(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
11 April 2019 (11.04.2019)

WIPO | PCT

(10) International Publication Number
WO 2019/069095 A1

(51) International Patent Classification:
H01M 8/1004 (2016.01) *H01M 8/1065* (2016.01)
H01M 4/86 (2006.01) *C25B 11/04* (2006.01)
H01M 8/1018 (2016.01)

(21) International Application Number:
PCT/GB2018/052854

(22) International Filing Date:
05 October 2018 (05.10.2018)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
1716428.6 06 October 2017 (06.10.2017) GB

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(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JO, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

(54) Title: FUEL CELL OR ELECTROLYSER ASSEMBLY

(57) Abstract: A membrane electrode assembly (8), suitable for use in a fuel cell or electrolyser, comprising: - an anion exchange membrane (4); - two catalyst containing layers (3, 5) each disposed either side of the anion exchange membrane (4), and; - two gas diffusion layers (2, 6), each in contact with one of said catalyst containing layers 3, 5), - wherein the anion exchange membrane (4) comprises a solid state electrolyte, - at least one catalyst containing layer (3, 5) comprises particulates of the solid state electrolyte material of the anion exchange membrane (4).

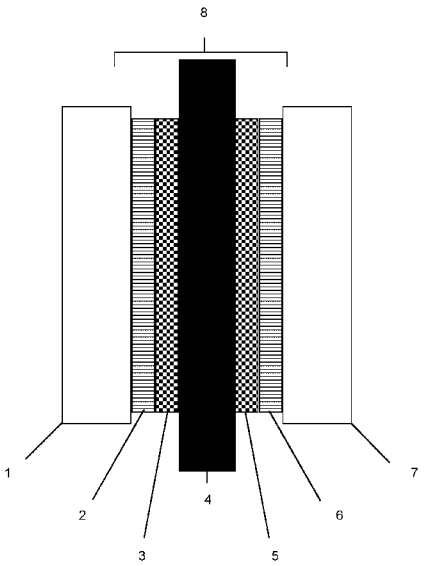

Fig. 1

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[Continued on next page]

Declarations under Rule 4.17:

— *as to the identity of the inventor (Rule 4.17(i))*
— *as to applicant's entitlement to apply for and be granted a patent (Rule 4.17(ii))*
— *as to the applicant's entitlement to claim the priority of the earlier application (Rule 4.17(iii))*

Published:

— *with international search report (Art. 21(3))*

FUEL CELL OR ELECTROLYSER ASSEMBLY

The present invention relates to Anion Exchange Membrane (AEM) fuel cells and/or electrolysers, electrode assemblies of such fuel cells and/or electrolysers, and to catalyst containing layers of such electrode assemblies.

INTRODUCTION

Fuel cells have been identified as a relatively clean and efficient source of electrical power. Alkaline Fuel Cells (AFCs) are of interest because they operate at relatively low temperatures, are efficient and mechanically and electrochemically durable.

Anion Exchange Membrane fuel cells (AEMFCs) are functionally similar to AFCs but employ a solid electrolyte, whereas AFCs use aqueous potassium hydroxide as an electrolyte. AEMFCs are of particular interest because, among other advantages, they are less prone to carbonate precipitation on electrodes, and, in alkaline environments, can potentially allow the use of relatively inexpensive, non-noble metal catalysts, such as silver or iron phthalocyanines for the cathode and nickel for the anode, due to the more facile oxygen reduction reaction (ORR) kinetics at the cathode.

Such AEMFCs comprise electrode assemblies familiar to those skilled in the art; a solid electrolyte Anion Exchange Membrane (AEM) with a catalyst containing layer on either side of the AEM, and further Gas Diffusion Layers (GDLs) applied on either side over the catalyst containing layers. The catalyst containing layer may be a layer discrete from the GDL layer (albeit in intimate contact), comprising catalyst, carbon and a hydrophobic material. In a known alternative, the catalyst layer on each side of the AEM may simply be a coating of electrocatalyst material on a face of the GDL layer. GDLs are typically composed of porous materials comprising a dense array of carbon fibres in the form of a cloth or paper, or alternatively may comprise or be composed of a metallic material, and provide an electrically conductive pathway for current collection. In some embodiments they also comprise a hydrophobic material which may also serve the function of a binder.

One of the challenges of AEMFCs is achieving OH- ion conductivity comparable to H+ conductivity observed in (Proton Exchange Membrane Fuel Cells (PEMFCs).

It is therefore beneficial to provide improvements to AEMFCs which address or mitigate problems in the prior art.

Surprisingly, we have found that by dispersing a solid electrolyte on or through the catalyst containing layer, in particular in the form of an interpenetrating network of solid electrolyte particles, an alkaline fuel cell or electrolyser with improved results can be achieved.

SUMMARY OF THE INVENTION

Accordingly, in a first aspect there is provided, a membrane electrode assembly suitable for use in a fuel cell, comprising:

- an anion exchange membrane;
- two catalyst containing layers each disposed either side of the anion exchange membrane; and
- two gas diffusion layers, each in contact with one of said catalyst containing layers;

wherein the anion exchange membrane comprises a solid state electrolyte, and at least one catalyst containing layer comprises particulates of the solid state electrolyte material of the anion exchange membrane.

The presence of electrolyte in the catalyst containing layer provides for anion exchange capacity deep into the catalyst containing layer, improving OH- ion conductivity in the electrode. This should increase the power performance of a fuel cell or decrease the amount of power required for electrolysis (the devices become more efficient).

Further, where particulates of a solid state electrolyte are present within the catalyst layer, the similarity of materials further provides for enhanced ionic conductivity into and/or through the catalyst containing layer.

Further, it will be appreciated that in traditional AFCs, KOH is present as a liquid electrolyte both in the membrane layer and in the catalyst containing layers. Advantageously, the invention enables the use of a fuel cell or electrolyser without liquid KOH present. Much previous work has been performed on ways to mitigate the corrosive and reactive effects of liquid KOH in fuel cells. Here, where solid state electrolyte is used both as the AEM and is also 'infused' into the catalyst containing layer, the requirement

to use liquid KOH is beneficially avoided.

In an embodiment, particulates of solid state electrolyte are distributed on or through the catalyst containing layer.

In an embodiment, at least one catalyst containing layer comprises particulates of the solid state electrolyte material of the anion exchange membrane.

Where the solid state electrolyte particulates are distributed on or through the catalyst containing layer, this maximises the opportunity for good ionic conductivity. In an embodiment, this effect is improved further by disposing the particulate material within the catalyst layer such that an interpenetrating network of electrolyte material is present. In this way, channels of ionic conductivity are provided deep into the catalyst layer, maximising contact with the catalyst particles themselves, and so maximising performance.

In an embodiment, the catalyst containing layer would be of the order of 500μm thick, or thinner, and the particulates of solid state electrolyte that are embedded within the catalyst would be in the size range 10μm to 250μm. Suitable ranges may also include, in some embodiments, 10μm to 50μm, 10μm to 100μm, 10μm to 150μm, 5μm to 50μm, 5μm to 100μm, 5μm to 150μm, 5μm to 250μm, 50μm to 100μm, 50μm to 150μm, 50μm to 200μm, or 50μm to 250μm, for example.

In an embodiment, the particulates of anion exchange material embedded in the catalyst layer may comprise whiskers or tubes of anion exchange material. Ionic conductivity is improved by the presence of these types of particulates.

In an embodiment, the catalyst containing layer further comprises an electro-catalyst. Such electro-catalysts may comprise platinum, palladium, silver, nickel, or alloys thereof. Ceramic and carbon based catalysts are also available.

In embodiments, the catalyst containing layer may contain fullerene or fullerene based materials, such as carbon nanotubes or graphene, or other carbon materials such as carbon black. Further, the catalyst containing layer may comprise a fluid-permeable hydrophobic material such as PTFE. Another suitable material is polyvinylidene fluoride (PVdF), which may be used instead of or in addition to the PTFE. Another possibility is a

perfluoroalkoxy (PFA) polymer or copolymer. A fluorinated ethylene/propylene copolymer (FEP) or an ethylene/tetrafluoroethylene copolymer (ETFE) are other options.

It will be appreciated that fuel cells and electrolysers are often constructed in fuel cell stacks, where membrane electrode assemblies (MEAs) are sandwiched between pairs of bipolar plates. These plates connect and separate individual MEAs, conduct electrical current from one MEA to the next, provide physical support, facilitate the distribution of fuel and oxidant to the MEA, facilitate water management, and additionally may provide clamping forces to press the various layers of MEAs together. In an embodiment, therefore, the MEA of the present invention is disposed between a pair of bipolar plates. Aspects and embodiments of the present invention may advantageously require less clamping force to be applied across the MEAs; this being the case, the bipolar plates in a fuel cell or electrolyser, or fuel cell or electrolyser stack, comprising MEAs in accordance with aspects and embodiments as described herein may be thinner and lighter than might otherwise be required. This is advantageous in terms of both potential weight saving in a fuel cell or electrolyser device and also in terms of material cost.

A fuel cell stack comprises multiple fuel cells arranged as a stack, in order to provide a higher output power. Such a stack may include between two cells and two hundred cells, more typically between eight cells and one hundred cells.

In an aspect there is provided a fuel cell comprising a membrane electrode assembly as described in other aspects and embodiments.

In an aspect there is provided a fuel cell stack comprising a membrane electrode assembly as described in other aspects and embodiments.

It will be recognised by the skilled person that fuel cells in principle have the capacity to run in reverse mode and perform electrolysis. Such a device is often termed a regenerative fuel cell or is simply known as an electrolyser. The present membrane electrode assembly is suitable for use in such an electrolyser as well as in a fuel cell.

An electrolyser stack comprises multiple electrolyser cells arranged as a stack, in order to produce a greater rate of oxygen / hydrogen production. Such a stack may include between two cells and two hundred cells, more typically between eight cells and one hundred cells.

Accordingly, in an aspect there is provided an electrolyser comprising a membrane electrode assembly as described in other aspects and embodiments.

There is further provided, in an aspect, an electrolyser stack comprising an electrolyser or a membrane electrode assembly as described in other aspects and embodiments.

In another aspect, there is provided a method of manufacturing the present assembly, the method comprising:

- providing a GDL and affixing the catalyst-containing layer to provide a GDL-catalyst layer;
- affixing the GDL-catalyst layer to first and second sides of the AEM so that the catalyst containing layer contacts the AEM;
- and optionally pressing one or more bipolar plates to the GDL layers.

In another aspect, there is provided a method of manufacturing the present assembly, the method comprising:

- providing an AEM and applying or affixing a catalyst-containing layer to first and second sides of the AEM to provide a catalyst-AEM-catalyst layer;
- applying or affixing a GDL to the catalyst-containing layers;
- and optionally pressing one or more bipolar plates to the GDL layers.

BRIEF DESCRIPTION OF THE DRAWINGS

The invention will now be further and more particularly described, by way of example only, and with reference to the accompanying drawings, in which;

Figure 1 shows a diagrammatic cross-sectional view through one cell of a fuel cell or electrolyser stack comprising an Anion exchange membrane electrode assembly in accordance with the invention. The figure shows:

1. Bipolar plate
2. Gas diffusion layer
3. Catalyst containing layer
4. Anion exchange membrane

5. Catalyst containing layer
6. Gas diffusion layer
7. Bipolar plate
8. Membrane electrode assembly

Figure 2 shows a catalyst layer structure according to an aspect of the invention. Figures 2a, 2b, 2c and 2d show diagrammatic cross-sectional partial views through a portion of the catalyst layer (5) in accordance with various embodiments of the invention. The figures show:

5. Catalyst containing layer
100. Particulates of solid state electrolyte material
101. Whiskers of particulates
102. Tubes of particulates

Figure 3 shows a representative graph indicating expected test results of a fuel cell constructed in an analogous manner to a fuel cell in accordance with embodiments described herein. The Figure shows:

30. Typical set of polarisation curves and the corresponding power curves
31. Potential axis
32. Current Density Axis
33. Power Density axis
34, 35, 36. Polarisation curves
37, 38, 39. Power curves

Figure 4 shows a diagram of a test cell as is used to produce the results of the test cell in Figure 3. The Figure shows:

1. Bipolar plate
2. Gas diffusion layer
4. Anion exchange membrane
6. Gas diffusion layer
7. Bipolar plate
40. Anode catalyst layer
41. Cathode catalyst layer
42. Hydrogen inlet
43. Hydrogen outlet

44. Oxygen / air inlet
45. Oxygen/ air outlet and water outlet

Figure 5 shows a cross-sectional view through the structural components of a cell for a fuel cell comprising an assembly in accordance with the invention as may be used in a fuel cell or electrolyser, with the components separated for clarity. Figure 1 of Patent publication GB2508649 shows a typical arrangement of a fuel cell stack, and it will be apparent that the arrangement of components in such a stack may be modified in accordance with the arrangement shown in Figure 5. Further, the person skilled in the art will recognise the changes required to run such an assembly as an electrolyser.

DETAILED DESCRIPTION

Referring to Figure 1, a fuel cell comprises a membrane electrode assembly (8) between two bipolar plates (1, 7). At the centre of the membrane electrode assembly is a solid state anion exchange membrane (4). On either side of the anion exchange membrane is a catalyst layer (3, 5), and on the outside faces of the catalyst layers are gas diffusion layers (2, 6). All the layers are in intimate contact, and are pressed together by a clamping force (not shown).

Typical materials for a catalyst layer include carbon, with a hydrophobic binder which may be polytetrafluoroethylene (PTFE), and an appropriate active catalytic material in particulate form.

Typical materials for a gas diffusion layer include a carbon material such as carbon paper or carbon cloth. The carbon is often mixed with a hydrophobic binder which may be polytetrafluoroethylene (PTFE).

Referring to Figure 2a, there is shown a catalyst layer (5) on which particulates (100) are deposited.

Referring to Figure 2b, there is shown a catalyst layer (5) in which particulates (100) are distributed through the catalyst containing layer. In this instance, the penetration of the particles through the catalyst containing layer is incomplete.

Referring to Figure 2c, there is shown a catalyst layer (5) in which particulates (100) are distributed through the catalyst containing layer. In this Figure, the penetration of the particles through the catalyst containing layer is complete and they form an interpenetrating network.

Referring to Figure 2d, there is shown a catalyst layer (5) in which particulates (100) are distributed throughout the catalyst containing layer and comprise whiskers (101) and tubes (102).

Figures 3 and 4 are described in more detail in the following Example.

Referring now to Figure 5 there is shown a cross-sectional view through the structural components of a cell with the components separated for clarity. The Membrane Electrode Assembly (8) is shown, which is as essentially described in relation to Figure 1 and comprises GDL (2), catalyst containing layer (3), Anion Exchange Membrane (4), catalyst containing layer (5) and GDL (6). The Bipolar plates (1, 7) are shown here in more detail; each defines rectangular blind recesses (21, 27) on the inner face to act as gas chambers, surrounded by a frame comprising a shallow (22, 28) surrounding each gas chamber.

Accordingly, when the assembly is clamped together, the GDL and catalyst layers (2, 3 and 5, 6) locate in the shallow frame recesses of each bipolar plate, with the GDL layers (2, 6) facing the gas chambers and the catalyst-containing layers (3, 5) facing the Anion Exchange Membrane (4).

Before assembly of the cell, the opposing surfaces of the bipolar plates are provided with a resilient sealing element (25). The sealing element ensures that gases cannot leak out of the gas chambers and so remain on their respective face of the Membrane Electrode Assembly. The gases are supplied into the gas chambers by means of ducts through the bodies of the bipolar plates (not shown) in accordance with means known in the art, or as shown in principle in Figure 4. Outlets are also provided for unused reactants and/or reaction products.

After assembly, the components are secured together using for example a strap, a clamp or bolts, as known in the art.

It will be appreciated that a cell as shown in Figure 5 may be run as a fuel cell or part of a fuel cell stack, or alternatively as an electrolyser or as part of an electrolyser stack.

EXAMPLE

By way of demonstrating the principle of the invention, a fuel cell test system (Figure 4) may be set up in the following manner.

A solid Anion Exchange Membrane (4) prepared by soaking in a 1Mol KOH solution. An AEM such as a FUMASEP® product from FUMATECH™ would be a suitable representative test subject that would be familiar to a person skilled in the art.

Each catalyst-containing layer (3, 40, 5, 41) and GDL layer (2, 6) pair are affixed together ensuring intimate contact between the layers and providing GDL-catalyst layers.

A test cell is constructed from the soaked AEM, with the catalyst containing layers and GDL layers, in the form of GDL-catalyst layers, placed either side of the AEM such that the catalyst containing layers are in contact with each side of the AEM.

The whole Anion Exchange Membrane Electrode assembly (GDL, catalyst containing layer, soaked AEM, catalyst containing layer, GDL) is in turn pressed between a pair of bipolar plates (1, 7) which are clamped together with a clamping force of, for example, about 5.5 Nm torque.

The contact between the soaked AEM and catalyst layers, and the clamping force supplied, ensures that the KOH solution is partially squeezed out of the AEM and absorbed by the catalyst containing layers. This provides a cell demonstrative of the inventive concept in that the catalyst containing layers are thus provided with a like electrolyte and thus good ionic connectivity between the AEM and the catalyst containing layers.

The fuel cell temperature is controlled at 40°C or 60°C, and supplied with pure H_2 (42) into one GDL, via an inlet at the anode side (3, 40), and pure O_2 (44) into the other GDL via an inlet at the cathode side (5, 41), with flow rates of 1L/min for both gases, with no back-pressure, and both gases at 100% Relative Humidity (RH). The temperatures of the

gases are controlled at either 36°C, 38°C or 60°C. Outlets are provided to allow for an outlet of hydrogen (43) from the anode side and an outlet of oxygen and water (45).

Polarisation curves (34, 35, 36) are obtained by scanning the voltage and measuring the potentiodynamic current in order to obtain indicative power densities (37, 38, 39).

Figure 3 shows an example of a typical set of power curves as expected to be obtained by these tests, where:
Graph 30 comprises three axes; a Potential axis 31 measuring Volts, a Current Density Axis 32 measuring milliamps per cm^2, and a Power Density axis 33 measuring milliwatts per cm^2.

A number of tests of the cell are run at various times during the lifecycle of the cell, and these are illustratively represented by lines 34, 35, 36, 37, 38, and 39 of Figure 3.

In a first test, a forward voltage scan as represented by line 34, at a cell temperature of 40°, a hydrogen temperature of 36°, and an oxygen temperature of 36°, is performed, and a result for power density is obtained as represented by line 37.

In a second test, a forward voltage scan as represented by line 35, at a cell temperature of 40°, a hydrogen temperature of 38° and an oxygen temperature of 38° is performed and a result for power density is obtained as represented by line 38.

In a third test, a forward voltage scan as represented by line 36, at a cell temperature of 60°, a hydrogen temperature of 60° and an oxygen temperature of 60° is performed and a result for power density is obtained as represented by line 39.

As can be seen, over the course of these tests, a continuous drop in performance is observed. At least part of this degradation is anticipated as being due to KOH being 'flushed' from the AEM and catalyst containing layers by water that is produced as a result of the reaction, as well as water from the humid oxygen and hydrogen gas streams.

Thus, the positive effect of the presence of KOH in the catalyst containing layers as well as the AEM is indicated by the higher peak power obtained in the first test (power density line 37) as opposed to the third test (power density line 39). This is seen as analogous to the power density improvement that can be expected from a cell constructed in

accordance with aspects and embodiments described in the present specification, where for example the anion exchange membrane comprises a solid state electrolyte, and the catalyst containing layers comprise particulates of the solid state electrolyte material of the anion exchange membrane.

In order to run a cell as illustrated in Figure 4 as an electrolyser, it will be appreciated that various changes will be required, including but not limited to:

- water being supplied to the cathode side (44, 41);
- a means of collecting hydrogen at the cathode (41)
- a means of collecting oxygen at the anode (40).

CLAIMS

1. A membrane electrode assembly, suitable for use in a fuel cell or electrolyser, comprising:
- an anion exchange membrane;
- two catalyst containing layers each disposed either side of the anion exchange membrane, and;
- two gas diffusion layers, each in contact with one of said catalyst containing layers,
wherein the anion exchange membrane comprises a solid state electrolyte and at least one catalyst containing layer comprises particulates of the solid state electrolyte material of the anion exchange membrane.

2. A membrane electrode assembly as claimed in claim 1, wherein the particulates are distributed on or through the catalyst containing layer.

3. A membrane electrode assembly as claimed in claim 1 or 2, wherein the particulate distribution provides an interpenetrating network of electrolyte material.

4. A membrane electrode assembly as claimed in any one of claims 1, 2, or 3, wherein the catalyst containing layer is of thickness of 500μm or less and the particulate size is in the range 10μm to 250μm.

5. An assembly as claimed in any one of the preceding claims, in which the particulate material comprises whiskers or tubes of anion exchange membrane material.

6. An assembly as claimed in any previous claim wherein the catalyst containing layer further comprises an electro-catalyst.

7. An assembly as claimed in any previous claim wherein the catalyst containing layer further comprises a fullerene or fullerene-based material or carbon black.

8. An assembly as claimed in any of the previous claims wherein the catalyst containing layer further comprises a fluid-permeable hydrophobic material.

9. An assembly as claimed in any previous claim wherein the assembly is disposed between a pair of bipolar plates, wherein each plate is in contact with one of the two gas diffusion layers.

10. A fuel cell comprising an assembly as claimed in any one of the preceding claims.

11. A fuel cell stack comprising a fuel cell as claimed in claim 10.

12. An electrolyser comprising an assembly as claimed in any one of claims 1 to 9.

13. An electrolyser stack comprising an electrolyser as claimed in claim 12.

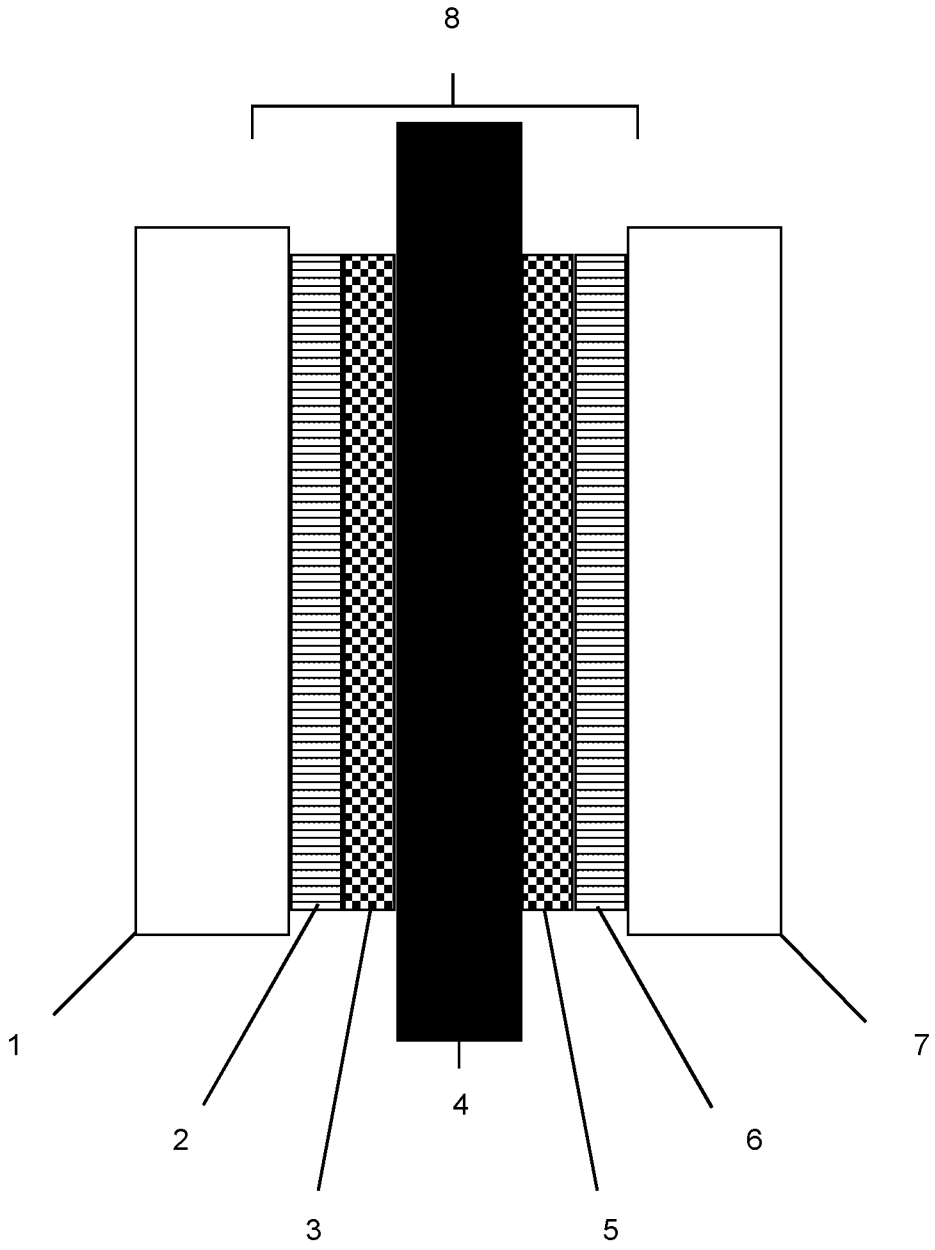

Fig. 1

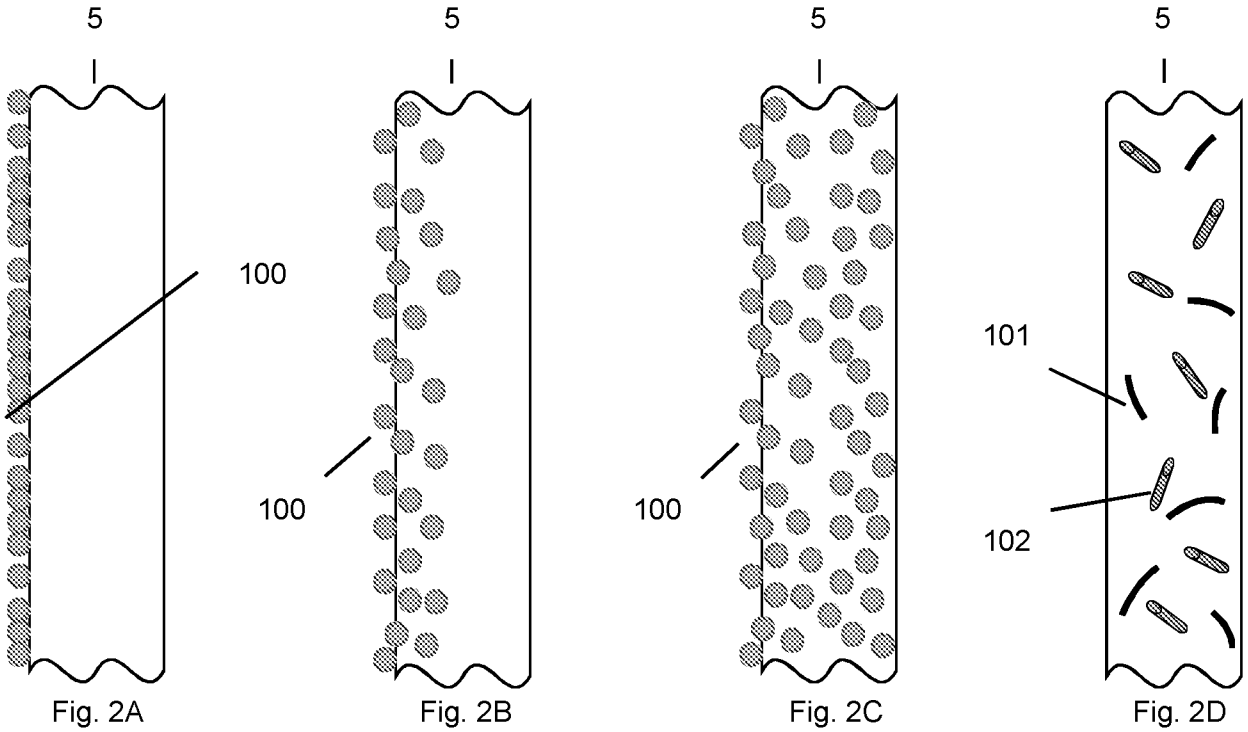

Fig. 2

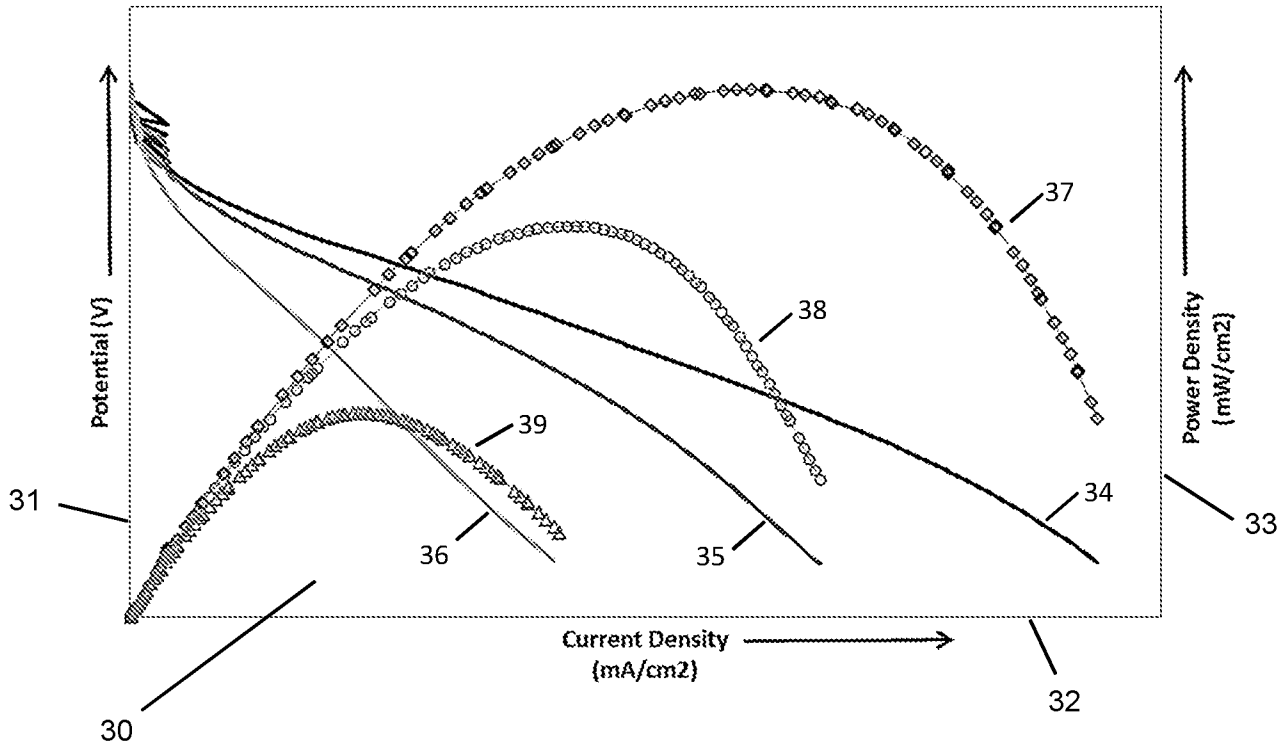
Potential (V)
Power Density (mW/cm2)
Current Density (mA/cm2)
31
30
32
33
34
35
36
37
38
39

Fig. 3

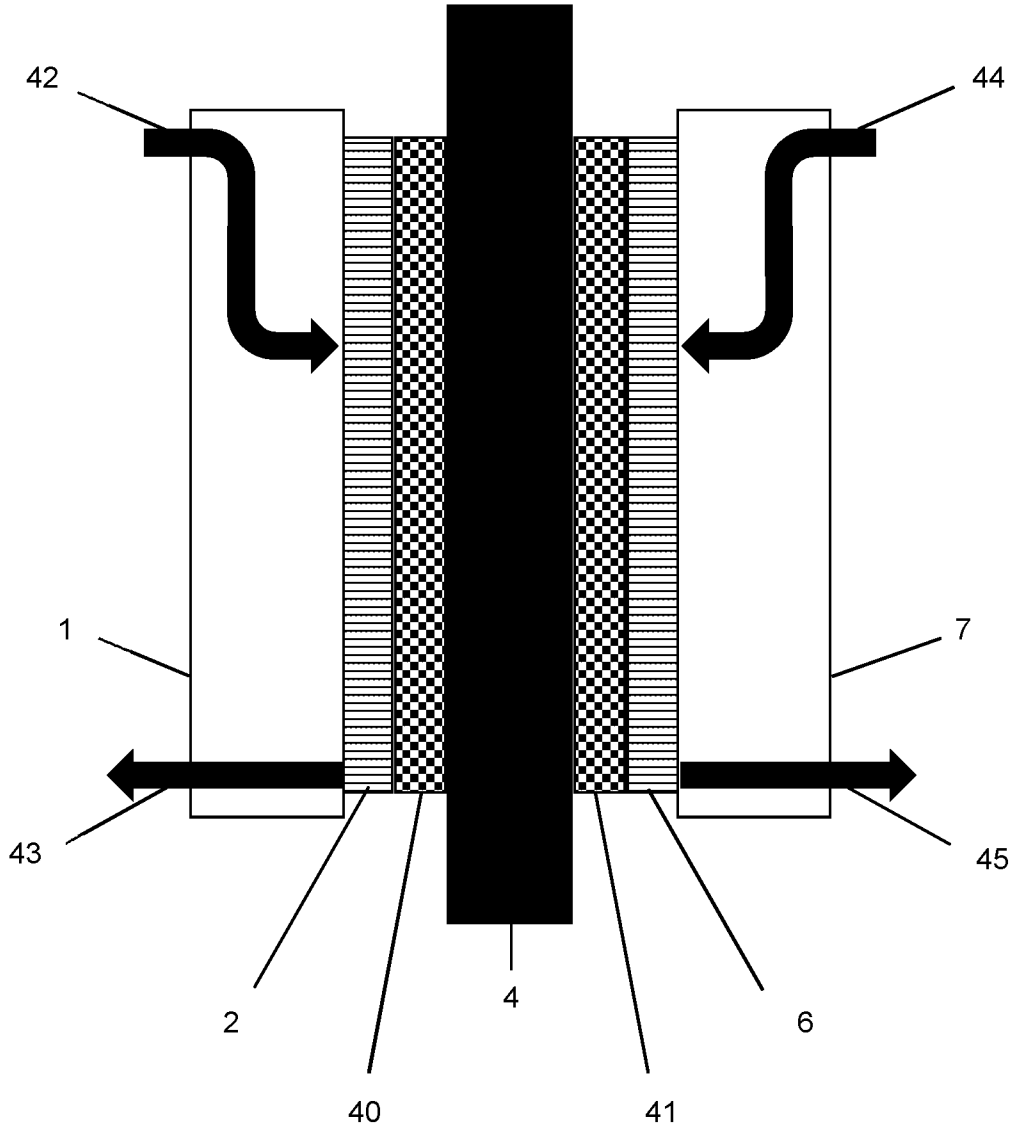

Fig. 4

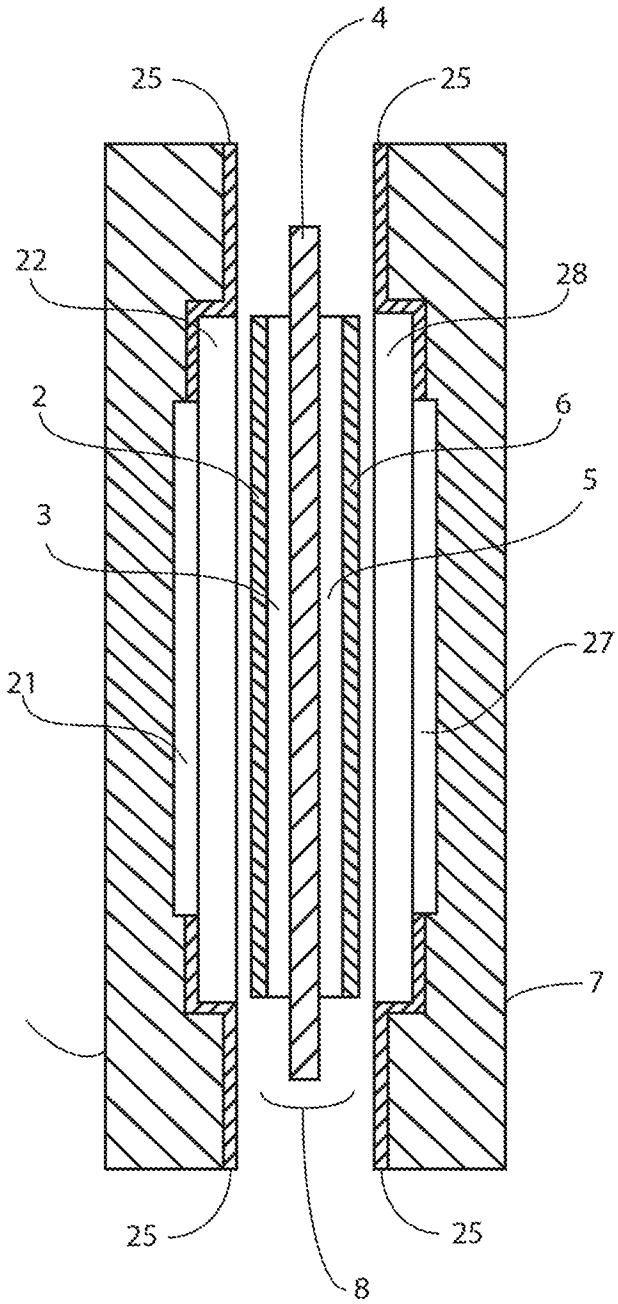

Fig. 5

INTERNATIONAL SEARCH REPORT

International application No
PCT/GB2018/052854

A. CLASSIFICATION OF SUBJECT MATTER
INV. H01M8/1004 H01M4/86
ADD. H01M8/1018 H01M8/1065 C25B11/04

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)
H01M C25B

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

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2015/099207 A1 (YAMAGUCHI TAKEO [JP] ET AL) 9 April 2015 (2015-04-09)	1-3,5-11
A	figures 2,3 paragraphs [0069] - [0071], [0150]	4,12,13
X	EP 2 876 713 A1 (TOKUYAMA CORP [JP]) 27 May 2015 (2015-05-27)	1-3,6-13
A	paragraphs [0020] - [0023], [0040] - [0045] paragraphs [0061] - [0065] paragraphs [0093], [0094]	4,5
A	US 2015/349368 A1 (ARGES CHRISTOPHER G [US] ET AL) 3 December 2015 (2015-12-03) paragraphs [0011] - [0022]	1,10-13

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[X] Further documents are listed in the continuation of Box C.

[X] 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

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"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

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Date of the actual completion of the international search	Date of mailing of the international search report
30 November 2018	13/12/2018

Name and mailing address of the ISA/	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Jacquinot, Patrick

1

Form PCT/ISA/210 (second sheet) (April 2005)

INTERNATIONAL SEARCH REPORT

International application No
PCT/GB2018/052854

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 2006/257641 A1 (CHO SUNG-YONG [KR] ET AL) 16 November 2006 (2006-11-16) paragraphs [0031], [0035] claims 5,8 -----	7

INTERNATIONAL SEARCH REPORT

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