Internal distribution code:
(A) [ ] Publication in OJ
(B) [ ] To Chairmen and Members
(C) [X] To Chairmen
(D) [ ] No distribution

Datasheet for the decision of 26 January 2012

Case Number: T 1921/08 - 3.3.05

Application Number: 01129420.4

Publication Number: 1217680

IPC: H01M 8/10, C25B 9/00

Language of the proceedings: EN

Title of invention:
An ion exchange and electrode assembly for an electrochemical cell

Applicant:
JAPAN GORE-TEX, INC.

Headword:
Electrode assembly/JAPAN GORE-TEX, INC.

Relevant legal provisions:
EPC Art. 54, 56, 123

Keyword:
"Added subject-matter (no)"
"Novelty (yes)"
"Inventive step (yes): Method steps not derivable from the prior art"

Decisions cited:
-

Catchword:
-

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 1921/08 **-** 3.3.05

D E C I S I O N
of the Technical Board of Appeal 3.3.05
of 26 January 2012

Appellant: (Applicant)
JAPAN GORE-TEX, INC.
42-5, 1-chome
Akazutsumi
Setagaya-ku
Tokyo-To 156 (JP)

Representative:
Moreland, David
Marks & Clerk LLP
Aurora
120 Bothwell Street
Glasgow G2 7JS (GB)

Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 2 April 2008 refusing European patent application No. 01129420.4 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman: G. Raths
Members: H. Engl
C. Vallet

Summary of Facts and Submissions

I. The appeal lies against the decision of the examining division posted on 2 April 2008 to refuse European patent application EP 01129420.4.

II. The documents cited during the examination procedure included the following:

D1: WO-A-89-06 055

D2: Mark W. Verbrugge *et al.*, *"Composite membranes for Fuel-Cell Applications"*, AICHE Journal, vol. 38, no. 1, 1992, pages 93 to 100

D3: EP-A-0 572 810

D6: US-A-4 865 930

III. The European patent application was refused in examination proceedings on the ground that claim 1 lacked an inventive step (Article 56 EPC).

The examining division argued in particular that:

- D2 represented the closest prior art;
- The problem to be solved was to provide a catalytically active PTFE-supported solid polymer ion exchange membrane/electrode assembly suitable for fuel cells;
- The claimed solution, namely to attach electrodes directly to composite membranes such as known from D2, was obvious in view of D1 and/or D3.

IV. The notice of appeal was filed with letter dated 30 May 2008. The grounds for appeal were received under cover of a letter dated 31 July 2008, accompanied by a main request and auxiliary requests 1 to 6.

V. In a communication dated 11 October 2011 the board provisionally raised objections under Article 123(2) EPC against the claims of the first, fifth and sixth auxiliary requests.

The board also addressed a potential problem of double-patenting arising in connection with granted European patent EP-B-718 903, in the name of the appellant.

Regarding inventive step, D2 appeared to disclose ***inter alia*** fully impregnated, non-porous composite membranes comprising Nafion as a solid polymer ion exchange resin and expanded porous PTFE (Gore-Tex) as a membrane support film. The board raised the question of whether the bonding of an electrode to such a composite membrane was an obvious step when the technical problem consisted in providing a membrane/electrode assembly for an electrochemical cell.

VI. Under cover of a letter dated 21 December 2011 the appellant filed amended claims as a main request and as auxiliary requests 1 to 7, replacing the requests previously on file.

VII. Oral proceedings took place on 26 January 2012. After discussion of the main request and auxiliary requests 1 to 4, the appellant filed auxiliary request 5 as a new main request.

VIII. Independent method claims 1 and 2 of said main request read:

"1. A method for producing a unitary assembly for an electrochemical cell; wherein the method comprises:

providing at least one pre-formed membrane support film of porous expanded polytetrafluoroethylene;

providing a first electrode comprising two planar surfaces;

superposing the membrane support film on the electrode; then

impregnating the pores of the membrane support film by solid polymer ion exchange resin, such that the pores of the expanded polytetrafluoroethylene are essentially completely filled with and made non-porous by solid polymer ion exchange resin so as to form a composite nonporous solid polymer ion exchange membrane and simultaneously intimately joining the membrane support film to a first planar surface of the first electrode by the said polymer ion exchange resin."

"2. A method for producing a unitary assembly for an electrochemical [sic]; wherein the method comprises:

separately forming a composite solid polymer ion exchange resin filled membrane support film; and subsequently

intimately bonding the composite solid polymer ion exchange resin filled membrane to a first planar surface of the first electrode by the said polymer ion exchange resin; wherein

separately forming a composite solid polymer ion exchange resin filled membrane support film comprises providing at least one pre-formed membrane support film of porous expanded polytetrafluoroethylene and impregnating the pores of the membrane support film by polymer ion exchange resin, such that the pores of the expanded polytetrafluoroethylene are essentially

completely filled with and made non-porous by solid polymer ion exchange resin."

Dependent method claims 3 to 8 concern specific embodiments of the methods of claims 1 and/or 2.

IX. The appellant essentially argued as follows:

Inventive step

D3 disclosed membrane/electrode assemblies for electrochemical cells wherein the membrane consisted of a radiation-grafted cationic exchange material such as fluorosulfonic acid membranes, acrylic acid membranes and methacrylic acid membranes. The membrane was plasma etched and coated with a catalytic material (Pt, Pd or Pt/C dispersed in a PTFE dispersion).

D2 described the synthesis of composite polymer membranes by depositing perfluorosulfonic acid (PSA) onto porous PTFE. Said composite Gore-Tex/Nafion membranes ranged from essentially pure Nafion to nearly pure Gore-tex. It was concluded that membranes having an intermediate Nafion content and a high porosity were potentially useful for fuel cells. Therefore, the person skilled in the art starting from D3 and considering the teaching of D2 would follow the explicit teaching of D2 that membranes having an intermediate PSA content should be used as cell membranes. Such membranes are porous. The skilled person would be led away from using membranes having a very high or very low Nafion content which were provided in D2 for experimental comparison only. The mere theoretical possibility of using a high PSA

content membrane was insufficient to lead a skilled person to consider such membranes, in particular as this went against the teaching of D2 that intermediate porous PSA membranes should be used.

The appellant argued that D2 did not teach a substantial filling of the pores with ion exchange material. Figure 8 of D2 did not suggest or imply such filling of the pores but rather a coating of the expanded PTFE with ion exchange resin. Therefore, claim 1 of the first auxiliary request which called for a substantial filling of the pores of the expanded PTFE with solid ion exchange material was not obvious in view of D2.

The appellant also argued that the prior art did not teach bonding or joining of electrode structures to a composite PTFE membrane using the same kind of solid polymer ion exchange resin as was used in filling the pores of the expanded PTFE membrane.

D6 disclosed an ion and gas permeable membrane obtained by fully impregnating a porous polymer substrate with an ion conducting polymer material to form a composite and subsequently stretching said composite to form pores. A skilled person looking for a membrane for use in a system that did not require transport of gas between anode and cathode would not look to D6, as D6 was specifically concerned with problems relating to such systems that comprise a gas reactant. Furthermore it would not be obvious to use an intermediate non-porous material. The suggestion to omit the stretching step would only be made with hindsight.

X. Requests:

The appellant requested that the decision under appeal be set aside and a patent be granted on the basis of the claims of the main request filed during oral proceedings.

Reasons for the Decision

1. Amendments (Article 123(2) EPC)

1.1 Claims 1 and 2 of the main request are based on claim 1 and the description, column 9, lines 29 to 41, as originally filed and published as EP-A-1 217 620. This passage of the description discloses the two alternative methods of forming the membrane/electrode assembly in accordance with the invention, as defined in independent claims 1 and 2, respectively.

The claim feature

"*such that the pores of the expanded polytetrafluoroethylene are essentially completely filled with and made non-porous by solid polymer ion exchange resin*"

is based on column 9, lines 21 to 23, of the application documents as originally filed.

1.2 Original disclosure for the subject-matter of claim 3 is found in column 9, lines 21 to 23, of the description, disclosing that the impregnation steps may be repeated.

Dependent claims 4 and 5 are based on the original disclosure of column 6, lines 29 to 35.

Claims 6 and 7 are based on the description, column 7, line 53 to column 8, line 8.

A basis for claim 8 is found in the description, column 9, lines 15 to 21.

1.3 The board is thus satisfied that the amended claims meet the requirements of Article 123(2) EPC.

2. Novelty

2.1 Novelty was not under dispute. None of the available documents disclose methods for producing a unitary assembly comprising the steps of providing a pre-formed expanded PTFE membrane support film impregnated and made non-porous by a solid polymer ion exchange resin, and joining or bonding an electrode to said membrane support film by the said solid polymer ion exchange resin, either simultaneously with the impregnating step (claim 1) or in a separate step (claim 2).

D1 discloses electrode-membrane assemblies comprising a Nafion®-impregnated composite of expanded Teflon and a bonded electrode of Pt/C. See pages 22, 23, Example 4. However, these composites are porous (gas-permeable).

D6 discloses a method for making a composite electrode structure comprising impregnating a substrate of porous expanded PTFE (Gore-Tex®) with a solid polymer ion exchange resin (Nafion) such that substantially all the

pores are filled with the ion-conducting polymer (column 3, lines 39 to column 4, line 3) and stretching said intermediate structure until the pores re-open or the ion exchange polymer exposes the pores (see column 4, lines 4 to 34; column 4, line 62 to column 5, line 20). The final composite membrane structure obtained in accordance with D6 is therefore porous.

The remaining documents are further removed from the claimed subject matter. They do not disclose composite membranes comprising expanded PTFE and solid polymer ion exchange resin of the kind used in the presently claimed methods.

2.2 In conclusion, the subject-matter of claims 1 and 2 of the main request is novel in view of the cited prior art. The same applies to the subject-matter of the dependent claims.

The requirements of Article 54 EPC are thus met.

3. Inventive step

3.1 The invention is concerned with methods for producing a unitary membrane/electrode assembly for electrochemical cells.

3.2 *Closest prior art*

The board considers that D2 represents the closest prior art document.

Said document is concerned with a study of composite membrane structures comprising a support film of

porous, stretched polytetrafluoroethylene (PTFE) (Gore-Tex® membrane) impregnated with varying amounts of a solid polymer ion-exchange resin (perfluorosulfonic acid; PSA) (Nafion®) (see page 93, abstract; page 93, right hand column, last line to page 94, left hand column, line 28; Figure 1; page 95, left hand column, lines 15 to 18; Figure 5; page 98, right hand column, line 26). The authors of D2 come to the conclusion that composite membranes of very low PSA content had characteristics similar to the hydrophobic PTFE substrate and were not of interest for fuel cells. However, for high PSA contents, the membrane behaviour was similar to that of the PSA polymer, except that the water permeability was reduced. From a fuel-cell membrane perspective, the optimal geometry corresponded to the point at which the membrane thickness was essentially determined by the Gore-Tex support and all of the Nafion was anchored inside the film (see Figure 8; page 99, left hand column, last paragraph; page 99, right hand column "Conclusions").

The method of producing said impregnated PSA/PTFE composite membranes involves the steps of providing small disks of a Gore-Tex membrane (expanded PTFE) (void volume about 80%, thickness about 50 µm, pore size about 0.2 µm), cleaning them thoroughly, treating the membrane disks with PSA polymer solution containing solubilised Nafion polymer) and evaporating the treatment solution at 160°C for 5 hrs (see the paragraph bridging pages 94 and 95). D2 thus already discloses a method for producing a composite membrane comprising a porous pre-formed support film of expanded PTFE which is impregnated with a solid polymer ion exchange resin.

3.3 *Problem underlying the application*

The technical problem of the application in suit in the light of D2 is to provide a method for producing a unitary assembly for an electrochemical cell consisting of a composite membrane and a bonded electrode.

3.4 *Solution*

As a solution to this technical problem, the application proposes a method according to claim 1, characterized in that the pores of the expanded PTFE membrane support film are impregnated by solid polymer ion exchange resin, such that the pores of the expanded polytetrafluoroethylene are essentially completely filled with and made non-porous by solid polymer ion exchange resin so as to form a composite nonporous solid polymer ion exchange membrane and simultaneously intimately joining the membrane support film to a first planar surface of the first electrode by the said polymer ion exchange resin.

The application in suit furthermore proposes a method according to claim 2, characterized in that a composite solid polymer ion exchange resin filled membrane support film is separately formed; a composite solid polymer ion exchange resin filled membrane support film comprising at least one pre-formed membrane support film of porous expanded polytetrafluoroethylene is separately formed and the pores of the membrane support film are impregnated by solid polymer ion exchange resin, such that the pores of the expanded polytetrafluoroethylene are essentially completely

filled with and made non-porous by solid polymer ion exchange resin; and in that subsequently the composite solid polymer ion exchange resin filled membrane is intimately bonded to a first planar surface of the first electrode by the said polymer ion exchange resin.

3.5 *Success of the solution*

The examples of the application in suit provide experimental evidence illustrating the claimed methods for producing the desired membrane/electrode assemblies.

The board is therefore satisfied that the underlying problem is successfully solved.

3.6 *Obviousness*

It remains to be decided whether the claimed solution was obvious having regard to the prior art.

3.6.1 During examination proceedings it was under dispute whether or not D2 disclosed a full impregnation with Nafion, ***i.e.*** an impregnation yielding an essentially non-porous membrane. The examining division relied in this respect ***inter alia*** on Figure 8 (page 99) of D2 as showing, in the left hand part of the Figure, a schematic representation of a fully impregnated, non-porous composite membrane.

The appellant essentially argued that it was not clear that said Figure 8 depicted a cross-section perpendicular to the film surface or rather a view from above onto the film's surface. The appellant argued

that the membrane shown in the left-hand part of Figure 8 should be interpreted as a porous membrane which was superficially coated with PSA, without however completely filling the pores of the membrane, because the coating solution at high PSA concentrations was too viscous to penetrate in the membrane pores.

In the board's view, Figure 8 of D2 may be best interpreted as a view from above (or below) onto the membrane's plane. The grid structure appears to schematically represent the porous expanded PTFE support. The hatched area clearly represents the Nafion (as is indeed explained in Figure 8 itself). This interpretation is consistent with the explanations in D2 that the impregnation with PSA was varied so as to range from practically pure Nafion to nearly pure Gore-Tex (page 95, passage bridging the left and right hand columns). Pure Nafion behaviour would be expected when the PTFE support was fully covered with Nafion, as shown in the left-hand embodiment of Figure 8 of D2. The right-hand part of Figure 8 shows a porous PTFE support only partially impregnated, with droplets of Nafion attached to the PTFE network.

3.6.2 In the left hand part of Figure 8, relating to a "High Nafion" situation, Nafion covers the porous PTFE support completely so as to make it apparently non-porous. However, the appellant argued plausibly that the relatively high viscosity of the polymer-containing impregnation solution would not be able to essentially completely fill the pores of the membrane, unless the impregnation is repeated. D2 does not disclose such a repeated impregnation, in contrast to the present application (see column 9, lines 21 to 24; example 2:

column 13, lines 15 to 17; example 3: column 14, lines 16 to 19). The board also considers that the nominal pore size of the expanded PTFE membrane in accordance with the present application is preferably between 0.05 to 5 µm (see claim 4). The pore size of the membrane used in D2 is 0.2 µm at a membrane thickness of about 50 µm (see page 93, right hand column, last line; page 99, right hand column, last paragraph). At this ratio of pore size to pore length the board considers it plausible that a conventional one-step impregnation with a relatively viscous fluid would not - or not essentially completely - fill the said pores with solid polymer ion exchange resin. Thus, it cannot be excluded that the porous support is only superficially coated with Nafion, leaving the pores internally open. D2 does not explicitly mention the resulting porosity of the PSA-coated composite membrane. There is no unambiguous and direct disclosure in D2 that the pores of the expanded polytetrafluoroethylene are essentially completely filled with and made non-porous by solid polymer ion exchange resin and that the claimed invention thus differs in this respect from D2. Therefore, D2 did not give the skilled person a clear hint at completely filling the pores of the support with ion exchange resin.

3.6.3 In the board's view a complete filling of the pores with solid polymer ion exchange resin according to the present application is evidently advantageous in that it facilitates the ion transport across the composite membrane.

3.6.4 The claimed invention furthermore proposes to join or bond a planar electrode to the composite membrane,

using the same solid polymer ion exchange resin as is used for impregnating the porous membrane support film, the bonding or joining being carried out either separately or simultaneously with the impregnation step.

3.6.5 As to the bonding of an electrode to a polymeric membrane, the following methods are known in the art.

a) D3 discloses plasma-etching of the surfaces of a radiation grafted polymeric membrane (selected from fluorosulfonic acid membranes, benzenesulfonic acid membranes and acrylic or methacrylic acid membranes) to be bonded to the cathode and anode, respectively, using an oxygen plasma at 100 millitorr and 200 W energy. After the plasma etching, the membrane is bonded to the anode and cathode layers using conventional thermal bonding practice (*e.g.* 200 °C temperature and 140.7 kg/cm^2 pressure). Optionally, prior to bonding, the anode and cathode may be impregnated with liquid Nafion solution to enhance the surface area of the membrane and to minimize resistance to ion movement through the membrane. See D3, page 3, lines 38 to 58; examples 1 to 6; claims 1, 2, 6, 11, 12, 14). D3 does therefore not hint at bonding electrodes to a membrane using a solid polymer ion exchange resin. The optional impregnation with Nafion solution does not facilitate the bonding, but reduces ion transport resistance.

b) As already discussed above, document Dl discloses electrode-membrane assemblies comprising a Nafion-impregnated composite of expanded Teflon and a bonded electrode of Pt/C. The impregnation with Nafion is only partial. These composites are porous and gas-permeable (see page 23, lines 1 to 15; claim 1). According to one

embodiment disclosed in D1, porous (expanded) PTFE backed fuel cell electrodes coated with 5% Nafion solution were pressed together with the PTFE surfaces in contact and the assembly cured at 120 °C for half an hour. The resulting porous structure comprised a sandwich structure of (from the outside): two electrodes; adjacent regions of porous Teflon containing islands of Nafion; and a central region containing islands of Nafion. Alternatively, a Nafion-impregnated Teflon composite may be simply pressed against fuel cell electrodes. See page 18, lines 1 to 17. Therefore, D1 does not suggest bonding of electrodes to a composite membrane using a solid ion exchange polymer resin as a bonding agent.

c) As mentioned above, D6 discloses composite membrane-electrode assemblies which are porous. Electrodes are then attached to the porous structure. D6 does not, however, disclose any details about the process of bonding the electrodes to the stretched composite membrane. Therefore, the skilled person would not derive from D6 the idea of joining or bonding an electrode to said membrane support film by the said solid polymer ion exchange resin, either simultaneously with the impregnating step (current claim 1) or in a separate step (current claim 2).

3.6.6 The remaining documents are further removed from the claimed subject matter. They do not disclose or suggest composite membranes of the kind used in the presently claimed methods.

3.7 The subject-matter of claims 1 and 2 therefore involves an inventive step.

3.8 The dependent claims 3 to 8 define preferred embodiments of the inventive process and derive their patentability from claim 1 or claim 2.

The requirements of Article 56 EPC are thus met.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the department of first instance with the order to grant a patent on the basis of claims 1 to 8 of the main request filed during oral proceedings and a description to be adapted.

The Registrar The Chairman

C. Vodz G. Raths