

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

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
17 January 2002 (17.01.2002)

PCT

(10) International Publication Number
WO 02/05373 A1

(51) International Patent Classification⁷: **H01M 8/02**,
8/04, 8/10

[SE/SE]; Kungshamra 41-215, S-170 70 Solna (SE).
JAOUEN, Frédéric [SE/SE]; Kungshamra 41-312, S-170
70 Solna (SE).

(21) International Application Number: PCT/SE01/01514

(74) Agent: **FRÖHLING, Werner**; Volvo Teknisk Utveckling
AB, Patent Department 06820, CTP, S-412 88 Göteborg
(SE).

(22) International Filing Date: 29 June 2001 (29.06.2001)

(81) Designated States (national): AE, AG, AL, AM, AT, AU,
AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU,
CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM,
HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK,
LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX,
MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL,
TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

(25) Filing Language: English

(84) Designated States (regional): ARIPO patent (GH, GM,
KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian
patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European
patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE,

(26) Publication Language: English

(30) Priority Data:
0002601-3 7 July 2000 (07.07.2000) SE

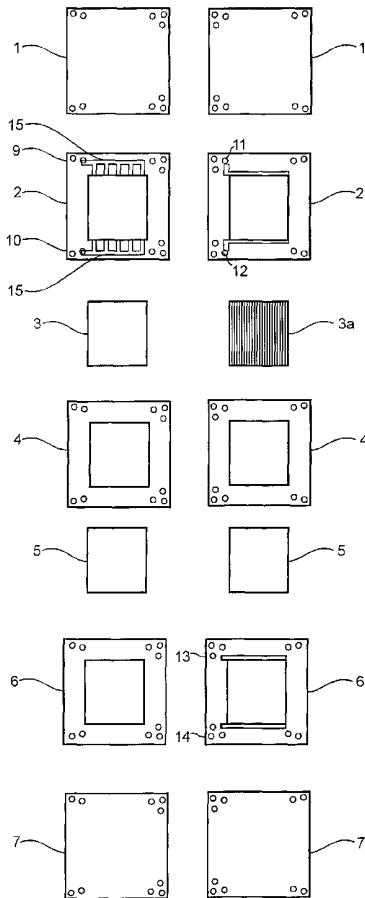
(71) Applicant (for all designated States except US): **AB
VOLVO** [SE/SE]; S-405 08 Göteborg (SE).

(72) Inventors; and

(75) Inventors/Applicants (for US only): **IHONEN, Jari**

[Continued on next page]

(54) Title: POLYMER FUEL CELL STRUCTURE



(57) Abstract: The invention discloses a polymer electrolyte fuel cell structure comprising a proton exchange membrane (4). An anode catalyst layer (1; 16) is located on one side of the proton exchange membrane. A cathode catalyst layer (7) is located on the opposite side of the proton exchange membrane, and a gas distribution layer (3, 5) is arranged on each side of the proton exchange membrane (4). The anode side gas distribution layer (3) is a flat, porous structure having water channels (3a) formed in the surface facing the membrane (4). The anode side gas distribution layer (3) is enclosed by a coplanar, sealing plate (2) with water inlet channels coupled to said water channels (3a) in the gas distribution layer.

WO 02/05373 A1



IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

Published:

- *with international search report*
- *before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments*

TITLE:

Polymer fuel cell structure

5

TECHNICAL FIELD

The present invention generally relates to fuel cells, and in particular to improvements in performance of polymer fuel cells.

10

BACKGROUND OF THE INVENTION

Polymer fuel cells are on the fringe of commercialization. The progress made in catalyst and membrane research in the last few years has enabled very high power densities (15) ($>1W/cm^2$) with moderate efficiencies for the fuel cell (40%). The catalyst loading of electrodes has been reduced to 0.1 mg Pt/cm² while maintaining a high performance. The price of the perfluorinated sulfonic acid membranes such as Nafion (R) is expected to decrease, with increasing (20) production, whilst other proton conducting membrane candidates have been discovered.

However, serious problems are encountered when polymer fuel cell technology is scaled up to larger cells and stacks.

(25) One of the main problems, in the stacks themselves, is the water management, since the proton conducting membrane must be kept well humidified under operating conditions.

The dominating component, at the internal resistance loss (30) in the stack, is due to the limiting proton conductivity of the membrane. Membranes tend to dry out, especially on the anode side, at high current densities, since proton migration drags water molecules away from the anode.

Drying of anode does not only affect resistance but also the kinetics of hydrogen reduction reaction (HRR) at the anode.

5 Therefore, in attempts to remedy this problem the anode side is often humidified more intensively than the cathode side. The cathode side of the cell can also be pressurized to use the pressure gradient over the membrane to press the water back to the anode. However, it is important that the
10 water management does not impede the gas flow inside the cells.

One solution for this problem would be to use thinner membranes, but this approach has limitations since
15 mechanical rigidity of the membrane must be sufficient.

Another solution is to have a direct water contact with the membrane at the anode side, since the water content and conductivity of the membrane are much higher when membrane
20 is in equilibrium with water. Also, when liquid is evaporated inside the fuel cell a considerable amount (40-50%) of the heat can be removed from the cell with the produced water vapor.

25 In US-5,958,613 (Hamada et al) relates to such direct water humidification of fuel cell membranes. Therein is disclosed a polymer fuel cell system with a capability to moisten the solid-polymer film without providing a special humidifier which humidifies the fuel gas or the oxidizer gas, and that
30 cools down the main cell body without providing cooling channels. In this patent there is no disclosure of specified operation principles for a fuel cell stack, when the direct humidification is applied.

In US-5,935,726 (Chow et al) there is disclosed a method and apparatus for improved humidification of membranes in polymer fuel cells, by periodically reversing the flow direction of the oxidant stream through a fuel cell flow field. However, this patent is not concerned with cooling of the fuel cell.

SUMMARY OF THE INVENTION

Despite the numerous attempts to improve the water management in polymer fuel cells, there is still room for improvements.

Thus, the object of the present invention is to provide means for achieving better humidification, at low cost and low cell complexity. The trade off between performance and cost should be acceptable.

In the cell structure of the present invention, an aqueous phase, preferably water, is used for direct humidification of the membrane. The polymer electrolyte fuel cell structure according to the invention comprises a proton exchange membrane, an anode catalyst layer on one side of the proton exchange membrane, a cathode catalyst layer on the opposite side of the proton exchange membrane and a gas distribution layer on each side of the proton exchange membrane. It is characterized in that the anode side gas distribution layer is a flat, porous structure having water channels formed in the surface facing the membrane, and that the anode side gas distribution layer is enclosed by a coplanar, sealing plate with water inlet channels coupled to said water channels in the gas distribution layer.

Thereby, it is possible to maintain a direct water contact with the membrane at the anode side. This is beneficial for

the operation of the cell, since the water content and the conductivity of the membrane are much higher when the membrane is in equilibrium with water. Also, when water is evaporated inside the fuel cell, a considerable amount of
5 the heat, produced in the cell, can be removed from the cell by means of the produced water vapor.

BRIEF DESCRIPTION OF THE DRAWINGS

The invention will now be further described in the
10 following, in a non-limiting way with reference to the accompanying drawings in which:

Fig. 1 is a perspective explosion view showing the elements of a polymer fuel cell structure in accordance with the invention,

15 Fig. 2 shows in plane view from above and below, all elements of the structure in Fig. 1,

Fig. 3 is a section in larger scale, through a gas distribution layer which is included in the structure of Fig. 1 and 2,

20 Fig. 4 shows a second embodiment of the gas distribution layer,

Fig. 5 shows a third embodiment of the gas distribution layer,

25 Fig. 6 shows, in the same manner as Fig. 2, an alternative structure layout, and

Fig. 7 shows still another embodiment of the gas distribution layer.

DETAILED DESCRIPTION OF THE INVENTION

30 A preferred embodiment of the fuel cell structure according to the invention is shown in figure 1 and 2. The fuel cell comprises a conductive anode plate 1. An anode sealing frame 2 is provided adjacent the bipolar plate 1. This frame is provided with a central, rectangular opening for

an anode gas distribution layer 3. The frame 2 is also provided with an anode gas inlet 9 and an outlet 10 and distribution channels are formed as well as water inlets and outlets 11, 12 respectively. The anode gas distribution 5 layer 3 is provided with a plurality of narrow water channels 3a on the opposite side of the layer 3, with reference to the anode plate 1. A proton exchange membrane 4 is arranged for cooperation with the plate 1 for sandwiching the frame 2 and the diffusion layer 3 between 10 themselves.

The cathode side of the fuel cell is structured in a similar manner as the anode side. Thus, the opposite side of the membrane 4 is arranged for cooperation with a 15 conductive cathode plate 7 for sandwiching a cathode sealing frame 6 and a cathode gas distribution layer 5 between themselves. The cathode diffusion layer 5 is not provided with any water channels as the anode diffusion layer 3. The cathode sealing frame 6 is provided with a 20 cathode gas inlet 13 and an outlet 14.

In figure 2 the detailed structure of water channels and how the water distribution is organized in a stack is shown. The left-hand side of the figure shows the upside 25 and the right-hand side of the figure shows the down side.

Each sealing frame 2 in a stack has a number of holes made through it. The holes located in the corners are for clamping bolts used when assembling a number of cell units 30 to a cell stack. The remaining holes, together with corresponding holes in the other components of a stack, form channels through the stack for water, fuel gas and oxidant gas respectively.

Furthermore, the upper side (as defined above) of the sealing 2 has gas channels 15 running along the inner edge of the frame like structure. A number of distribution apertures (in the figure there are five) are diverted from 5 each channel 15, so as to distribute incoming gas into the diffusion/distribution material located in the frame. The second hole from left (in the figure) in the upper array of holes is the inlet channel 9 for incoming gas, and the second hole from left in the lower array of holes is the 10 outlet channel 10 for gas exiting from the cell on the anode side. The anode sealing 2 has the same configuration of gas channels regardless of position in the stack.

On the down side (as defined above) of each sealing 2 there 15 are provided channels for water, having a common water inlet 11 and a common water outlet 12.

In the middle of the stack the membrane 4 is arranged, separating the anode and cathode parts of the stack. On the 20 cathode side, a cathode gas distribution layer 5 is provided, and then there is sealing 6 for cathode wherein cathode gas inlet and outlet 13, 14 are formed, in a similar way as in the anode sealing 2.

25 Figure 3 shows a more detailed structure of a gas distribution layer. The layer 3 is provided with water channels 3a adjacent the membrane 4. In a typical embodiment of the invention, the water channels 3a may have a width of about 50-100 μ m, a depth of about 100-300 μ m and 30 the channels may be separated by a distance of about 200-1000 μ m. By making the water channels narrow, blocking of the channels due to membrane expansion is avoided. One possible method of producing the channels 3a would be to press the gas distribution layer against a template having a ridge

structure surface corresponding to the desired water channel structure.

Figure 4 shows an embodiment of the invention, where the gas 5 distribution layer 3 is provided with a catalyst layer 16. A non-porous or almost non-porous proton conducting polymer layer 17 is arranged so that it lines the water channel.

In this embodiment, a hydrogen peroxide or other oxygen 10 evolving compounds may be added to the humidification and cooling water, which is fed into the cell on the anode side. Since the oxygen is released in the vicinity of the catalyst, CO adsorption at the anode catalyst may be avoided, in a manner which is effective and which leads to 15 less consumption of oxygen. Part of the hydrogen peroxide will be decomposed at the electrode surface to generate oxygen with the reaction $H_2O_2 \rightarrow H_2O + 1/2O_2$. In this system possible benefits of hydrogen peroxide are achieved even if the decomposition is not complete. The path of the hydrogen 20 peroxide and evolved oxygen is marked as arrows in Fig. 4. However, this method can be applied to other direct water humidification systems in polymer fuel cells.

Figure 5 shows a gas distribution layer 3, the edges of 25 which has been treated with a hydrophobic polymer to prevent the water from entering the cell gas chamber. In this structure there are no gas channels in the bipolar plates 1, 7 or in the gas distribution layers 3, 5. The gas distribution layers can have a porosity exceeding 90% and 30 they should be good electrical conductors and have proper corrosion resistance against acid proton conducting membrane.

The present invention may be combined with the conventional serpentine channel structure. The principle of this is illustrated in figure 6. The same reference numbers has been used as in the embodiment according to 5 Fig. 2. The anode layer side of each bipolar plate 1, 7 may be provided with an anode gas channel 19 and at least one water inlet 20. A water outlet 21 may also be provided. The cathode layer side of each bipolar plate is provided with at least one cathode gas channel 22.

10

An alternative structure for the water channels is presented in Fig. 7. In this embodiment of the invention, the catalyst layer 16 is located on top of the membrane 4. A hydrophobous layer 18 is positioned between the membrane and the gas 15 distribution layer. The function of this layer 18 is to let gas diffuse to the electrode (catalyst layer) but not let the water to escape from the water channel 3a.

The embodiment according to Fig. 7 may be used for operation 20 of a liquid-gas direct methanol fuel cell. In such an embodiment of the invention, the anode side of the cell is fed with a liquid water-methanol mixture, which is totally or partially evaporated in the cell. The liquid mixture is fed in such a way that most part of the anode electrode is 25 in contact with a thin film of liquid methanol-water mixture. The remaining area of the anode electrode is in contact with the gas phase free from liquid. This in order to enable both fast release of gaseous carbon dioxide as well as for humidifying of membrane by water vapor reactant 30 to remaining part of the anode area. Water and methanol are transferred from fuel feeding channels to the anode electrode both directly and via gas phase. This is illustrated by means of arrows in Figure 7. The above

described method may also be applied to other types of fuel cell structures which are direct liquid cooled.

5 The water channel structure is preferably applied to the anode side. However, this structure can also be applied to the cathode side or to both sides simultaneously.

10 The invention is not limited to the above described embodiments, instead several modifications are possible within the scope of the following patent claims.

CLAIMS

1. A polymer electrolyte fuel cell structure comprising:

5 a proton exchange membrane (4);

an anode catalyst layer (1; 16) on one side of the proton exchange membrane;

10 a cathode catalyst layer (7) on the opposite side of the proton exchange membrane;

15 a gas distribution layer (3, 5) on each side of the proton exchange membrane (4),

characterized in

that the anode side gas distribution layer (3) is a flat, porous structure having water channels (3a) formed in the 20 surface facing the membrane (4), and that

the anode side gas distribution layer (3) is enclosed by a coplanar, sealing plate (2) with water inlet channels coupled to said water channels (3a) in the gas distribution layer.

20

2. A structure according to claim 1,

characterized in

that the water channels (3a) have a width of about 50-100 μ m.

25

3. A structure according to claim 1 or 2,

characterized in

that the water channels (3a) have a depth of about 100-300 μ m.

30

4. A structure according to any one of claims 1-3,

characterized in

that the water channels (3a) are separated by a distance of about 200-1000 μ m.

5. A structure according to any one of claims 1-4,
characterized in
that the cathode side gas distribution layer (5) is a
porous structure which is enclosed by a coplanar, sealing
5 plate (6).

6. A structure according to claim 5,
characterized in
that the cathode side gas distribution layer (3) is
10 provided with water channels formed in the surface facing
the membrane (4), and that it is enclosed by a coplanar,
sealing plate (2) with water inlet channels coupled to said
water channels (3a) in the gas distribution layer.

15 7. A structure according to any one of claims 1-6,
characterized in
that the water channels (3a) are provided with a
hydrophobic coating.

20 8. A structure according to any one of claims 1-7,
characterized in
that a bipolar plate (1; 7) is located at the side of each
sealing plate (2; 6) which is distant from the membrane
(4).

25 9. A structure according to claim 8,
characterized in
that each bipolar plate (1, 7) is provided with the
anode catalyst layer on one side and the cathode
30 catalyst layer on the opposite side.

10. A structure according to claim 9,
characterized in

that the cathode layer side of each bipolar plate (1; 7) is provided with at least one cathode gas channel (22), and that the anode layer side is provided with an anode gas channel (19) and at least one water inlet (20).

5

11. A structure according to any one of claims 1-7, characterized in that the anode catalyst layer (16) is located on the anode side gas distribution layer (3).

10

12. A structure according to claim 11, characterized in that the cathode catalyst layer is located on the cathode side gas distribution layer (5).

15

13. A structure according to claim 11 or 12, characterized in that water is retained in the water channels (3a) by means of a comparatively non-porous layer (17) of a proton conducting material.

20

14. A structure according to any one of claims 1-7, characterized in that at least one of the catalyst layers (16) is located on the membrane (4).

25

15. A structure according to claim 14, characterized in that another catalyst layer is located on the gas distribution layer at the opposite side of membrane (4).

30

16. A structure according to claim 14 or 15, characterized in

that water is retained in the water channels (3a) by means of a comparatively non-porous layer (18) of a highly gas permeable material.

5 17. A method of operating a polymer electrolyte fuel cell comprising:

a proton exchange membrane (4);

an anode catalyst layer (1; 16) on one side of the proton exchange membrane;

10 a cathode catalyst layer (7) on the opposite side of the proton exchange membrane;

a gas distribution layer (3, 5) on each side of the proton exchange membrane (4), wherein a cooling and humidification liquid is fed to the anode side of the fuel cell,

15 characterized in
the step of adding an oxygen evolving compound to the liquid.

20 18. A method according to claim 17,

characterized in

that the oxygen evolving compound is hydrogen peroxide.

25 19. A method of operating a polymer electrolyte fuel cell comprising:

a proton exchange membrane (4);

an anode catalyst layer (1; 16) on one side of the proton exchange membrane;

30 a cathode catalyst layer (7) on the opposite side of the proton exchange membrane;

a gas distribution layer (3, 5) on each side of the proton exchange membrane (4), wherein a cooling and humidification liquid is fed to the anode side of the fuel cell,

characterized in
the step of adding a liquid methanol fuel to the
liquid.

5 20. A method according to claim 19,
characterized in
the step of distributing a water-methanol mixture in
such a way that one part of the anode catalyst layer is
in contact with a thin film of liquid methanol-water
10 mixture and a remaining part of the anode catalyst
layer is in contact with a gas phase free from liquid.

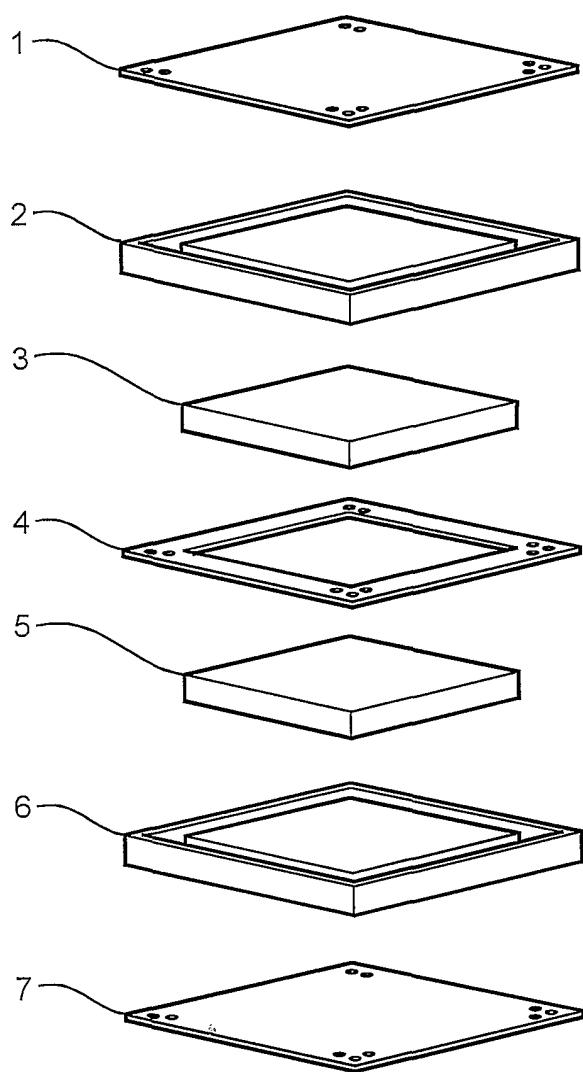


Fig.1

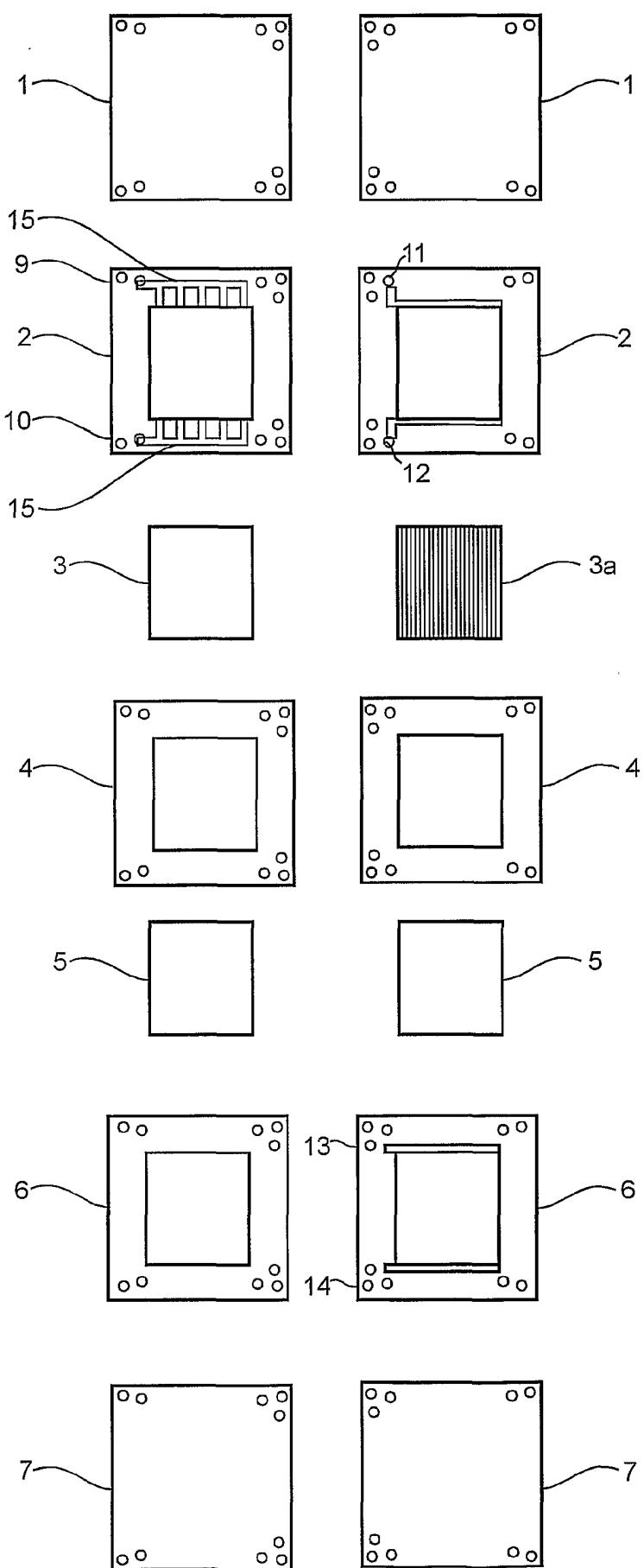


Fig.2

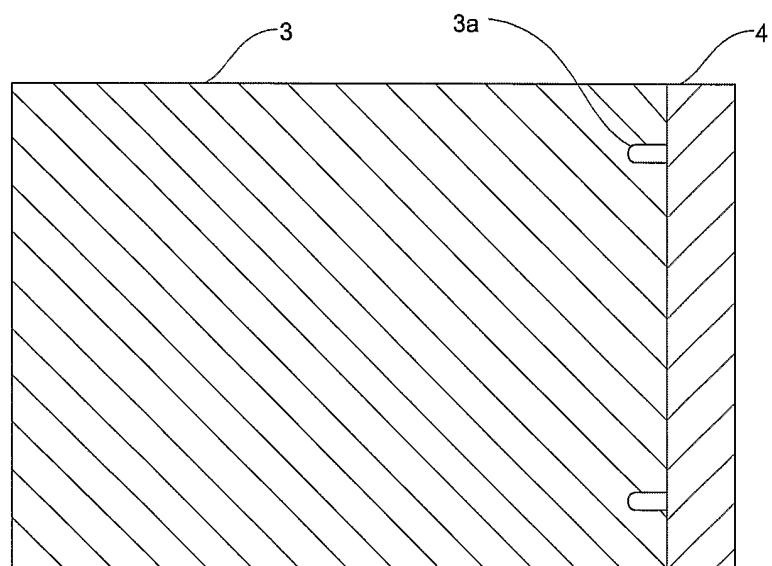


Fig.3

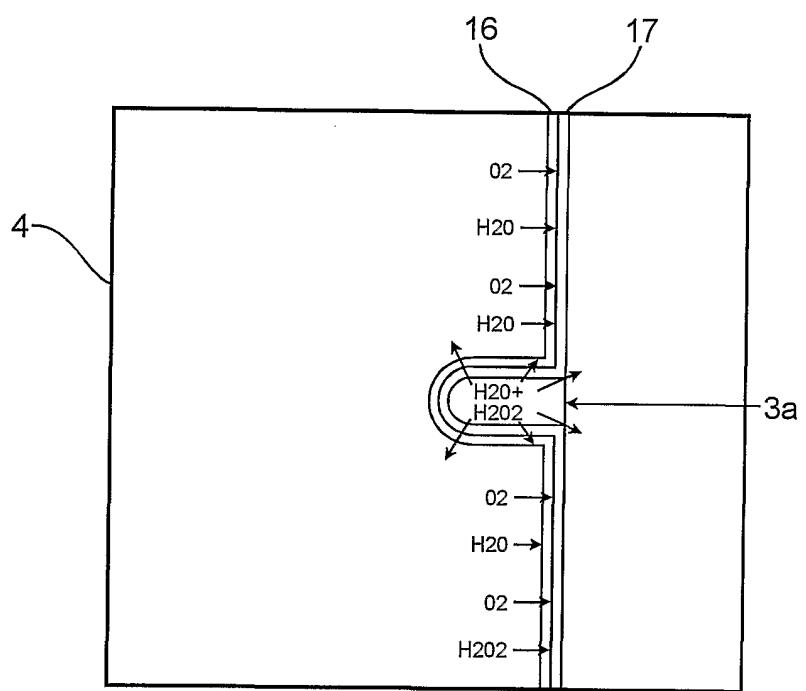


Fig.4

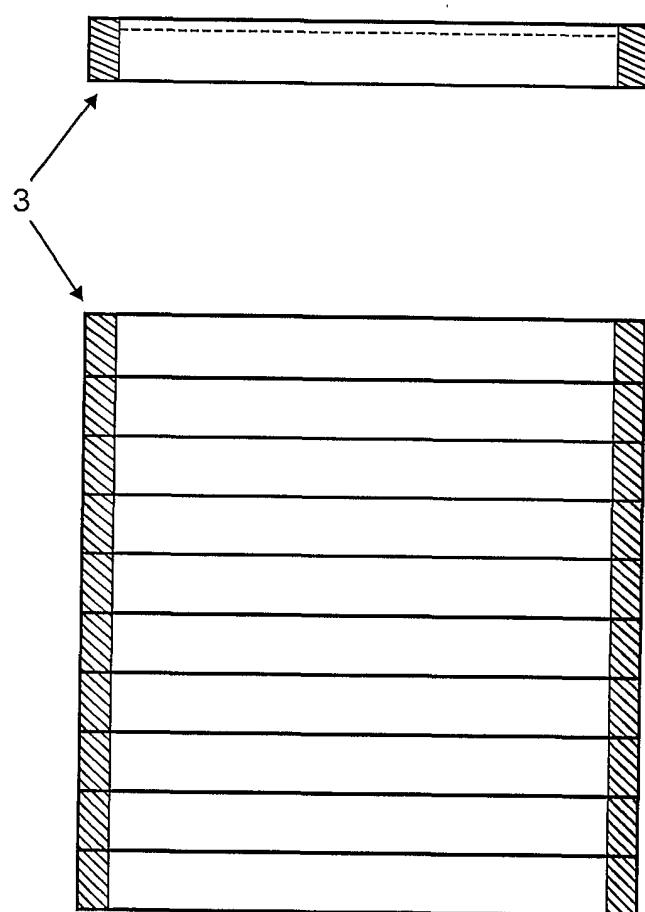


Fig.5

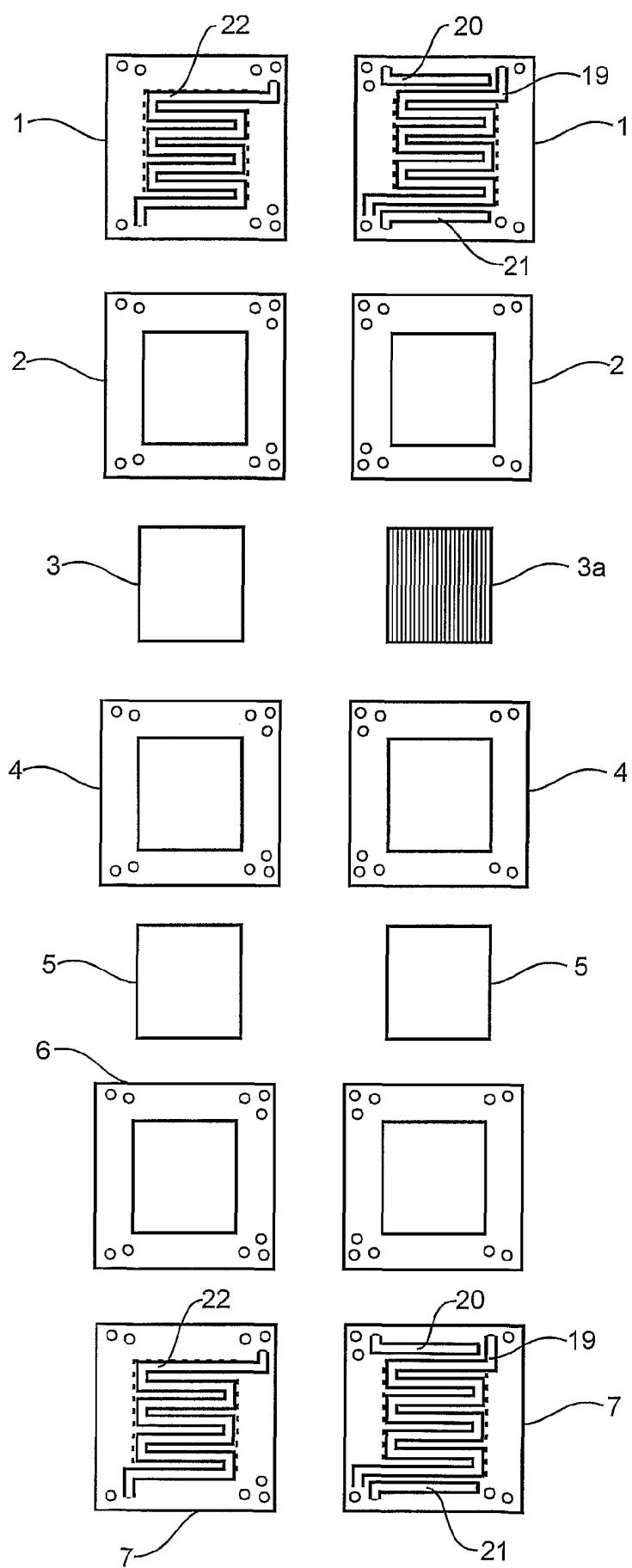


Fig.6

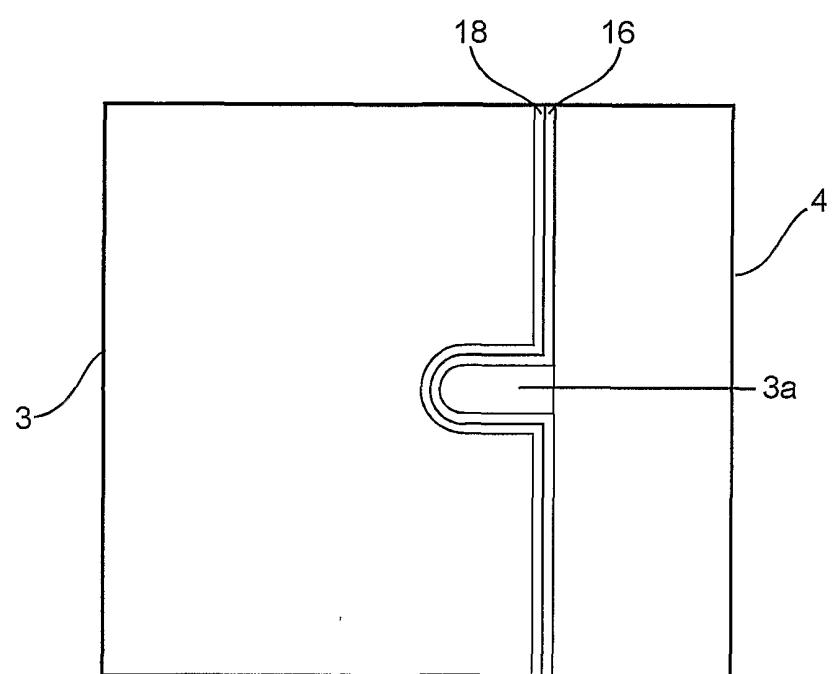


Fig.7

INTERNATIONAL SEARCH REPORT

International application No.

PCT/SE 01/01514

A. CLASSIFICATION OF SUBJECT MATTER

IPC7: H01M 8/02, H01M 8/04, H01M 8/10

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)

IPC7: H01M

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

SE,DK,FI,NO classes as above

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

EPO-INTERNAL, WPI DATA, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	WO 9852241 A1 (LOUGH-BOROUGH UNIVERSITY INNOVATIONS LIMITED), 19 November 1998 (19.11.98), page 1, line 14 - line 17; page 3, line 1 - line 10; page 5, line 10 - page 6, line 11 --	1,5-6
Y	US 5879826 A (PETER A. LEHMAN ET AL), 9 March 1999 (09.03.99), column 9, line 55 - column 10, line 60 --	1,5-6
A	EP 0975039 A2 (MATSUSHITA ELECTRIC INDUSTRIAL CO., LTD.), 26 January 2000 (26.01.00), column 5, line 27 - column 6, line 55 --	1-16

 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 another 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	Date of mailing of the international search report
7 December 2001	05-12-2001
Name and mailing address of the ISA/ Swedish Patent Office Box 5055, S-102 42 STOCKHOLM Facsimile No. + 46 8 666 02 86	Authorized officer Ulla Granlund/MP Telephone No. + 46 8 782 25 00

INTERNATIONAL SEARCH REPORTInternational application No.
PCT/SE01/01514**Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)**

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

See extra sheet

1. As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

The additional search fees were accompanied by the applicant's protest.
 No protest accompanied the payment of additional search fees.

II.

According to PCT Rule 13.2 the requirement of unity shall be fulfilled only when there is a technical relationship among the claimed inventions involving one or more of the same or corresponding "special technical features."

Invention 1, claims 1-16, relates to a polymer electrolyte fuel cell structure. The special technical feature of the invention is a gas distribution layer having water channels formed in the surface. The layer is enclosed by a sealing plate.

Invention 2, claims 17-18, relates to a method for operating a polymer electrolyte fuel cell. The special technical feature of the invention is the step of adding an oxygen evolving compound to a cooling and humidification liquid.

Invention 3, claim 19-20, relates to a method for operating a polymer electrolyte fuel cell. The special technical feature of the invention is the step of adding a liquid methanol fuel to a cooling and humidification liquid.

The special technical features of invention 1 and 2-3 are therefore not so linked as to form a singel general inventive concept.

INTERNATIONAL SEARCH REPORT

Information on patent family members

06/11/01

International application No.

PCT/SE 01/01514

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
WO 9852241 A1	19/11/98	AU	7439498 A	08/12/98
		EP	0996988 A	03/05/00
		GB	9709541 D	00/00/00
		GB	9720822 D	00/00/00
US 5879826 A	09/03/99	NONE		
EP 0975039 A2	26/01/00	JP	2000100454 A	07/04/00