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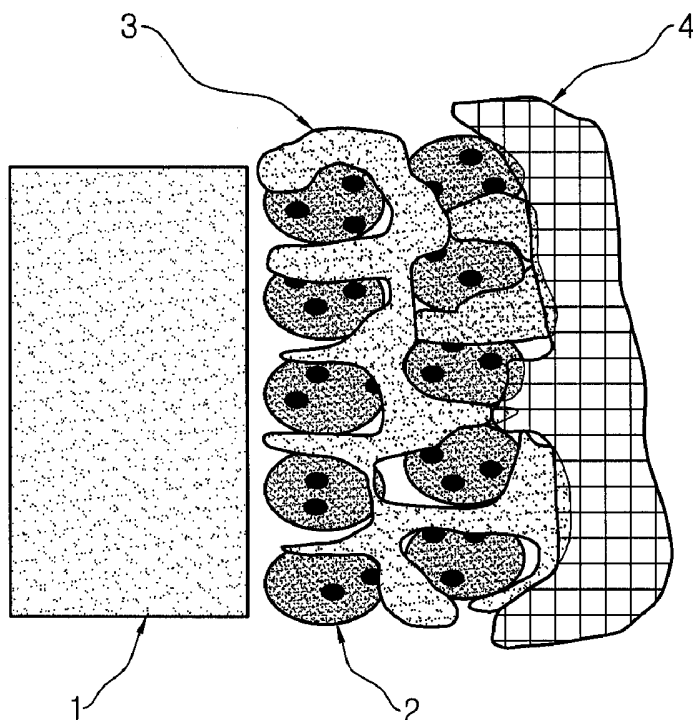
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(54) Title: MULTI-LAYERED ELECTRODE FOR FUEL CELL AND METHOD FOR PRODUCING THE SAME



(57) Abstract: Disclosed are a multi-layered electrode for fuel cell and a method for producing the same, wherein the electrode can be operated under non-humidification and normal temperature, the flooding of the electrode catalyst layer can be prevented, and the long-term operation characteristic can be increased due to the prevention of the loss of the electrode catalyst layer.



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Description

MULTI-LAYERED ELECTRODE FOR FUEL CELL AND METHOD FOR PRODUCING THE SAME

Technical Field

- [1] The present invention relates to a multi-layered electrode for fuel cell and a method for producing the same, and particularly to a multi-layered electrode for fuel cell and a method for producing the same, wherein a distribution of the equivalent weight (hereinafter referred to "EW") of ionomer at an inside of an electrode catalyst layer and on a surface of an electrode catalyst layer is regulated so that the operation under non-humidification and normal temperature is possible, the flooding of the electrode catalyst layer can be prevented, and the long-term operation characteristic can be increased due to the prevention of the loss of the electrode catalyst layer.

Background Art

- [2] Techniques for producing an electrode for fuel cell are known in the art as follows:
- [3] For example, a method for producing an electrode has been proposed by P. Driven et al., wherein water repellency treated carbon powders are applied on a water repellency treated porous carbon paper to form an intermediate layer, on which layer a mixture of catalyst and polymer electrolyte, i.e., high EW ionomer (e.g., EW=1,100), is then applied thinly, thereby preparing an electrode.
- [4] Then, a commercial electrolyte membrane is placed between the anode and the cathode as prepared, and hot-pressing is performed at above glass transition temperature of the electrolyte and under a certain pressure to produce a MEA <see P. Driven and W. Engelen, US Patent No. 5,561,000 (1996)>.
- [5] In producing an electrode using such method, an addition of a nafion ionomer to the electrode catalyst layer makes three-interface advantageously increased. Further, the prevention of the flooding comes to be possible.
- [6] However, such method has problems that due to the poor water absorbency of the nafion ionomer, water produced by electrochemical reaction in cathode can not be used sufficiently so that the non-humidification operation is substantially impossible to thus require an external humidifier, which correspondingly causes additional costs and volume increase.
- [7] Furthermore, such method has another problems that due to the insufficient indirect annealing of electrode by hot-pressing, the nafion ionomer is leaked together with the catalyst during the operation of the fuel cell, which is disadvantageous in long-term operation of the fuel cell.
- [8] Meanwhile, a three-layered structure catalyst layer of a cathode electrode has been

proposed by Yoshitake et al., wherein the first catalyst layer directly contacting the first electrolyte membrane includes an ion conductive polymer ionomer with high ion exchange capacity, i.e., with low EW, the second catalyst layer adjacent to the first catalyst layer includes an ion conductive polymer ionomer with high oxygen solubility, and the third catalyst layer includes soluble fluoropolymer <see M. Yoshitake, I. Terada, H. Shimoda, A. Watanabe, K. Yamada, K. Min and Y. Kunisa, "2002 Fuel Cell Seminar Abstracts", 826(2002)>.

- [9] In producing an electrode using such method, a nafion ionomer with high EW is added to the first and second catalyst layers so that in comparison with the prior electrode, interface resistance between the electrode and the electrolyte membrane can be advantageously reduced and the oxygen concentration in the catalyst layer can be increased to improve the performance of the fuel cell.
- [10] However, such method has problems that due to the generation of flooding resulted from high water absorbency, the performance can be abruptly reduced. Further, the method still has problems that due to the insufficient indirect annealing of electrode by hot-pressing, the nafion ionomer comes to be leaked together with the catalyst during the operation of the fuel cell, which is disadvantageous in long-term operation of the fuel cell.
- [11] Specifically, according to the above prior methods, the MEA was produced with the hot pressing under the conditions of 120~140°C temperature, 100~200 atm pressure and 30 seconds to 2 minutes time. Since the phase inversion of the nafion ionomer in the electrode catalyst layer does not occur sufficiently under such conditions, the nafion ionomer serving to transfer hydrogen ion between the electrode and the electrolyte membrane and to bind the catalyst comes to be re-solved by wet reaction gas and leaked out together with the catalyst during the operation of the fuel cell, so that the electrode and the electrolyte membrane are separated from each other to increase resistance, thereby causing the decrease of the performance of fuel cell and giving disadvantageous effects to the long-term operation thereof.

Disclosure of Invention

Technical Problem

- [12] The present invention is to solve the above problems, and an object of the invention is to provide a multi-layered electrode for fuel cell and a method for producing the same, in which the operation under non-humidification and normal temperature is possible, the flooding of an electrode catalyst layer is prevented, and a long-term operation characteristic is improved due to the prevention of the loss of the electrode catalyst layer.

Technical Solution

- [13] In order to accomplish the object, there is provided a multi-layered electrode for fuel cell comprising: a gas diffusing layer; a catalyst layer formed on the gas diffusing layer and applied with a catalyst slurry containing a solution of a nafion ionomer with a specific equivalent weight (EW) and a catalyst; and a coating layer formed on the catalyst layer and coated with a solution of a nafion ionomer with lower EW than the specific EW of the former nafion ionomer.
- [14] In order to accomplish the object, there is also provided a method for producing a multi-layered electrode for fuel cell comprising the steps of: (S1) preparing a catalyst slurry containing a catalyst and a solution of a nafion ionomer with specific equivalent weight (EW); (S2) applying the catalyst onto a gas diffusing layer; (S3) first-drying the catalyst slurry applied on the gas diffusing layer to form a catalyst layer; (S4) applying on the surface of the dried catalyst layer a solution of a nafion ionomer with the lower EW than the specific EW of the nafion ionomer of the catalyst slurry; (S5) second-drying the applied solution of the nafion ionomer to form an electrode; and (S6) annealing the dried electrode.

Advantageous Effects

- [15] According to a multi-layered electrode for fuel cell and a method for producing the same, the operation under non-humidification and normal temperature is possible, the flooding of an electrode catalyst layer can be prevented, and a long-term operation characteristic comes to be improved due to the prevention of the loss of the electrode catalyst layer.

Brief Description of the Drawings

- [16] Fig. 1 is a schematic view of a structure of a multi-layered electrode for fuel cell according to the present invention.
- [17] Fig. 2 is a schematic view of a structure of a MEA for fuel cell according to the present invention.
- [18] Fig. 3 is a graph showing performances in case of a comparative example and an example under 80°C standard humidification operation condition according to an experiment 1 of the present invention, wherein ■ indicates the case of the MEA of the example, and ▲ indicates the case of the MEA of the comparative example.
- [19] Fig. 4 is a graph showing performances in case of a comparative example and an example under non-humidification operation condition according to an experiment 2 of the present invention, wherein ■ indicates the case of the MEA of the comparative example and ▲ indicates the case of the MEA of the example.

Best Mode for Carrying Out the Invention

- [20] Hereinafter, a multi-layered electrode for fuel cell and a method for producing the same according to the present invention will be described in detail.

- [21] According to the present invention, a nafion ionomer with relatively high equivalent weight (EW) to that of a nafion ionomer, which will be used on a surface of a catalyst layer, is put inside the catalyst layer so that the foolding of the catalyst can be prevented. Further, the nafion ionomer with relatively low EW, i.e., with high water absorbency and ionic conductivity to that of said former nafion ionomer put inside the catalyst layer is applied on the surface of the catalyst layer so that an interface resistance between an electrode and an electrolyte membrane can be reduced while improving a performance of a fuel cell by using water produced by an electrochemical reaction, thereby enabling the non-humidification operation. Finally, mechanical properties of the nafion ionomers inside the catalyst layer and on the surface of the catalyst layer are made identical to that of a commercial electrolyte membrane by annealing the electrode before assembling the electrode and the electrolyte membrane to form the MEA so that the catalyst and the nafion ionomer come not to be leaked out during the operation of the fuel cell, which ensures a long-term operation.
- [22] Fig. 1 is a schematic view of the structure of the multi-layered electrode for fuel cell according to the present invention.
- [23] As shown in Fig. 1, the multi-layered electrode for fuel cell of the present invention includes Pt/C catalysts 2 applied on a gas diffusing layer 1, a nafion ionomer 3 with specific equivalent weight (EW) applied between the catalysts 2, and a coating layer containing a nafion ionomer 4 with lower EW than specific EW of the former nafion ionomer 3 on the surface of a catalyst layer including the nafion ionomer 3 and the catalysts 2.
- [24] The multi-layered electrode for fuel cell is provided such that a perfluorinated sulfonic acid nafion ionomer, which has relatively high EW, i.e., relatively low ionic conductivity and water absorbency to the following nafion ionomer, is included inside the catalyst layer, i.e., the catalyst layer on the gas diffusing layer, and then a coating layer of the nafion ionomer solution, which has relatively low EW, i.e., relatively high ionic conductivity and water absorbency to the former nafion ionomer, is formed on the surface of the catalyst layer prepared as described above.
- [25] Further, the prepared multi-layered electrode is annealed at 120~170°C temperature for 0.5~24 hours.
- [26] Fig. 2 is a schematic view of the structure of the MEA for fuel cell according to the present invention.
- [27] As shown in Fig. 2, the prepared multi-layered electrode and a commercial nafion electrolyte membrane 5 are assembled together to make the MEA.
- [28] The present invention follows the below exemplary process in order to produce the multi-layered electrode for fuel cell.
- [29] First, a catalyst slurry containing a nafion ionomer with specific EW is prepared

(S1).

- [30] That is, to prepare the catalyst slurry, 20~60wt% Pt/C (Vulcan XC-72R) is used. The total amount of the nafion ionomer used in the present invention (total amount of the nafion ionomer used in the catalyst layer and on the surface of the catalyst layer) is 20~50% of the weight of the Pt/C catalyst. In preparing the catalyst slurry (i.e., this is the case where the nafion ionomer is used in the catalyst layer), a nafion ionomer with 1,000~1,300 EW, 5wt% or less and 1/12~4/12 of the total amount of the nafion ionomer is stirred with IPA solvent or EtOH solvent for 20 minutes to 24 hours.
- [31] The most excellent performance is shown in case that the amount of 1/12~4/12 of the total amount of the nafion ionomer is used in the catalyst layer. If used less than 1/12 of the total amount of the nafion ionomer, three-interface comes to be less-formed, and if used more than 4/12 of the total amount of the nafion ionomer, the catalyst comes to be covered completely.
- [32] Herein, it is preferable that 20~40wt% Pt/C (Vulcan XC-72R) and a nafion ionomer with 1,100~1,200 EW, 5wt% or less and 1/12~2/12 of the total amount of the nafion ionomer are stirred together with IPA solvent or EtOH solvent for 20 minutes to 24 hours.
- [33] Next, the catalyst slurry is applied onto the gas diffusing layer (S2).
- [34] Namely, the catalyst slurry as prepared above is applied in a loading quantity of 0.04~1.2mg-Pt/C/cm² onto 20~60wt% PTFE carbon paper or carbon fabric, which is the gas diffusing layer, by means of spraying method or tape casting method. Herein, it is preferable to use 20~40wt% PTFE carbon paper as the gas diffusing layer and set the loading quantity to be 0.4~0.7mg-Pt/C/cm².
- [35] Next, the catalyst slurry applied on the gas diffusing layer is first-dried (S3).
- [36] That is, after applying the catalyst as described above, a drying is performed in an oven at 60~80°C temperature for 30 minutes to 24 hours. Herein, it is preferable to perform the drying in the oven at 60~80°C temperature for 1~2 hours.
- [37] Next, a solution of a nafion ionomer with lower EW than the specific EW of the nafion ionomer of the catalyst slurry is applied onto the dried surface of the catalyst layer (S4).
- [38] That is, a nafion ionomer which is 5wt% or less and has 400~1,000 EW (or a nafion ionomer with 1,100~1,200 EW, and if the nafion ionomer of the catalyst slurry has 1,100~1,200 EW, the lower EW should be chosen) and a remainder of the total amount of the nafion ionomer (i.e., 20~50wt% of the weight of the Pt/C catalyst) excepting the amount of the nafion ionomer of the catalyst slurry [i.e., 1/12~4/12 of the total amount of the nafion ionomer (preferably 1/12~2/12)] is stirred with IPA solvent or EtOH solvent for 20 minutes to 24 hours, thereby preparing a nafion ionomer solution.
- [39] The nafion ionomers used in the steps S1 and S4 are different each other in EW at

the inside and the surface of the catalyst layer, wherein the EW is relatively low at the surface of the catalyst layer. As described above, the used total amount of the nafion ionomer is 20~50% of the weight of the Pt/C catalyst, wherein 1/12~4/12 (preferably 1/12~2/12) of the total amount of the nafion ionomer exists inside the catalyst layer and the remainder of the total amount of the nafion ionomer excepting the amount of the nafion ionomer of the catalyst slurry exists on the surface of the catalyst layer.

[40] Next, performed is the second drying of what was applied with the nafion ionomer solution (S5).

[41] That is, after the applying of the nafion ionomer solution, a drying is performed in an oven at 60~80°C temperature for 30 minutes to 24 hours. Herein, it is preferable to perform the drying at 60~80°C temperature for 1~2 hours.

[42] Next, the prepared electrode is annealed in a vacuum oven (S6).

[43] That is, the electrode is annealed in the vacuum oven at 120-170°C temperature for 0.5~24 hours, whereby the phase inversion occurs so that a mechanical strength of the nafion becomes identical to that of the commercial nafion electrolyte membrane.

[44] Herein, it is more preferable to perform the annealing in the vacuum oven at 150~170°C temperature for 6~18 hours.

[45] In this way, if the annealing is performed in the vacuum oven at above glass transition temperature (120~170°C) for 0.5~24 hours, the phase inversion occurs in the SCM (solution cast membrane) prepared by the nafion ionomer solution and the nafion ionomer of the electrode catalyst layer, the mechanical properties are improved like the commercial membrane, and strong bonding to each other are obtained so as to prevent the loss of the catalyst and the nafion ionomer during the operation of the fuel cell. Accordingly the interface resistance during the operation of the fuel cell can be maintained at the substantially same level as an initial state in the MEA production, which means advantages in the long-term operation of the fuel cell.

[46] In addition, if annealed at above glass transition temperature for a sufficient time, the nafion ionomer comes to have improved mechanical properties since the main chains composed of C-F are fused to each other and sulfonic groups ($-\text{SO}_3\text{H}$) are rotated to orient from outside to inside and form an ionic bonding therebetween.

[47] The MEA with the nafion commercial membrane is prepared through such annealing, so that its mechanical properties come to increase and its lifetime extends to 1,000~100,000 hours.

[48] The present invention will be described in detail by comparing the preferred example of the invention with comparative example. However, the present invention is not limited to the following example and various modification can be realized within the scope of the appended claims. The following example is intended to completely explain the present invention as well as to render the persons skilled in the art to easily

perform the present invention.

[49] <MEA production of example>

[50] As for a catalyst material in an electrode, Pt/C powders (electrocam or E-tech), in which 20wt% Pt is supported in a carbon black carrier (Vulcan XC-72R, Cabot), were used, and as for a catalyst support, a carbon paper (TGPH-060) with 20wt% PTFE (polytetrafluorethylene) was used in order to provide a water repellency.

[51] In order to prepare an ink for electrode, a Pt/C catalyst and a nafion ionomer (EW=1,100), which is 5wt% and has the amount of 4/12 of the total quantity of the nafion ionomer if the used total quantity of the nafion ionomer is 33% of the weight of the Pt/C catalyst, were dispersed into isopropyl alcohol (IPA). For homogeneous dispersion, ultrasonic treatment was performed.

[52] The prepared catalyst ink was coated onto the carbon paper using an air brush gun, on which carbon paper the nafion ionomer (EW=1,000), which is 5wt% and has the remainder of the total quantity of the nafion ionomer excepting the nafion ionomer with 1,100 EW and is diluted with IPA, was additionally applied onto the surface of the catalyst to thus form the catalyst layers of anode and cathode. The Pt loading quantity of the catalyst was $0.4\text{mg}/\text{cm}^2$ at anode and $0.7\text{mg}/\text{cm}^2$ at cathode.

[53] The prepared anode and cathode were placed at both sides of the pre-treated nafion 115 polymer electrolyte membrane (EW=1,100). Then, a hot-pressing was performed to produce an MEA. Herein, a temperature was 140°C , a pressure 200atm, and hot pressing time 90 seconds.

[54] <MEA production of comparative example>

[55] As for a catalyst material in an electrode, Pt/C powders (electrocam or E-tech), in which 20wt% Pt is supported in a carbon black carrier (Vulcan XC-72R, Cabot), were used, and as for a catalyst support, a carbon paper (TGPH-060) with 20wt% PTFE (polytetrafluorethylene) was used in order to provide a water repellency.

[56] In order to prepare an ink for electrode, a Pt/C catalyst and a nafion ionomer (EW=1,100), which is 5wt% and has the amount of 4/12 of the total quantity of the nafion ionomer if the used total quantity of the nafion ionomer is 33% of the weight of the Pt/C catalyst, were dispersed into isopropyl alcohol (IPA). For homogeneous dispersion, ultrasonic treatment was performed.

[57] The prepared catalyst ink was coated onto the carbon paper using an air brush gun, on which carbon paper the nafion ionomer (EW=1,100), which is 5wt% and has the remainder of the total quantity of the nafion ionomer excepting the nafion ionomer with 1,100 EW and is diluted with IPA, was additionally applied onto the surface of the catalyst to thus form the catalyst layers of anode and cathode. The Pt loading quantity of the catalyst was $0.4\text{mg}/\text{cm}^2$ at anode and $0.7\text{mg}/\text{cm}^2$ at cathode.

[58] The prepared anode and cathode were placed at both sides of the pre-treated nafion

115 polymer electrolyte membrane (EW=1,100). Then, a hot-pressing was performed to produce an MEA. Herein, a temperature was 140°C, a pressure 200atm, and hot pressing time 90 seconds.

[59] <Experiment 1: 80°C standard humidification operation condition>

[60] In order to evaluate the performances of the MEAs of the example and the comparative example, an unit cell experiment was performed. In the experiment 1, the performances under 80°C standard humidification operation condition were measured and compared with each other.

[61] Fig. 3 is a graph showing performances in case of the comparative example and the example under 80°C standard humidification operation condition according to this experiment 1 of the present invention, wherein ■ indicates the case of the MEA of the example, and ▲ indicates the case of the MEA of the comparative example.

[62] As shown in Fig. 3, it can be seen that the MEA of the example is excellent in performance in that its current density increases at the same voltage.

[63] <Experiment 2: normal temperature and non-humidification operation condition>

[64] In the experiment 2, the performances under normal temperature and non-humidification operation condition were measured and compared with each other.

[65] Fig. 4 is a graph showing performances in case of the comparative example and the example under non-humidification operation condition according to this experiment 2 of the present invention, wherein ■ indicates the case of the MEA of the comparative example and ▲ indicates the case of the MEA of the example.

[66] As shown in Fig. 4, it can be seen that the MEA of the example is remarkably excellent in performance.

Industrial Applicability

[67] According to the present invention, a multi-layered electrode for fuel cell and a method for producing the same are provided, wherein a distribution of the equivalent weight (EW) of ionomer is regulated at the inside of the electrode catalyst layer and on the surface of the electrode catalyst layer so that the prepared electrode can be operated under non-humidification and normal temperature, the flooding of the electrode catalyst layer can be prevented, and the long-term operation characteristic can be increased due to the prevention of the loss of the electrode catalyst layer.

Claims

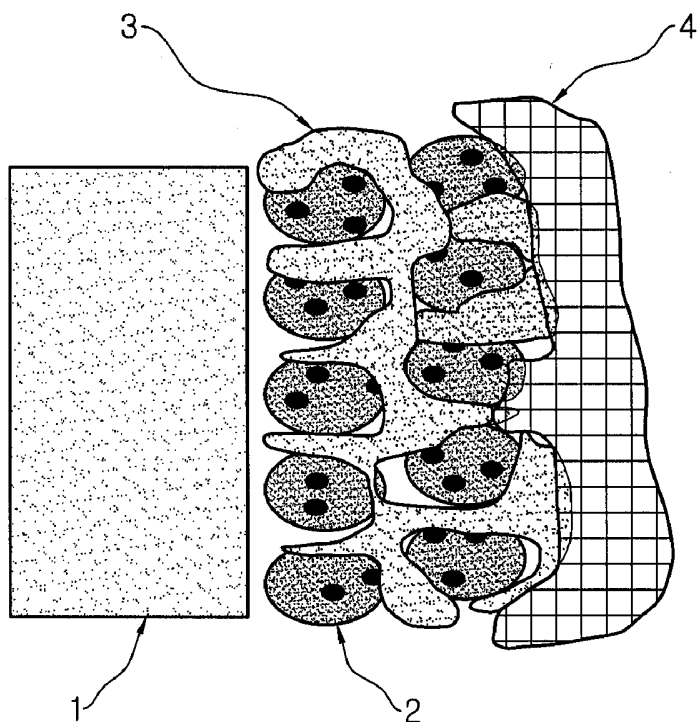
- [1] A multi-layered electrode for fuel cell comprising:
a gas diffusing layer 1;
a catalyst layer formed on the gas diffusing layer 1 and applied with a catalyst slurry containing a catalyst 2 and a solution of a nafion ionomer 3 with a specific equivalent weight (EW); and
a coating layer formed on the catalyst layer and coated with a solution of a nafion ionomer 4 with lower EW than the specific EW of the former nafion ionomer 3.
- [2] The multi-layered electrode according to claim 1, wherein the electrode is annealed at a temperature of 120~170°C.
- [3] The multi-layered electrode according to claim 1 or 2, wherein the gas diffusing layer 1 is 20~60wt% PTFE carbon paper or carbon fabric.
- [4] The multi-layered electrode according to claim 1 or 2, wherein in the catalyst slurry, the catalyst 2 is 20~60wt% Pt/C, and the solution of the nafion ionomer 3 is a solution of the nafion ionomer 3 with EW of 1,000~1,300 which, in case that the total quantity of the nafion ionomer 3 of the catalyst layer and the nafion ionomer 4 of the coating layer amounts to 20~50% of the weight of Pt/C catalyst, has the amount of 1/12~4/12 of the total quantity of the nafion ionomer and is 5wt% or less.
- [5] The multi-layered electrode according to claim 4, wherein the solution of the nafion ionomer 4 is a solution of the nafion ionomer 4 with EW of 400~1,000 which has the remainder of the total quantity of the nafion ionomer excepting the quantity of the nafion ionomer 3 of the catalyst layer and is 5wt% or less.
- [6] A method for producing multi-layered electrode for fuel cell comprising the steps of:
(S1) preparing a catalyst slurry containing a catalyst and a solution of a nafion ionomer with a specific equivalent weight (EW);
(S2) applying the catalyst slurry onto a gas diffusing layer;
(S3) first-drying the catalyst slurry applied on the gas diffusing layer to form a catalyst layer;
(S4) applying on the surface of the dried catalyst layer a solution of a nafion ionomer with the lower EW than the specific EW of the nafion ionomer of the catalyst slurry;
(S5) second-drying the applied nafion ionomer solution to form an electrode; and
(S6) annealing the dried electrode.
- [7] The method according to claim 6, wherein in the step S1, the catalyst slurry is prepared by stirring 20~60wt% Pt/C catalyst; the nafion ionomer with EW of

1,000~1,300 which, in case that the total quantity of the nafion ionomer in the step S1 and the nafion ionomer in the step S4 amounts to 20~50% of the weight of Pt/C catalyst, has the amount of 1/12~4/12 of the total quantity of the nafion ionomer and is 5wt% or less; and IPA or EtOH solvent.

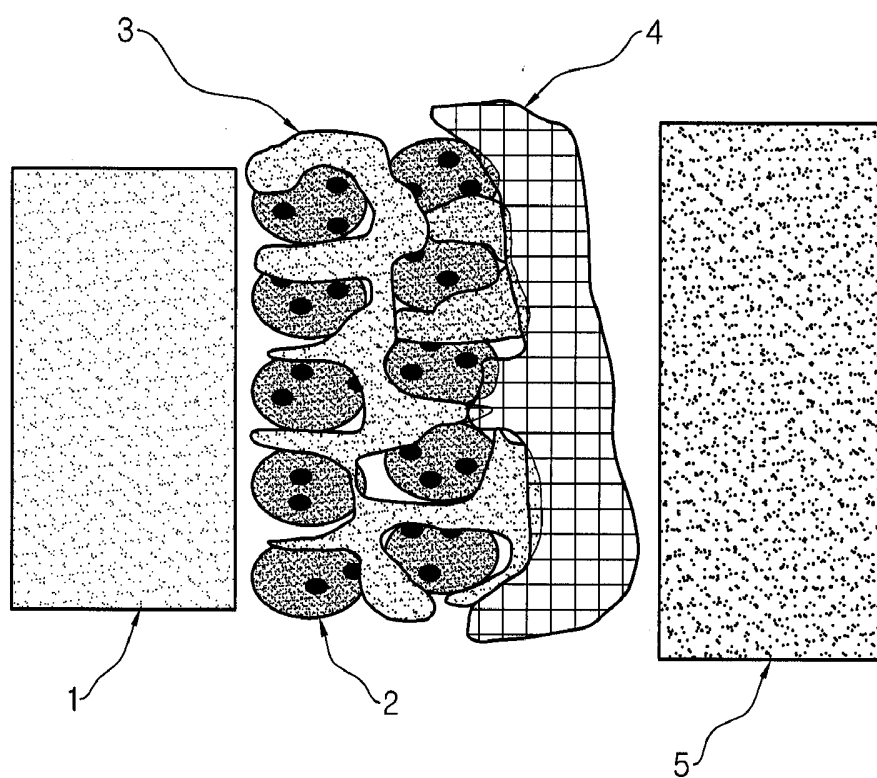
- [8] The method according to claim 7, wherein in the step S1, the catalyst slurry is prepared by stirring 20~40wt% Pt/C catalyst; the nafion ionomer with EW of 1,100~1,200 which has the amount of 1/12~2/12 of the total quantity of the nafion ionomer and is 5wt% or less; and IPA or EtOH solvent.
- [9] The method according to claim 7, wherein in the step S2, the catalyst slurry is applied in a loading quantity of 0.04~1.2mg-Pt/C/cm² onto the gas diffusing layer composed of 20~60wt% PTFE carbon paper or carbon fabric.
- [10] The method according to claim 9, wherein in the step S2, the catalyst slurry is applied in a loading quantity of 0.4~0.7mg-Pt/C/cm² onto the gas diffusing layer composed of 20~40wt% PTFE carbon paper or carbon fabric.
- [11] The method according to claim 9, wherein in the step S3, the drying is performed at a temperature of 60~80°C for 30 minutes to 24 hours.
- [12] The method according to claim 11, wherein in the step S3, the drying is performed at a temperature of 60~80°C for 1~2 hours.
- [13] The method according to claim 11, wherein in the step S4, the solution of the nafion ionomer is prepared by stirring the nafion ionomer with EW of 400~1,000 which has the remainder of the total quantity of the nafion ionomer excepting the quantity of the nafion ionomer in the step S1 and is 5wt% or less; and IPA or EtOH solvent.
- [14] The method according to claim 11, wherein in the step S4, in case that the catalyst slurry is prepared by stirring 20~40wt% Pt/C catalyst; the nafion ionomer with EW of 1,100~1,200 which has the amount of 1/12~2/12 of the total quantity of the nafion ionomer and is 5wt% or less; and IPA or EtOH solvent in the step S1, the solution of the nafion ionomer is prepared by stirring the nafion ionomer with EW of 1,100~1,200 which has the remainder of the total quantity of the nafion ionomer excepting the quantity of the nafion ionomer in the step S1 and is 5wt% or less; and IPA or EtOH solvent.
- [15] The method according to claim 13, wherein in the step S5, the drying is performed at a temperature of 60~80°C for 30 minutes to 24 hours after applying the nafion ionomer solution.
- [16] The method according to claim 15, wherein in the step S5, the drying is performed at a temperature of 60~80°C for 1~2 hours after applying the nafion ionomer solution.
- [17] The method according to claim 15, wherein in the step S6, the annealing is

- performed in a vacuum oven at a temperature of 120~170°C for 0.5~24 hours.
- [18] The method according to claim 17, wherein in the step S6, the annealing is performed at a temperature of 150~170°C for 6~18 hours.

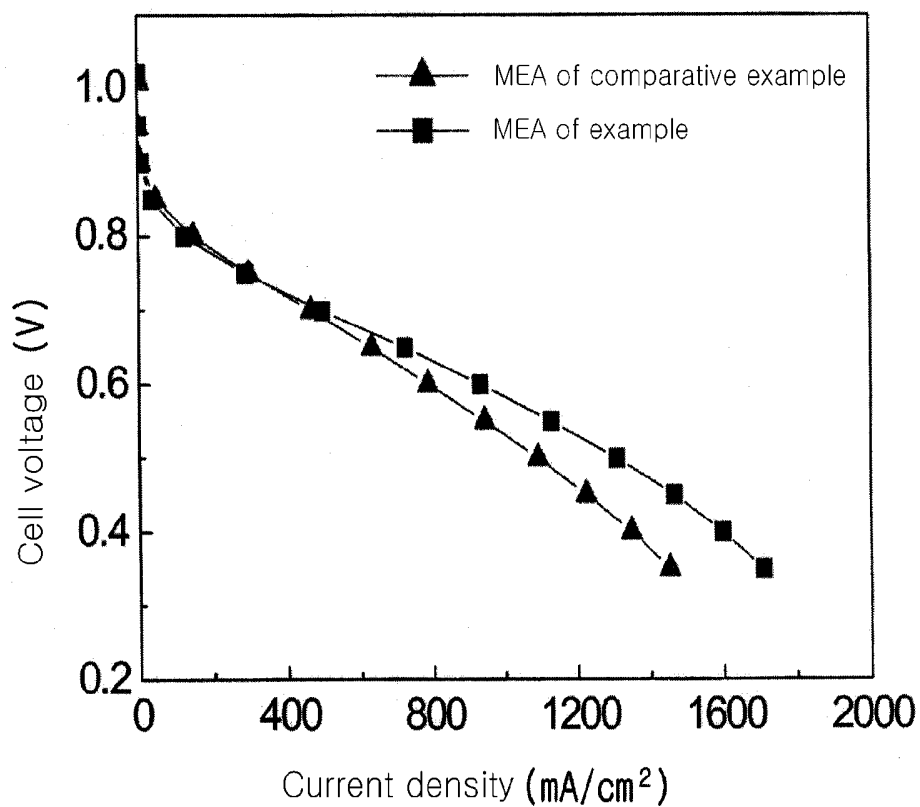
[Fig. 1]



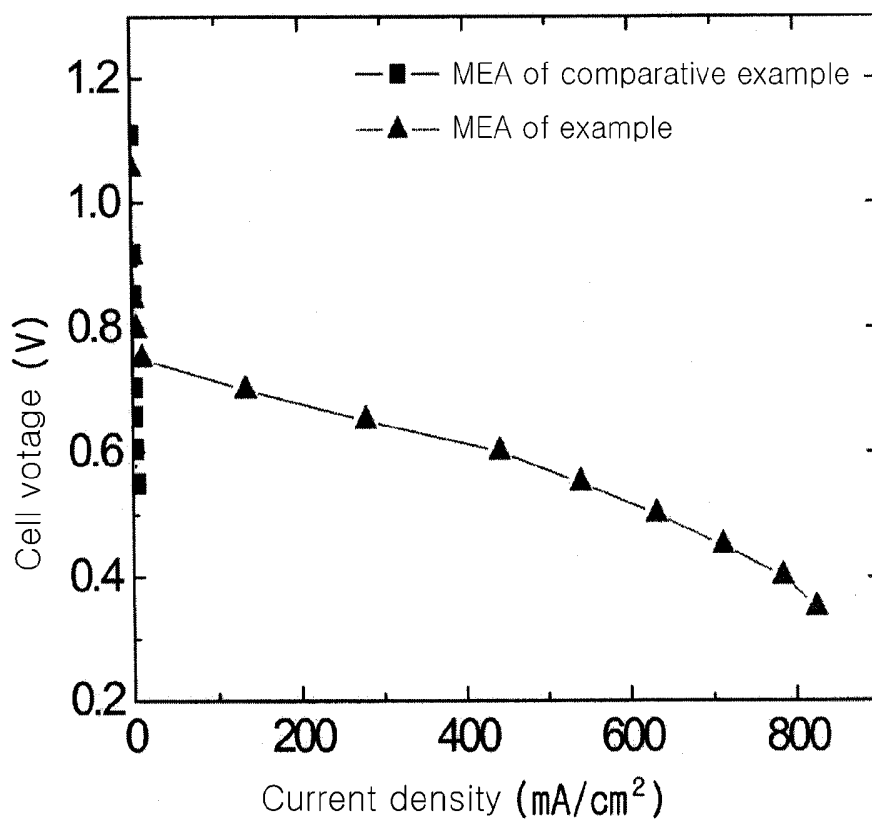
[Fig. 2]



[Fig. 3]



[Fig. 4]



PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference OF05P323	FOR FURTHER ACTION see Form PCT/ISA/220 as well as, where applicable, item 5 below.	
International application No. PCT/KR2005/003635	International filing date (<i>day/month/year</i>) 31 OCTOBER 2005 (31.10.2005)	(Earliest) Priority Date (<i>day/month/year</i>) 27 JANUARY 2005 (27.01.2005)
Applicant Korea Institute of Science and Technology et al		

This International search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 3 sheets.

☐ It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

- a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ The international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, see Box No. I.

2. ☐ **Certain claims were found unsearchable** (See Box No. II)

3. ☐ **Unity of invention is lacking** (See Box No. III)

4. With regard to the **title**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. With regard to the **drawings**,

- a. the figure of the **drawings** to be published with the abstract is Figure No. 1

☒ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

- b. ☐ none of the figure is to be published with the abstract.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2005/003635**A. CLASSIFICATION OF SUBJECT MATTER****H01M 4/86(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)

IPC8 H01M

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

Korean Patents and applications for inventions since 1975

Korean Utility models and applications for Utility models since 1975

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

KR : IPC as above

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 6,187,467 B1 (Juijun Zhang, Richmond) 13 February 2001 See abstract, claim1 and Figure1	1-18
A	US 6,749,892 B2 (Hyuk Chang) 15 June 2004 See abstract, claim1 and Figure1	1-18
A	US 6,696,382 B1 (Piotr Zelenay) 24 February 2004 See abstract, claim1 and Figure1	1-18



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

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of citation or other special reason (as specified)

"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

"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

"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

"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

"&" document member of the same patent family

Date of the actual completion of the international search

02 MARCH 2006 (02.03.2006)

Date of mailing of the international search report

02 MARCH 2006 (02.03.2006)

Name and mailing address of the ISA/KR

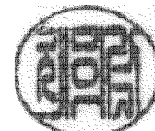
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Authorized officer

CHAE, Heui Gak

Telephone No. 82-42-481-5544



INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/KR2005/003635

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 6,187,467 B1	13. 02. 2001	US 17384598	16. 10. 1998
		US 22250798	29. 12. 1998
US 6,749,892 B2	15. 06. 2004	NONE	
US 6,696,382 B1	24. 02. 2004	NONE	