxyl groups

and extremely high surface energy, agglomeration is prone to occur. The isopropanol is a weakly polar solvent, and the compatibility between the two is poor. Therefore, it is necessary to add the acetylacetone to appropriately reduce the surface energy of the nano-silica, promoting the formation of a stable colloidal system of nano-silicon dioxide in the isopropanol, thereby obtaining the silica suspension.

[0145] In some embodiments, the pH may also be adjusted using a strong acid such as the concentrated sulfuric acid. The hydroxyl groups on the surface of the nano-silica are protonated into positively charged silanulates, enhancing the repulsive force between the nano-silica through electrostatic repulsion, thereby further preventing agglomeration of the nano-silica.

[0146] In some embodiments, a sonication frequency of the initial sonication is 40Hz, a sonication temperature of the initial sonication is 10°C, a sonication duration of the initial sonication is 10min; and a sonication frequency of the second sonication is 40Hz, a sonication temperature of the second sonication is 5°C, a sonication duration of the second sonication is 30min.

[0147] In some embodiments, the sonication frequency of the initial sonication may also be one of 40 Hz, 30 Hz, or 50 Hz.

[0148] In some embodiments, the sonication frequency of the initial sonication may also be one of 30-40 Hz, 40-50 Hz, or 30-50 Hz.

[0149] In some embodiments, the sonication temperature of the initial sonication may also be one of 10°C, 5°C, or 15°C.

[0150] In some embodiments, the sonication temperature of the initial sonication may also be one of 5-10°C, 10-15°C, or 5-15°C.

[0151] In some embodiments, the sonication duration of the initial sonication may also be one of 10 min, 5 min, or 15 min.

[0152] In some embodiments, the sonication duration of the initial sonication may also be one of 5-10 min, 10-15 min, or 5-15 min.

[0153] In some embodiments, the sonication frequency of the second sonication may also be one of 40 Hz, 30 Hz, or 50 Hz.

[0154] In some embodiments, the sonication frequency of the second sonication may also be one of 30-40 Hz, 40-50 Hz, or 30-50 Hz.

[0155] In some embodiments, the sonication temperature of the second sonication may also be one of 5°C, 10°C, or 15°C.

[0156] In some embodiments, the sonication temperature of the second sonication may also be one of 5-10°C, 10-15°C, or 5-15°C.

[0157] In some embodiments, the sonication duration of the second sonication may also be one of 30 min, 20 min, or 40 min.

[0158] In some embodiments, the sonication duration of the second sonication may also be one of 20-30 min, 30-40 min, or 20-40 min.

[0159] In some embodiments, the sonication frequency of the initial sonication is 40Hz, the sonication temperature of the initial sonication is 10°C, the sonication duration of the initial sonication is 10min; the sonication frequency of the second sonication is 40Hz, the sonication temperature of the second sonication is 5°C, the sonication duration of the second sonication is 30min. The homogeneity of the silica suspension can be ensured, and agglomeration and coagulation of the nano-silica can be avoided.

[0160] In some embodiments, mass ratios of the silica, the acetylacetone, and the isopropanol in the silica suspension may be determined based on actual needs.

[0161] In some embodiments, the mass ratios of the silica, the acetylacetone, and the isopropanol in the silica suspension are 2%, 5%, and 93%, respectively; and a particle size of the silica in the silica suspension is 20nm.

[0162] In some embodiments, the mass ratios of the silica, the acetylacetone, and the isopropanol in the silica suspension may also be one of 2.5%:5%:92.5%, 1.5%:5.5%:93%, 2%:4.5%:93.5%, 3%:5%:92%, or 2%:5.5%:92.5%.

[0163] In some embodiments, the particle size of the silica in the silica suspension may also be one of 16 nm, 18 nm, 22 nm, 24 nm, or 26 nm.

[0164] In some embodiments, by setting the mass ratios of the silica, the acetylacetone, and the isopropanol in the silica suspension are 2%, 5%, and 93%, respectively; and the particle size of the silica in the silica suspension is 20 nm, the suspension stability of the silica suspension can be ensured, resulting in a stable and homogeneous silica suspension.

[0165] S5, processing using the second flat-plate hot pressing process to obtain the membrane electrode.

[0166] In some embodiments, after the bonding step of S4, a five-layer composite structure (the anode diffusion layer, the anode catalyst layer, the proton exchange membrane, the cathode catalyst layer, and the cathode diffusion layer) is obtained. To further enhance the bonding force between the anode diffusion layer and the cathode

diffusion layer, it is also necessary to further tightly bond the cathode diffusion layer and the anode diffusion layer through the second flat-plate hot pressing process to obtain the finished membrane electrode..

[0167] In some embodiments, the second flat-plate hot pressing is conducted with a pressure below 0.3 MPa, a temperature ranging from 130 to 140 °C, and a duration of 30s to 2 min.

[0168] In some embodiments, the second flat-plate hot pressing is conducted with the pressure may also be one of 0.1 MPa, 0.15 MPa, 0.2 MPa, or 0.25 MPa.

[0169] In some embodiments, the second flat-plate hot pressing is conducted with the pressure may also be one of 0.1 MPa, 0.15 MPa, 0.2 MPa, or 0.25 MPa. may also be one of 0.1-0.2 MPa, 0.2-0.3 MPa, or 0.1-0.25 MPa.

[0170] In some embodiments, the second flat-plate hot pressing is conducted with the temperature may also be one of 130°C, 132°C, 135°C, 138°C, or 140°C.

[0171] In some embodiments, the second flat-plate hot pressing is conducted with the temperature may also be one of 130-135°C, 135-140°C, 130-132°C, 132-138°C, or 138-140°C.

[0172] In some embodiments, the second flat-plate hot pressing is conducted with the duration may also be one of 30 s, 60 s, 90 s, or 120 s.

[0173] In some embodiments, the second flat-plate hot pressing is conducted with the duration may also be one of 30-60 s, 60-90 s, 90-120 s, 30-90 s, or 60-120 s.

[0174] The present disclosure achieves efficient water management by adding a hydrophilic conductive polymer (the sulfonated lignin-doped conductive polymer nanoparticles) to the anode catalyst ingredients and adding a hydrophobic material (the polytetrafluoroethylene) to the cathode catalyst ingredients. Specifically, the addition of the hydrophilic conductive polymer can absorb and retain moisture, reducing drying of the proton exchange membrane and ensuring sufficient moisture on the anode side; whereas the addition of the hydrophobic material polytetrafluoroethylene can effectively prevent flooding, ensuring rapid removal of moisture from the cathode side and maintaining good gas transport channels.

[0175] By optimizing the formulation of the catalyst ingredients, the present disclosure improves the electrical conductivity of the catalyst layer, thereby enhancing the electrochemical performance of the fuel cells. The hydrophilic conductive polymer and the nano-conductive particles can form a stable conductive network, improving the transport efficiency of electrons and protons; the addition of the high-dielectric small-

molecule alcohol can promote proton conduction, improve the ionic conductivity of the proton exchange membrane, and thereby enhance the overall performance of the fuel cells. By adding a hydrophilic binder (e.g., the glycerol, PEG) to the catalyst ingredients, the present disclosure ensures uniform dispersion of the catalyst particles in the raw materials, improving the uniformity and stability of the catalyst layer. The hydrophilic binder can reduce the interaction forces between catalyst particles, prevent particle agglomeration, ensure uniform distribution of the catalyst in the catalyst layer, thereby ensuring the thickness and uniformity of the catalyst layer and improving the performance of the catalyst layer.

[0176] In the preparation process, the present disclosure uses hydrophilic and relatively hydrophobic conductive adhesives for dispensing on the anode and cathode respectively, fixing the diffusion layers without sacrificing conductivity. Meanwhile, the frame-type encapsulation structure separately manages hydrophilicity and hydrophobicity on both sides, effectively enhancing performance.

[0177] The fuel cell catalyst layer membrane electrode and the preparation method of the membrane electrode provided in the present disclosure are further described below with reference to the accompanying drawings.

[0178] Embodiment 1:

[0179] Embodiment 1 provides a membrane electrode and a preparation method of the membrane electrode:

[0180] Ingredients are as follows:

[0181] The anode catalyst layer: 100 g of the deionized water, 100 g of the ethanol, 15 g of the Pt-C catalyst (the loading amount of Pt is 0.03 mg/cm²), 0.075 g of the sulfonated lignin-doped conductive polymer nanoparticles, 2 g of the glycerol, 40 g of the sulfonated poly(ether ether ketone) solution (containing 10 wt% sulfonated polyether ether ketone);

[0182] The cathode catalyst: 100 g of the deionized water, 100 g of the ethylene glycol, 15 g of the Pt-C catalyst (the loading amount of Pt is 0.3 mg/cm²), 1 g of the polytetrafluoroethylene, 31.5 g of the sulfonated polystyrene solution (containing 10 wt% sulfonated polystyrene).

[0183] Operations are as follows:

[0184] S1, the above ingredients of the anode catalyst layer and the cathode catalyst layer are mixed, respectively, and the mixing process is the ultrasonic mixing.

[0185] S2, the dispersed anode catalyst ingredients are blade-coated on one side of the proton exchange membrane using the screen printing; a dry film thickness of the prepared

membrane layer is 5 μm .

[0186] S3, the dispersed cathode catalyst ingredients are blade-coated on the other side of the proton exchange membrane using the screen printing; the dry film thickness of the prepared membrane layer is 5 μm .

[0187] S4, the sonication(40 Hz, 10°C, 10 min) is used to disperse 20 nm silica and the acetylacetone in the isopropanol, a mass ratio of the silica, the acetylacetone, and the isopropanol is 2%:5%:93%; the concentrated sulfuric acid is then added concentrated until pH is less than 1, and the sonication (40 Hz, 5°C) is continued for 30 min.

[0188] S5, the anode diffusion layer/the cathode diffusion layer (the dual-layer composite structure of carbon fiber and graphite, with the carbon fiber thickness of 150 μm and the graphite thickness of 20 μm) is immersed in the silica suspension for 10 seconds, then dried at 140°C for 10 minutes, the immersion process is repeated for 4 to 5 times until the silica content on the diffusion layer reaches 10%.

[0189] S6, the anode catalyst layer and the cathode catalyst layer are both sealed with a PEN frame with the hot-melt adhesive, edges of the anode catalyst layer and the cathode catalyst layer are respectively 0.5 cm away from the PEN frame; the initial flat-plate hot pressing (pressure: 0.2 MPa, temperature :100°C, time :0.5 min) is used to bond the PEN frame.

[0190] S7, bonding the anode diffusion layer: the conductive polymer-sulfonated lignin hydrogel is dispensed onto the peripheral area of the anode catalyst layer surface to form the anode diffusion layer, wherein the anode diffusion layer adheres to the edge of the anode catalyst layer; bonding the cathode diffusion layer: the Nafion-PEDOT conductive polymer is dispensed onto the peripheral area of the cathode catalyst layer surface to form the cathode diffusion layer, wherein the cathode diffusion layer adheres to the edge of the cathode catalyst layer.

[0191] S8, processing using the second flat-plate hot pressing process (with a pressure of 0.2 MPa, temperature of 130°C, and duration of 0.5 min) to obtain the catalyst layer for the fuel cells.

[0192] Embodiment 2:

[0193] Embodiments 2 provides a membrane electrode and a preparation method of the membrane electrode:

[0194] Ingredients are as follows:

[0195] The anode catalyst layer: 100 g of the deionized water, 400 g of the ethanol, 20 g of the Pt-C catalyst (the loading amount of Pt 0.3 mg/cm²), 1 g of the sulfonated lignin-

doped conductive polymer nanoparticles, 5 g of the PEG, and 56 g of the sulfonated polysulfone solution (containing 20 wt% sulfonated polysulfone).

[0196] The cathode catalyst: 100 g of the deionized water, 400 g of the ethylene glycol, 20 g of the Pt-C catalyst (the loading amount of Pt was 0.7 mg/cm²), 5 g of the polytetrafluoroethylene, and 48 g of the sulfonated polyethylene solution (containing 10 wt% sulfonated polyethylene).

[0197] operations are as follows:

[0198] S1, the above ingredients of the anode catalyst layer and the cathode catalyst layer are mixed, respectively, and the mixing process is the high-speed shear mixing.

[0199] S2, the dispersed anode catalyst ingredients are prepared on one side of the proton exchange membrane by a spraying process; a dry film thickness of the prepared membrane layer is 10 μm.

[0200] S3, the dispersed cathode catalyst ingredients are blade-coated on the other side of the proton exchange membrane by one of a wire rod blade-coating process, a blade-coating process, or a slot-die blade-coating process; a dry film thickness of the prepared membrane layer is 10 μm.

[0201] S4, the sonication(40 Hz, 10°C, 10 min) is used to disperse 20 nm silica and the acetylacetone in the isopropanol, the mass ratio of the silica, the acetylacetone, and the isopropanol is 2%:5%:93%; the concentrated sulfuric acid is then added concentrated until pH is less than 1, and the sonication (40 Hz, 5°C) is continued for 30 min.

[0202] S5, the anode diffusion layer/the cathode diffusion layer (the dual-layer composite structure of carbon fiber and graphite, with the carbon fiber thickness of 150 μm and the graphite thickness of 40 μm) is immersed in the silica suspension for 10 seconds, then dried at 140°C for 10 minutes, and the immersion process is repeated 4 to 5 times until the silica content on the diffusion layer reaches 40%.

[0203] S6, the anode catalyst layer and the cathode catalyst layer are both sealed with a PEN frame with the hot-melt adhesive, edges of the anode catalyst layer and the cathode catalyst layer are respectively 1 cm away from the PEN frame; the initial flat-plate hot pressing (pressure: 0.28 MPa, temperature:118°C, time :1 min) is used to bond the PEN frame.

[0204] S7, bonding the anode diffusion layer: a conductive polymer-sulfonated lignin hydrogel is dispensed onto a surface edge of the anode catalyst layer to bond the anode diffusion layer; bonding the cathode diffusion layer: a Nafion-PEDOT conductive polymer is dispensed onto a surface edge of the cathode catalyst layer to bond the cathode

diffusion layer.

[0205] S8. Processing using the second flat-plate hot pressing process (with a pressure of 0.28 MPa, temperature of 140°C, and duration of 2 min) to obtain the catalyst layer for the fuel cell.

[0206] Embodiment 3

[0207] Embodiments 3 provides a membrane electrode and a preparation method of the membrane electrode.

[0208] Ingredients are as follows:

[0209] The anode catalyst layer: 100 g of the deionized water, 200 g of the ethanol, 18 g of the Pt-C catalyst (the loading amount of Pt 0.2 mg/cm²), 0.36 g of the sulfonated lignin-doped conductive polymer nanoparticles, 4 g of a mixture of the PEG and the glycerol, and 43.2 g of a mixture of the sulfonated polysulfone and the sulfonated polystyrene (containing 10 wt% sulfonated polysulfone and 10 wt% sulfonated polystyrene);

[0210] The cathode catalyst: 100 g of the deionized water, 300 g of the ethylene glycol, 16 g of the Pt-C catalyst (the loading amount of Pt is 0.5 mg/cm²), 4 g of the polytetrafluoroethylene, 28 g of a mixed solution of the sulfonated polyethylene and the sulfonated polyether ether ketone (containing 10 wt% sulfonated polyethylene and 10 wt% sulfonated polyether ether ketone).

[0211] Operations are as follows:

[0212] S1, the above ingredients of the anode catalyst layer and the cathode catalyst layer are mixed, respectively, and the mixing process is the ball milling mixing.

[0213] S2, the dispersed anode catalyst ingredients is blade-coated on one side of the proton exchange membrane by a transfer printing process; a dry film thickness of the prepared membrane layer is 8 μm.

[0214] S3, the dispersed cathode catalyst ingredients are blade-coated on the other side of the proton exchange membrane by a transfer printing process; a dry film thickness of the prepared membrane layer is 8 μm.

[0215] S4, the sonication(40 Hz, 10°C, 10 min) is used to disperse 20 nm silica and the acetylacetone in the isopropanol, the mass ratio of the silica, the acetylacetone, and the isopropanol is 2%:5%:93%; the concentrated sulfuric acid is then added concentrated until pH is less than 1, and the sonication (40 Hz, 5°C) is continued for 30 min.

[0216] S5, the anode diffusion layer/the cathode diffusion layer (the dual-layer composite structure of carbon fiber and graphite, with the carbon fiber thickness of 150 μm and the

graphite thickness of 220 μm) is immersed in the silica suspension for 10 seconds, then dried at 140°C for 10 minutes, and the immersion process is repeated for 4 to 5 times until the silica content on the diffusion layer reaches 30%.

[0217] S6, the anode catalyst layer and the cathode catalyst layer are both sealed with a PEN frame with the hot-melt adhesive, the edges of the anode catalyst layer and the cathode catalyst layer are respectively 1 cm away from the PEN frame; the initial flat-plate hot pressing (pressure: 0.25 MPa, temperature: 110°C, time: 1.5 min) is used to bond the PEN frame.

[0218] S7, bonding the anode diffusion layer: the conductive polymer-sulfonated lignin hydrogel is dispensed onto a surface edge of the anode catalyst layer to form the anode-side adhesive layer, and the anode diffusion layer adheres to the edge of the anode catalyst layer; bonding the cathode diffusion layer: the Nafion-PEDOT conductive polymer is dispensed onto a surface edge of the cathode catalyst layer to form a cathode-side adhesive layer, and the cathode diffusion layer adheres to the edge of the cathode catalyst layer.

[0219] S8, processing using the second flat-plate hot pressing process (with a pressure of 0.25 MPa, temperature of 135°C, and duration of 1.5 min) to obtain the catalyst layer for the fuel cell.

[0220] COMPARATIVE Embodiment 1

[0221] Comparative Embodiment 1 provides a membrane electrode of a catalyst layer for fuel cells and a preparation method of the membrane electrode. The preparation method of the membrane electrode is similar to that of Embodiment 2, with the only difference being that sulfonated lignin is removed from the raw materials of the anode catalyst layer in Comparative Embodiment 1, and a conductive polymer hydrogel not containing sulfonated lignin is used when attaching the anode diffusion layer in S7 of Comparative Embodiment 1.

[0222] COMPARATIVE Embodiment 2

[0223] Comparative Embodiment 2 provides a membrane electrode of a catalyst layer for fuel cells and a preparation method of the membrane electrode. The preparation method of the membrane electrode is similar to that of Embodiment 2, with the only difference being that polytetrafluoroethylene is removed from the raw materials of the cathode catalyst in Comparative Embodiment 2, and the rest is the same as in Embodiment 2.

[0224] TEST DETECTION

[0225] The catalyst layers prepared in Embodiments 1-3 and Comparative Embodiments 1-2 were cut into 5 cm×5 cm single pieces, sealed by hot melt sealing, matched with a 180 μm diffusion layer, and used to prepare 30 membrane electrodes. The membrane electrodes were assembled into a stack, activated, and subjected to performance testing.

[0226] Activation conditions: hydrogen 0.1 MPa, air 0.1 MPa, flow rates of 700 mL/min and 2800 mL/min, 60% humidification, 50°C, 4 A, constant current discharge at every 2 A for 30 min, 10 A, constant current discharge at every 5 A for 30 min, 19 A, constant current discharge at every 2 A for 30 min.

[0227] Constant current discharge conditions: hydrogen 0.1 MPa, air 0.1 MPa, flow rates of 700 mL/min and 2800 mL/min, 60% humidification, 50°C, constant current discharge at 15 A for 110 h. The stack power and temperature were continuously recorded.

[0228] FIG. 1 is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to some embodiments of the present disclosure. For more intuitive understanding and illustration, FIG. 1 is split into FIG. 1a, FIG. 1b, FIG. 1c, and FIG. 1d.

[0229] FIG. 1a is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to Embodiment 1 of the present disclosure. FIG. 1b is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to Embodiment 3 of the present disclosure. FIG. 1c is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to Embodiment 2 and Comparative Embodiment 1 of the present disclosure. FIG. 1d is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to Embodiment 2 and Comparative Embodiment 2 of the present disclosure. In FIG. 1, FIG. 1a, FIG. 1b, FIG. 1c, and FIG. 1d, the horizontal coordinate represents time, and the vertical coordinate represents the temperature of the fuel cells. A lower temperature of the fuel cell indicates that the fuel cell converts more chemical energy directly into electrical energy instead of wasting it as thermal energy, resulting in a higher energy conversion efficiency.

[0230] According to the results shown in FIG. 1, the temperatures of the fuel cells prepared using the technical solutions of Embodiments 1-3 are significantly lower than the temperatures of the fuel cells prepared in Comparative Embodiments 1-2. The temperatures of the fuel cells prepared using the technical solutions of Comparative Embodiments 1-2 are higher and exhibit severe fluctuations. It can be seen that the catalyst layer and the membrane electrode of the fuel cells prepared in the present

disclosure have a good effect in reducing the temperature of the fuel cells, which can effectively suppress sudden temperature changes and abnormal fluctuations, and ensure thermal stability during long-term operation of the fuel cells.

[0231] FIG. 2 is a schematic diagram illustrating a variation of a temperature of a fuel cell with time according to some embodiments of the present disclosure. For more intuitive understanding and illustration, FIG. 2 is split into FIG. 2a, FIG. 2b, FIG. 2c, and FIG. 2d.

[0232] FIG. 2a is a schematic diagram illustrating a variation of a power of a fuel cell with time according to Embodiment 1 of the present disclosure. FIG. 2b is a schematic diagram illustrating a variation of a power of a fuel cell with time according to Embodiment 3 of the present disclosure. FIG. 2c is a schematic diagram illustrating a variation of a power of a fuel cell with time according to Embodiment 2 and Comparative Embodiment 1 of the present disclosure. FIG. 2d is a schematic diagram illustrating a variation of a power of a fuel cell with time according to Embodiment 2 and Comparative Embodiment 2 of the present disclosure. In FIG. 2, FIG. 2a, FIG. 2b, FIG. 2c, and FIG. 2d, the horizontal coordinate represents time, and the vertical coordinate represents the power of the fuel cell.

[0233] According to the results shown in FIG. 2, the power of the fuel cells prepared using the technical solutions of Embodiments 1-3 is significantly higher than the power of the fuel cells prepared in Comparative Embodiments 1-2. The power of the fuel cells prepared using the technical solutions of Comparative Embodiments 1-2 is lower and exhibits severe fluctuations. It can be seen that the catalyst layer and the membrane electrode of the fuel cells prepared in the present application have a good effect in improving the power of the fuel cells, can effectively suppress sudden power changes and abnormal fluctuations, and ensure power stability during long-term operation of the fuel cells.

[0234] In the description of the present application, it should be understood that the terms "upper," "lower," "bottom," "top," "front," "rear," "inner," "outer," "left," "right," etc., indicate orientations or positional relationships based on the orientations or positional relationships shown in the accompanying drawings. These terms are used only for ease of description of the present application and simplification of the description, and do not indicate or imply that the referred apparatus or element must have a specific orientation or be constructed and operated in a specific orientation. Therefore, these terms should not be construed as limiting the present application.

[0235] Although the present application is described herein with reference to specific embodiments, it should be understood that these embodiments are merely examples of the principles and applications of the present application. Therefore, it should be understood that numerous modifications may be made to the exemplary embodiments, and other arrangements may be devised, without departing from the spirit and scope of the present application as defined by the appended claims. It should be understood that different dependent claims and features described herein may be combined in a manner different from that described in the original claims. It should also be understood that features described in connection with individual embodiments may be used in other embodiments.

WHAT IS CLAIMED IS:

1. A membrane electrode with a catalyst layer for fuel cells, wherein, the catalyst layer includes an anode catalyst and a cathode catalyst, and the anode catalyst layer includes a Pt-C catalyst, a high-dielectric small-molecule alcohol, sulfonated lignin-doped conductive polymer nanoparticles, a hydrophilic binder, an ionomer solution, and deionized water;

the cathode catalyst layer includes a catalyst, the ionomer solution, the high-dielectric small-molecule alcohol, polytetrafluoroethylene, the deionized water;

in the anode catalyst layer, within the Pt-C catalyst, a loading amount of Pt is 0.03-0.3 mg/cm², mass of an effective component in the ionomer solution is 20-80% of mass of C in the Pt-C catalyst, mass of the Pt-C catalyst is 5-20% of mass of the deionized water, a mass ratio of the deionized water to the high-dielectric small-molecule alcohol is 1:(1-4), mass of the sulfonated lignin-doped conductive polymer nanoparticles is 0.5-5% of the mass of the Pt-C catalyst, and mass of the hydrophilic binder is 2-5% of the mass of the deionized water;

in the cathode catalyst layer, the catalyst of the cathode catalyst layer is at least one of a Pt-C catalyst and a Ni-Fe catalyst; in the Pt-C catalyst, a loading amount of Pt is 0.3-0.7 mg/cm², mass of an effective component in the ionomer solution is 30-80% of mass of C in the Pt-C catalyst, mass of the Pt-C catalyst is 5-20% of mass of the deionized water, a mass ratio of the deionized water to the high-dielectric small-molecule alcohol is 1:(1-4), and mass of the polytetrafluoroethylene is 1-5% of the mass of the deionized water;

a preparation process of the membrane electrode with the catalyst layer for the fuel cells includes:

a preparation of an anode-side adhesive layer: dispensing a conductive polymer-sulfonated lignin hydrogel onto a peripheral area of a surface of the anode catalyst layer, thereby forming the anode-side adhesive layer configured for bonding the anode catalyst layer;

a preparation of the cathode-side adhesive layer: dispensing the ionomer solution onto a peripheral area of a surface of the cathode catalyst layer, thereby forming the cathode-side adhesive layer configured for bonding a cathode diffusion layer, wherein the ionomer solution is a Nafion-PEDOT conductive polymer, and the membrane electrode with the catalyst layer for the fuel cells assembly employs a frame-like

encapsulation;

wherein the high-dielectric small-molecule alcohol includes at least one of ethanol, ethylene glycol, n-propanol, and isopropanol;

the ionomer solution includes at least one of a sulfonated poly(ether ether ketone) solution, a sulfonated polystyrene solution, a sulfonated polysulfone solution, a sulfonated polyethylene solution, and a perfluorosulfonic acid resin solution; and

the hydrophilic binder includes glycerol and polyethylene glycol (PEG).

2. A preparation method of the membrane electrode with the catalyst layer for the fuel cells according to claim 1, comprising:

S1, a preparation of the anode catalyst layer: mixing raw materials contained in the anode catalyst layer and preparing the mixed raw materials contained in the anode catalyst layer on one side of a proton exchange membrane;

a preparation of the cathode catalyst layer: mixing raw materials contained in the cathode catalyst layer and preparing the mixed raw materials contained in the cathode catalyst layer on the other side of the proton exchange membrane;

S2, using a PEN frame with a hot-melt adhesive to seal the anode catalyst layer and the cathode catalyst layer, wherein edges of the anode catalyst layer and the cathode catalyst layer are 0.5-1 cm away from the PEN frame; adhering the PEN frame by performing an initial flat-plate hot pressing;

S3, a preparation of the anode-side adhesive layer: dispensing the conductive polymer-sulfonated lignin hydrogel onto the peripheral area of the surface of the anode catalyst layer, thereby forming the anode-side adhesive layer configured for bonding the anode catalyst layer;

a preparation of the cathode-side adhesive layer: dispensing the ionomer solution onto the peripheral area of the surface of the cathode catalyst layer, thereby forming the cathode-side adhesive layer configured for bonding the cathode diffusion layer;

S4, laminating an anode diffusion layer onto a surface of the anode-side adhesive layer, and laminating the cathode diffusion layer onto a surface of the cathode-side adhesive layer;

S5, processing using a second flat-plate hot pressing process to obtain the membrane electrode with the catalyst layer for the fuel cells;

wherein each of the cathode diffusion layer and the anode diffusion layer includes nano-silica and a dual-layer composite structure of carbon fiber and graphite, and the

nano-silica coats the dual-layer composite structure of carbon fiber and graphite.

3. The preparation method of the membrane electrode with the catalyst layer for the fuel cells according to claim 2, wherein mass of the nano-silica is 10-40% of mass of the dual-layer composite structure of carbon fiber and graphite, a thickness of the carbon fiber is 150-310 μm , and a thickness of the graphite is 20-40 μm .

4. The preparation method of the membrane electrode with the catalyst layer for the fuel cells according to claim 2, wherein a total thickness of the PEN frame is 40-80 μm .

5. The preparation method of the membrane electrode with the catalyst layer for the fuel cells according to claim 2, wherein

the initial flat-plate hot pressing is conducted with a pressure of less than 0.3 MPa, a temperature below 120 $^{\circ}\text{C}$, and a duration of 30s to 1 min; and

the second flat-plate hot pressing is conducted with a pressure below 0.3 MPa, a temperature ranging from 130 to 140 $^{\circ}\text{C}$, and a duration of 30s to 2 min.

6. The preparation method of the membrane electrode with the catalyst layer for the fuel cells according to claim 2, wherein thicknesses of the anode catalyst layer and the cathode catalyst layer are 5-10 μm , respectively.

7. The preparation method of the membrane electrode with the catalyst layer for the fuel cells according to claim 1, wherein in the S1, a mixing process includes at least one of an ultrasonic mixing, a high-speed shear mixing, a ball milling mixing, and a high-pressure homogenization mixing.

8. The preparation method of the membrane electrode with the catalyst layer for the fuel cells according to claim 1, wherein in the S1, the method further includes:

applying mixed anode catalyst layer ingredients to one side of the proton exchange membrane using screen printing; and

blade-coating the mixed cathode catalyst layer ingredients onto the other side of the proton exchange membrane.

9. The preparation method of the membrane electrode with the catalyst layer for the

fuel cells according to claim 2, wherein the method further includes:

immersing the dual-layer composite structure of carbon fiber and graphite in a silica suspension, removing and drying the dual-layer composite structure of carbon fiber and graphite, and repeating the immersing and drying process multiple times to form the anode diffusion layer or the cathode diffusion layer.

10. The preparation method of the membrane electrode with the catalyst layer for the fuel cells according to claim 9, wherein an immersion time in the silica suspension is 10s, a drying temperature is 140 °C, a drying duration is 10min, and a number of cycles is 4-5.

11. The preparation method of the membrane electrode with the catalyst layer for the fuel cells according to claim 2, wherein a preparation process of the silica suspension includes:

obtaining a mixture by performing an initial sonication of silica, acetylacetone, and isopropanol;

adjusting pH of the mixture to less than 1 by adding concentrated sulfuric acid; and
obtaining the silica suspension by performing a second sonication of the mixture.

12. The preparation method of the membrane electrode with the catalyst layer for the fuel cells according to claim 11, wherein

a sonication frequency of the initial sonication is 40Hz, a sonication temperature of the initial sonication is 10°C, a sonication duration of the initial sonication is 10min; and

a sonication frequency of the second sonication is 40Hz, a sonication temperature of the second sonication is 5°C, a sonication duration of the second sonication is 30min.

13. The preparation method of the membrane electrode with the catalyst layer for the fuel cells according to claim 11, wherein

mass ratios of the silica, the acetylacetone, and the isopropanol in the silica suspension are 2%, 5%, and 93%, respectively; and

a particle size of the silica in the silica suspension is 20nm.