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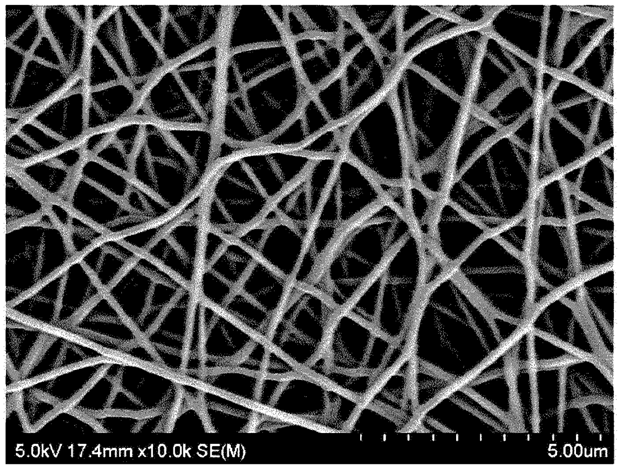
(54) GAS DIFFUSION LAYER FOR ANION EXCHANGE MEMBRANE ELECTROLYSIS, AND MANUFACTURING METHOD THEREFOR

(57) The present invention relates to a gas diffusion layer for anion exchange membrane electrolysis which is hydrophilic and exhibits gas permeability, and a manufacturing method therefor.

【FIG. 1】



EP 4 442 864 A1

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Description

[TECHNICAL FIELD]

CROSS-REFERENCE TO RELATED APPLICATION(S)

[0001] This application claims the benefit of Korean Patent Application No. 10-2021-0169324 filed on November 30, 2021 in the Korean Intellectual Property Office, the disclosure of which is incorporated herein by reference in its entirety.

[0002] The present invention relates to a gas diffusion layer for anion exchange membrane electrolysis and a manufacturing method therefor. Specifically, the present invention relates to a gas diffusion layer containing a reticular carbon nanofiber film, and a manufacturing method therefor.

[BACKGROUND]

[0003] The gas diffusion layer(GDL) serves as a passage that transmits reactants from a fuel cell(FC) and electrolysis(EC) to an electrode and simultaneously discharges products, and also is a key component that plays roles such as heat dissipation and electrode support.

[0004] Most of commercial GDLs are for polymer electrolyte membrane fuel cells(PEMFC) and have a structure in which a microporous layer(MPL) containing a hydrophobic polymer is coated onto a porous carbon paper. At this time, in the case of a fuel cell, a phenomenon (water flooding) may occur where water generated at the cathode blocks the pores of the gas diffusion layer, and thus the gas diffusion layer is required to have hydrophobicity. Therefore, the gas diffusion layer of the fuel cell uses PTFE and a binder as a polymer to impart hydrophobicity.

[0005] The gas diffusion layer for water electrolysis is an interface where gases such as hydrogen(H_2), oxygen(O_2), and water(H_2O), which are reactants and products of electrochemical reactions, moves freely between the catalyst layer and the electrode, and causes electrical conduction. Therefore, the porous carbon material is used, such as carbon fiber that have no side reactions with salts for electrochemical reactions, and have porosity and electrical conductivity.

[0006] However, a conventionally used carbon fiber paper had a problem of an interface contact resistance between the electrode layer and the catalyst layer due to its rough surface. To solve this problem, a microporous layer was formed by applying polymer materials such as powdered carbon paste or binder. However, the microporous layer containing polymer materials such as carbon paste or binder has a problem that its hydrophilicity and electrical conductivity are lowered due to the polymer component, which deteriorates the characteristics of the gas diffusion layer for electrolysis.

[0007] Therefore, a method has been devised to impart hydrophilicity by controlling the binder polymer contained in the microporous layer, but there still remains a problem that electrical conductivity is lowered, and an additional problem that gas movement is hindered.

[0008] Therefore, there is a need to develop a gas diffusion layer that is hydrophilic and has excellent electrical conductivity that can be used in anion exchange membrane electrolysis, and a method for manufacturing the same.

[DETAILED DESCRIPTION OF THE INVENTION]

[Technical Problem]

[0009] It is an object of the present invention to provide a gas diffusion layer for anion exchange membrane electrolysis and a manufacturing method therefor.

[0010] Specifically, it is an object of the present invention to provide a gas diffusion layer containing a reticular carbon nanofiber film which is hydrophilic and exhibits gas permeability, and a manufacturing method therefor.

[Technical Solution]

[0011] According to one embodiment of the present invention, there is provided a gas diffusion layer for anion exchange membrane electrolysis comprising a reticular carbon nanofiber film, wherein the carbon nanofiber has a diameter of 500 nm or less.

[0012] According to another embodiment of the present invention, there is provided a method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis, the method comprising: spinning a carbon precursor material to form a reticular molded product (step 1); and heat treating the reticular molded product to obtain a reticular carbon nanofiber film (step 2).

[Advantageous Effects]

[0013] A gas diffusion layer comprising a reticular carbon nanofiber film according to the present invention has higher porosity than polymers such as powdered carbon materials or binders, and thus is very advantageous for the movement of hydrogen gas and oxygen gas generated in the catalyst layer, which makes it easy to move gas from the electrolyte to the outside, thereby providing a gas diffusion layer for anion exchange membrane electrolysis that can improve the efficiency of an electrolytic cell.

[0014] In addition, the reticular carbon nanofiber film according to the present invention has high hydrophilicity and thus, can improve the wettability of the electrolyte in the gas diffusion layer.

[0015] Further, according to the present invention, it is possible to provide a method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis using a spinning method.

[0016] Further, according to the present invention, it is possible to provide a method of using and manufacturing the reticular carbon nanofiber film as the gas diffusion layer itself.

[0017] Further, according to the present invention, it is possible to provide a method of applying a conventionally used gas diffusion layer together in addition to the reticular carbon nanofiber film.

[BRIEF DESCRIPTION OF THE DRAWINGS]

[0018]

FIG. 1 is a SEM image of the surface structure of the gas diffusion layer of an embodiment of the present invention.

FIG. 2 is an SEM image of the lateral structure of the gas diffusion layer of an embodiment of the present invention.

FIG. 3 shows an experiment photograph which compares the contact angle of the gas diffusion layer of an embodiment of the present invention with the contact angle of the existing gas diffusion layer.

FIG. 4 shows a graph that measures and compares the gas permeability of the gas diffusion layer of an embodiment of the present invention and the gas permeability of the existing gas diffusion layer.

[DETAILED DESCRIPTION OF THE EMBODIMENTS]

[0019] The terms "first", "second", etc. may be used herein to describe various elements, and these terms are used to distinguish one element from another.

[0020] Also, the terminology used herein is for the purpose of describing particular example embodiments only and is not intended to be limiting of the invention. As used herein, the singular forms "a," "an" and "the" are intended to include the plural forms as well, unless the context clearly indicates otherwise. It will be further understood that the terms "include," "comprise," or "have" when used herein, specify the presence of stated features, numbers, steps, elements, or combinations thereof, but do not preclude the presence or addition of one or more other features, numbers, steps, elements, or combinations thereof.

[0021] Also, it means that, when each layer or element is referred to as being formed "on" or "top" of the respective layers or elements, each layer or element is formed on the top of the respective layers or elements, or the other layer or element may be additionally formed between the respective layers, or on the object or the substrate.

[0022] While the present invention is susceptible to various modifications and alternative forms, specific embodiments will be illustrated and described in detail as follows. It should be understood, however, that the description is not intended to limit the present invention to the particular forms disclosed herein, but the intention is to cover all modifications, equivalents, and replacements falling within the spirit and scope of the invention.

[0023] In the anion exchange membrane electrolysis, as the hydrophilicity of the gas diffusion layer increases, the wettability and permeability of the electrolytic solution increase, and thus the efficiency of the cell can increase. Therefore, the existing gas diffusion layer for fuel cells having a hydrophobic microporous layer reduces the wettability of the electrolyte solution, and at the same time, the thickness of the microporous layer, which reaches several tens of micrometers (μm), can also be a cause of decreasing the porosity of the porous support of the gas diffusion layer.

[0024] Therefore, the present inventors have now devised a method of using a reticular carbon nanofiber film as a gas diffusion layer without using a conventionally used polymer material such as a binder (binder-free) that imparts hydrophobicity.

[0025] According to the present invention, if a polymer for forming a paste with a reticular carbon nanofiber film material is not used, it is expected to reduce the interface roughness and interface resistance between the electrode and the catalyst. In addition, the porosity of carbon nanofibers is higher than that of polymers such as powdered carbon materials or binders, and thus it is very advantageous for the movement of hydrogen gas and oxygen gas generated in the catalyst layer, which makes it easy to move gas from the electrolyte to the outside. Thereby, it is expected that the efficiency of an electrolytic cell can be improved.

[0026] Below, the gas diffusion layer for anion exchange membrane electrolysis of the present invention will be discussed in more detail.

[0027] The gas diffusion layer for anion exchange membrane electrolysis of the present invention may be a reticular carbon nanofiber film itself. Further, the gas diffusion layer for anion exchange membrane electrolysis of the present invention includes a porous support; and a reticular carbon nanofiber film.

[0028] The gas diffusion layer of one embodiment of the present invention includes a reticular carbon nanofiber film, wherein the carbon nanofiber may have a diameter of 500 nm or less. Specifically, the gas diffusion layer may be a reticular carbon nanofiber film.

[0029] Specifically, the carbon nanofiber may have a diameter of 50 nm or more, 100 nm or more, or 150 nm or more, and 500 nm or less, 400 nm or less, 300 nm or less, or 200 nm or less.

[0030] The gas diffusion layer may have a thickness of 50 to 500 μm. Specifically, the gas diffusion layer may have a thickness of 50 μm or more, 100 μm or more, 150 μm or more, or 200 μm or more, and 500 μm or less, 450 μm or less, 400 μm or less, 350 μm or less, 300 μm or less, or 250 μm or less.

[0031] The gas diffusion layer for anion exchange membrane electrolysis may have a contact angle with water of 20° or less. Since the gas diffusion layer of the present invention exhibits hydrophilicity due to the carbon nanofiber, it is preferable that the lower limit of the water contact angle is not limited. Specifically, the water contact angle may be 0°. By exhibiting superhydrophilicity that the contact angle is 20° or less, it is possible to ensure wettability of an electrolytic solution when used as a gas diffusion layer for water electrolysis. This has the advantage that the electrolysis reaction of water is easy because the movement and internal circulation of the electrolyte are advantageous compared to the gas diffusion layer to which a hydrophobic microporous layer is applied.

[0032] In the present invention, the water contact angle refers to an angle made when a liquid and a gas is in thermodynamic equilibrium on a solid surface. The water contact angle of the present invention can be measured by placing a film-like sample on a flat surface, dropping 5 to 10 μm of distilled water onto it, and then observing its shape from the side using DSA to calculate the angle. According to FIG. 4, when compared with the existing gas diffusion layer, it can be confirmed that the gas diffusion layer of the present invention has a water contact angle of 0° which exhibits superhydrophilicity.

[0033] Further, the gas diffusion layer for anion exchange membrane electrolysis may have a gas permeability of 1×10^{-12} m^2 or more. The gas diffusion layer for anion exchange membrane electrolysis of the present invention is a better product as the gas permeability is higher. Specifically, the gas permeability may be 1×10^{-12} m^2 or more, 1.25×10^{-12} m^2 or more, 1.5×10^{-12} m^2 or more, 1.75×10^{-12} m^2 or more or 2×10^{-12} m^2 or more, and 4×10^{-12} m^2 or less, 3×10^{-12} m^2 or less, or 2.5×10^{-12} m^2 or less.

[0034] Considering that when the gas permeability is low, the mobility of oxygen and hydrogen, which must be discharged to the outside through the gas diffusion layer, is lowered, there may be a problem that when used for water electrolysis, and the like, the overall efficiency decreases.

[0035] The gas permeability of the present invention was measured using a device for evaluating GDL basic physical properties (CPRT 10, Korea Institute of Energy Technology's own standard). By randomly contacting the surface of the gas diffusion layer and performing measurements three times in total, the gas permeability can be determined using the following paper and Equation 1.

[0036] The gas permeability was determined using the Trough plane permeability method described in the paper "In-plane and through-plane gas permeability of carbon ber electrode backing layers (Jeff T. Gostick, et al., Sep 1 2006)," and the gas permeability (K, unit m^2) can be calculated according to the following Equation 1.

[Equation 1]

$$K = \frac{\mu}{A/t}\frac{m}{P_1 - P_2}\left(\frac{RT}{MP_{agg}}\right)$$

in Equation 1,

K is the gas permeability,
μ is the dynamic viscosity of the gas used,
A is the area of the cross section through which gas permeated,
t is the thickness of the gas diffusion layer through which the gas penetrated,
m is the flow rate (mass) of gas flowing through a unit area,
P_1 is the pressure before gas permeation,
P_2 is the pressure after gas permeation,
R is the gas constant,
T is the temperature,
M is the weight of the gas used, and
P_{avg} means the average value of P_1 and P_2.

[0037] Further, the gas diffusion layer for anion exchange membrane electrolysis may further include a porous support on one surface of the reticular carbon nanofiber film. Specifically, the gas diffusion layer may have a form in which a carbon nanofiber film is provided on one surface of a porous support.

[0038] The porous support is not limited as long as it is a porous material, but may be, for example, a porous carbon paper, a carbon fiber film, a carbon nanofiber film, and a carbon nanotube film. Further, the porous support may have a thickness of 50 to 500 ,um. Alternatively, the porous support may have a thickness of 100 to 500 μm.

[0039] According to the present invention, there can be provided a method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis, the method comprising: spinning a carbon precursor material to form a reticular molded product (step 1); and heat treating the reticular molded product to obtain a reticular carbon nanofiber film (step 2).

[0040] The carbon precursor material in the step 1 may be at least one selected from the group consisting of polyacrylonitrile, polyvinyl alcohol, polyvinylpyrrolidone, and precursors thereof, cellulose, lignin, and pitch.

[0041] The spinning of the (step 1) may be at least one selected from the group consisting of electrospinning, centrifugal jet spinning, melt-blown spinning, and spray spinning. Specifically, the spinning may be electrospinning.

[0042] When spinning is performed through electrospinning, a carbon precursor material in a solution state may be spun through a nozzle with a diameter of 1 to 5 mm. In addition, a voltage of 5 to 30 kV may be applied to the nozzle, and the distance between the nozzle and the ground may be 5 to 30 cm. At this time, the carbon precursor material in a solution state may be spun through the nozzle at a constant rate between 1 and 10 ml/hour. One or more nozzles can be used, and the number of nozzles can be increased or the spinning time can be increased to obtain the desired thickness.

[0043] Through the heat treatment of the (step 2), the reticular molded product formed in the spinning step is carbonized to form a reticular carbon nanotube film. The heat treatment step is a step of first stabilizing the reticular molded product under an air atmosphere or an oxygenmixed gas atmosphere in a temperature range of 200 to 400°C for 1 hour or more, and then carbonizing it using an inert gas at a temperature of 700 to 1500°C for 1 hour or more. Subsequently, it is cooled to room temperature under an inert gas atmosphere to obtain a reticular carbon nanotube film. At this time, cooling during carbonization, after the stabilization process, is carried out while maintaining an inert gas atmosphere.

[0044] In addition, spinning the carbon precursor material in the (step 1) may be spinning onto a porous support.

[0045] Alternatively, the method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis may further comprise a step of attaching the reticular carbon nanotube film obtained in (step 2) to a porous support. Specifically, a method of attaching a porous support to the reticular carbon nanotube film may be a method of applying an adhesive to one surface of the porous support and attaching it. The adhesive used at this time may contain a material having high electrical conductivity, or may contain a material having high viscosity, or may contain both.

[0046] The porous support is not limited as long as it is a porous material, but may be, for example, a porous carbon paper, a carbon fiber film, a carbon nanofiber film, a carbon nanotube film, and the like. Further, the porous support may have a thickness of 50 to 500 um. Alternatively, the porous support may have a thickness of 100 to 500 μm.

[0047] Hereinafter, preferable examples are presented for better understanding the present invention. However, the following examples are only for illustrating the present invention, and the present invention is not limited to or by them.

Example

[0048] In this example, a gas diffusion layer, which is a reticular carbon nanotube film, was manufactured using an electrospinning method. First, polyacrylonitrile, which is a carbon precursor material in a solution state, was spun at a constant speed of 5 ml per hour through a 3 mm diameter nozzle for 3 hours. At this time, aluminum foil is placed on the grounded collector so as to facilitate separation of the sample, and polyacrylonitrile was electrospun thereon. A voltage of 15 kV was applied to the nozzle. The distance between the nozzle and the ground was maintained at 15 cm. One or more nozzles can be used, but in this example, polyacrylonitrile was spun to a thickness of 500 μm using one nozzle to form a reticular molded product.

[0049] The reticular molded product was then stabilized under an atmospheric atmosphere (including oxygen) at a temperature of 270 to 300°C for 1 hour. Subsequently, it was carbonized under an inert gas N_2 atmosphere and in the temperature section of 1000°C for 1 hour. After the heat treatment process, the sample was cooled to room temperature to obtain a reticular carbon nanofiber film. The formed reticular carbon nanofiber film had a thickness of 200 μm.

Comparative Example

[0050] As a comparative example, a PEMFC gas diffusion layer product, which is commercially available under the product name JTN20-A6H available from JNTG, was used.

Experimental Example 1 - Surface observation

[0051] The surface and side surfaces of the manufactured reticular carbon nanofiber film were observed using a scanning electron microscope(SEM), and are shown in FIGS. 1 and 2, respectively.

[0052] As can be seen from the figure, it was confirmed that the carbon nanofiber film, which is the gas diffusion layer of the present invention, was formed into a reticular film shape and maintained its porosity.

Experimental Example 2 - Measurement of water contact angle

[0053] The water contact angle of the reticular carbon nanofiber film of Example with distilled water droplet was measured, and shown in FIG. 3. The water contact angle was measured by placing a film-like sample on a flat surface, dropping 5 to 10 $\mu\ell$ of distilled water, observing its shape from the side using DSA, and calculating the angle.

[0054] As can be seen from the figure, the reticular carbon nanofiber film of Example shows superhydrophilicity with a contact angle of 0° compared to the gas diffusion layer of Comparative Example.

Experimental Example 3 - Observation of gas permeability

[0055] The gas permeability of the present invention was measured using a device for evaluating GDL basic physical properties (CPRT 10, Korea Institute of Energy Technology's own standard). By randomly contacting the surface of the gas diffusion layer and performing measurements three times in total, the gas permeability was determined using the following paper and Equation 1.

[0056] The gas permeability was determined using the Trough plane permeability method described in the paper "In-plane and through-plane gas permeability of carbon ber electrode backing layers (Jeff T. Gostick, et al., Sep 1 2006)," and the gas permeability (K, unit m^2) was calculated according to the following [Equation 1], and shown in FIG. 4.

[Equation 1]

$$\mathrm{K} = \frac{\mu}{A/t}\frac{m}{P_1 - P_2}\left(\frac{RT}{MP_{agg}}\right)$$

in Equation 1,

K is the gas permeability,
μ is the dynamic viscosity of the gas used,
A is the area of the cross section through which gas permeated,
t is the thickness of the gas diffusion layer through which the gas penetrated,
m is the flow rate (mass) of gas flowing through a unit area,
P_1 is the pressure before gas permeation,
P_2 is the pressure after gas permeation,
R is the gas constant,
T is the temperature,
M is the weight of the gas used, and
P_{avg} means the average value of P_1 and P_2.
The test gas used at this time was dry air.

[0057] Comparing the gas permeability of the reticular carbon nanofiber film of Example and the gas diffusion layer of Comparative Example, it was confirmed that Example exhibited more excellent gas permeability.

Claims

1. A gas diffusion layer for anion exchange membrane electrolysis comprising a reticular carbon nanofiber film,
   wherein the carbon nanofiber has a diameter of 500 nm or less.

2. The gas diffusion layer for anion exchange membrane electrolysis according to claim 1, wherein:
   the gas diffusion layer for anion exchange membrane electrolysis has a contact angle with water of 20° or less.

3. The gas diffusion layer for anion exchange membrane electrolysis according to claim 1, wherein:
   the gas diffusion layer for anion exchange membrane electrolysis has a gas permeability of 1×10^{-12} m^2 or more.

4. The gas diffusion layer for anion exchange membrane electrolysis according to claim 1, wherein:
   the gas diffusion layer for anion exchange membrane electrolysis further comprises a porous support on one surface of the reticular carbon nanofiber film.

5. The gas diffusion layer for anion exchange membrane electrolysis according to claim 4, wherein:
   the porous support is at least one selected from the group consisting of a porous carbon paper, a carbon fiber film, a carbon nanofiber film, and a carbon nanotube film.

6. A method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis, the method comprising:

   spinning a carbon precursor material to form a reticular molded product (step 1); and
   heat treating the reticular molded product to obtain a reticular carbon nanofiber film (step 2).

7. The method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis according to claim 6, wherein:
   the carbon precursor material in the step 1 is at least one selected from the group consisting of polyacrylonitrile, polyvinyl alcohol, polyvinylpyrrolidone, and precursors thereof, cellulose, lignin, and pitch.

8. The method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis according to claim 6, wherein:
   the spinning of the (step 1) is at least one selected from the group consisting of electrospinning, centrifugal jet spinning, melt-blown spinning, and spray spinning.

9. The method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis according to claim 8, wherein:

   the electrospinning spins a carbon precursor material in a solution state through a nozzle with a diameter of 1 to 5 mm, and
   a voltage of 5 to 30 kV is applied to the nozzle.

10. The method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis according to claim 6, wherein:
    the heat treatment of step 2 is performed at a temperature of 700 to 1500°C for one hour or more.

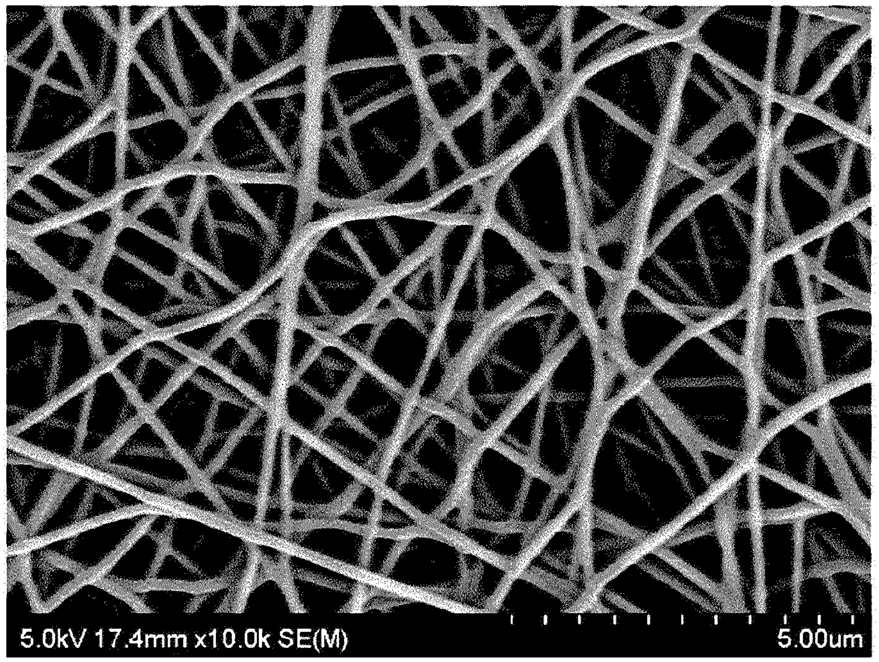
11. The method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis according to claim 6, wherein:
    spinning the carbon precursor material in the step 1 is spinning onto a porous support.

12. The method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis according to claim 6, wherein:
    the manufacturing method further comprises attaching the reticular carbon nanotube film obtained in step 2 to a porous support.

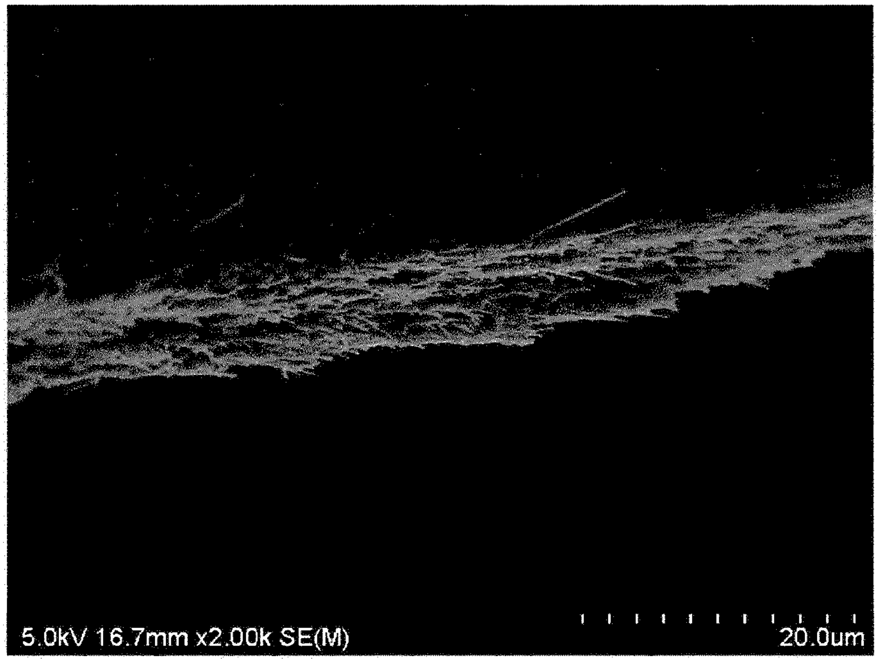
13. The method for manufacturing a gas diffusion layer for anion exchange membrane electrolysis according to claim 11 or 12, wherein:
    the porous support is at least one selected from the group consisting of a porous carbon paper, a carbon

fiber film, a carbon nanofiber film, and a carbon nanotube film.

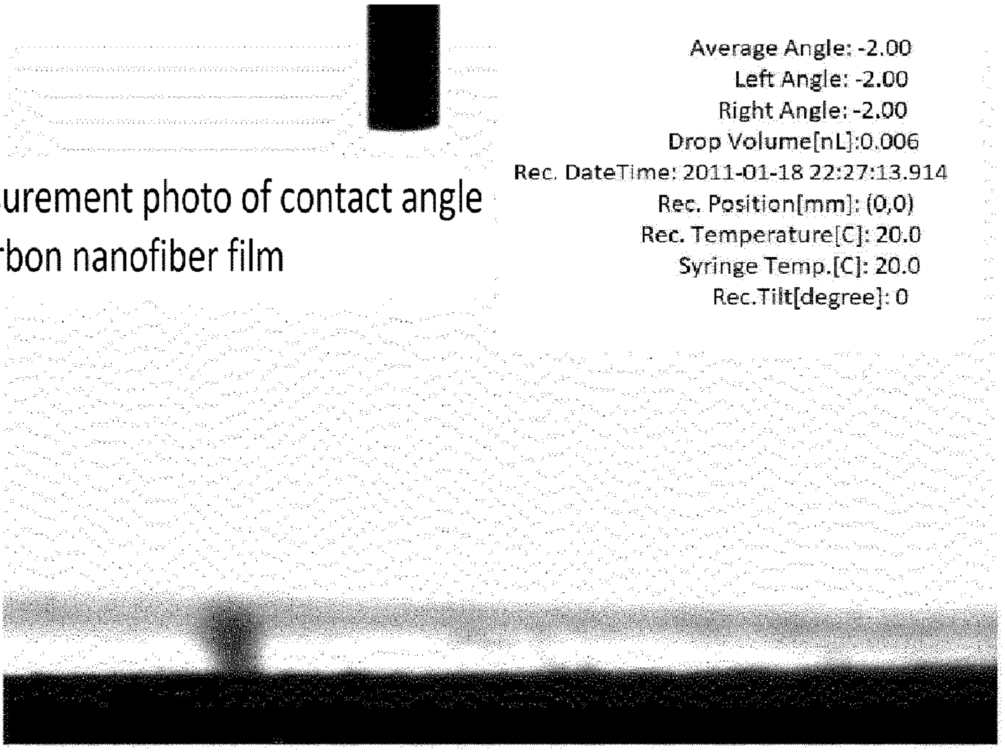
【FIG. 1】

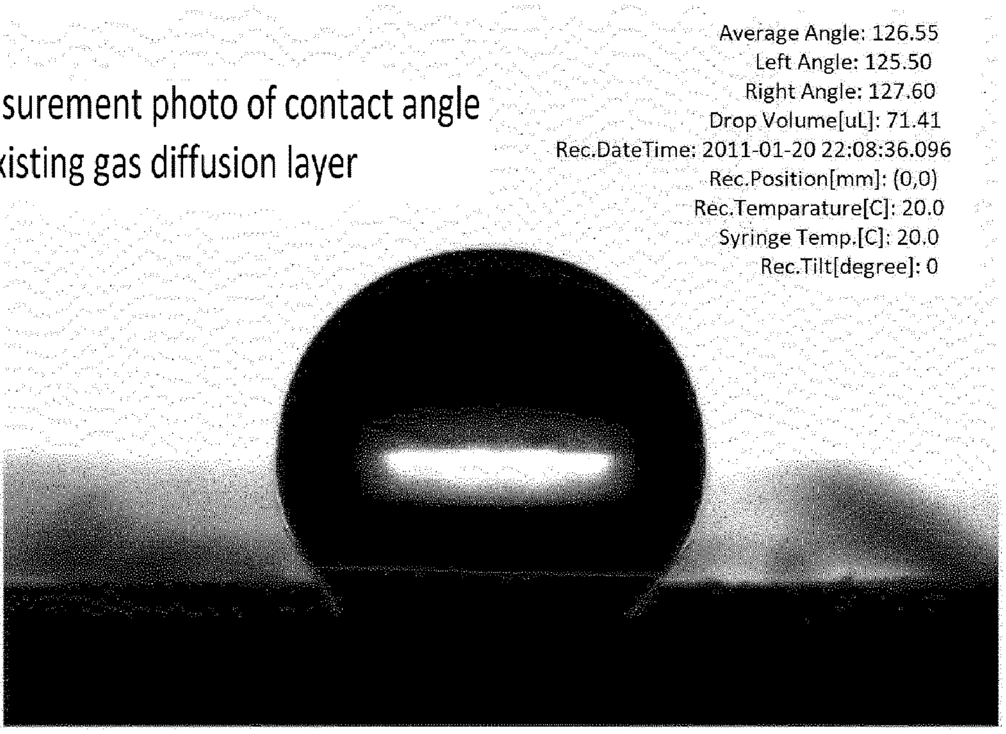


【FIG. 2】

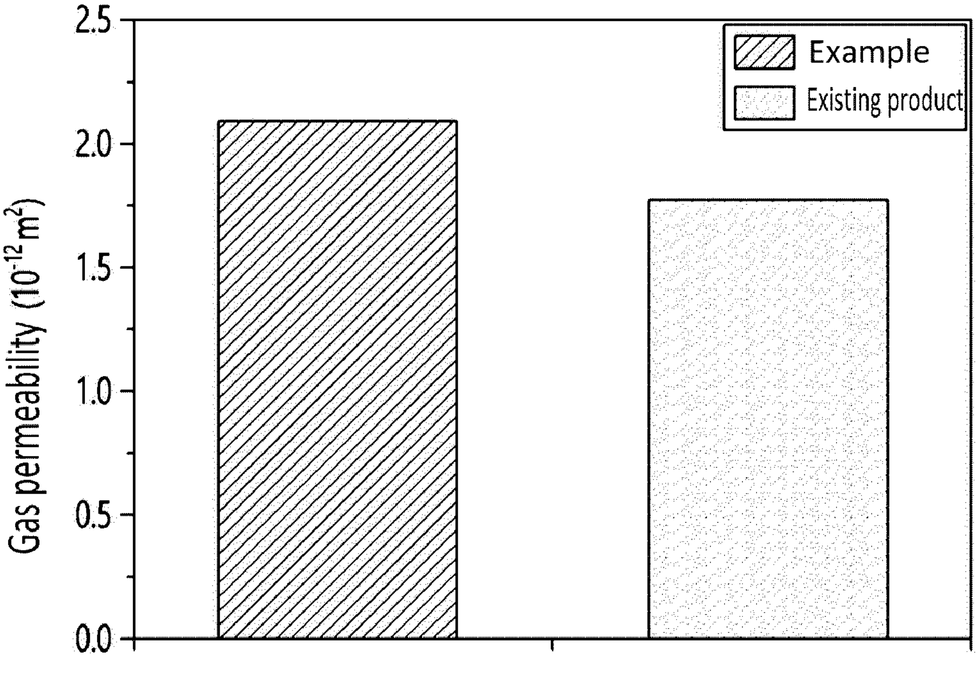


【FIG. 3】


Measurement photo of contact angle of carbon nanofiber film


Measurement photo of contact angle of existing gas diffusion layer

【FIG. 4】


2.5
2.0
1.5
1.0
0.5
0.0
Gas permeability (10^{-12} m^2)
Example
Existing product

INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2022/018750

A. CLASSIFICATION OF SUBJECT MATTER

C25B 11/032(2021.01)i; **C25B 1/04**(2006.01)i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C25B 11/032(2021.01); C25B 11/03(2006.01); C25B 11/04(2006.01); D01F 9/14(2006.01); H01M 4/88(2006.01); H01M 4/96(2006.01); H01M 8/0245(2016.01); H01M 8/1018(2016.01)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Korean utility models and applications for utility models: IPC as above
Japanese utility models and applications for utility models: IPC as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

eKOMPASS (KIPO internal) & keywords: 연료전지(fuel cell), 기체확산층(gas diffusion layer), 수전해(water electrolysis), 탄소나노섬유(carbon nano fiber)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 5672239 B2 (NISSHINBO HOLDINGS INC.) 18 February 2015 (2015-02-18) See claims 1, 3 and 4; paragraphs [0028] and [0029]; and figures 1 and 2.	1-13
Y	KR 10-2019-0021551 A (ELCHEM TECH CO., LTD.) 06 March 2019 (2019-03-06) See claims 1 and 4; and paragraph [0053].	1-13
A	JP 2018-204150 A (JAPAN VILENE CO., LTD.) 27 December 2018 (2018-12-27) See claims 1-15; and figures 1 and 3.	1-13
A	JP 5176021 B2 (JOHNSON MATTHEY PUBLIC LIMITED COMPANY) 03 April 2013 (2013-04-03) See entire document.	1-13
A	KR 10-2017-0139914 A (HYUNDAI MOTOR COMPANY) 20 December 2017 (2017-12-20) See entire document.	1-13

☐ Further documents are listed in the continuation of Box C. ☑ See patent family annex.

* Special categories of cited documents:	
"A" document defining the general state of the art which is not considered to be of particular relevance	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"D" document cited by the applicant in the international application	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E" earlier application or patent but published on or after the international filing date	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&" document member of the same patent family
"O" document referring to an oral disclosure, use, exhibition or other means	
"P" document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search **14 March 2023**	Date of mailing of the international search report **15 March 2023**
Name and mailing address of the ISA/KR **Korean Intellectual Property Office** **Government Complex-Daejeon Building 4, 189 Cheongsa-ro, Seo-gu, Daejeon 35208** Facsimile No. **+82-42-481-8578**	Authorized officer Telephone No.

Form PCT/ISA/210 (second sheet) (July 2022)

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/KR2022/018750

Patent document cited in search report	Publication date (day/month/year)	Patent family member(s)	Publication date (day/month/year)
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