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Datasheet for the decision of 4 December 2008

Case Number: T 0271/07 - 3.3.07

Application Number: 93114421.6

Publication Number: 0592809

IPC: B01D 71/02

Language of the proceedings: EN

Title of invention:
Composite mixed conductor membranes for producing oxygen

Patent Proprietors:
AIR PRODUCTS AND CHEMICALS, INC.

Opponents:
Praxair Technology, Inc.

Headword:
-

Relevant legal provisions (EPC 1973):
EPC Art. 56

Keyword:
"Inventive step - Problem originally solved (no)"
"Reformulated problem"
"Solution obvious (yes)"

Decisions cited:
G 0002/88, T 0932/99

Catchword:
-

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Case Number: T 0271/07 **-** 3.3.07

D E C I S I O N
of the Technical Board of Appeal 3.3.07
of 4 December 2008

Appellants: (Opponents)
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Decision under appeal:
Interlocutory decision of the Opposition Division of the European Patent Office posted 13 December 2006 concerning maintenance of European patent No. 0592809 in amended form.

Composition of the Board:

Chairman: S. Perryman
Members: F. Rousseau
G. Santavicca

Summary of Facts and Submissions

I. The appeal lies from an interlocutory decision of the Opposition Division of 13 December 2006, according to which, account being taken of the amendments made by the proprietors during the opposition proceedings, European patent 0 592 809 (granted on European patent application N° 93 114 421.6) and the invention to which it relates were found to meet the requirements of the EPC.

II. The European patent in suit was granted with a set of sixteen claims, with three independent claims reading as follows:

"1. A membrane capable of separating oxygen from an oxygen-containing gaseous mixture, which membrane comprises a dense layer having no connected through porosity and a plurality of porous layers having an average pore radius of less than about 10 micrometers wherein the average pore radius of each respective porous layer is larger than the average pore radius of the preceding layer as function of distance away from the dense layer, the porous and the dense layer which are independently formed from a multicomponent metallic oxide capable of conducting electrons and oxygen ions at temperatures greater than about 500°C.

8. A membrane capable of separating oxygen from an oxygen-containing gaseous mixture, which membrane comprises a first porous layer formed from a multicomponent metallic oxide having an average

pore radius of less than about 10 micrometers which is deposited to a second porous layer having an average pore radius greater than the radius of the first layer but less than about 10 µm which is not a mixed conducting oxide, the first porous layer being contiguous with a dense layer having no connected through porosity comprising a multicomponent metallic oxide, said multicomponent metallic oxides being capable of conducting oxygen ions at temperatures greater than about 500°C.

16. A membrane capable of separating oxygen from an oxygen-containing gaseous mixture, which membrane comprises a first porous layer and a second porous layer having an average pore radius of less than about 10 micrometers which are separated by and contiguous with a dense layer having no connected through porosity wherein the first porous layer, the second porous layer and the dense layer are independently formed from a multicomponent metallic oxide capable of conducting electrons and oxygen ions at temperatures greater than 500°C."

III. An opposition was filed against the patent by the present appellants, which opposition was rejected by a first decision of the Opposition Division posted on 15 July 1999. Following an appeal against that decision the case was remitted to the Opposition Division for further prosecution by appeal decision T 0932/99 of 3 August 2004.

IV. The second decision of the Opposition Division of 13 December 2006, against which the present appeal lies, was based on a set of amended claims 1 to 22 submitted

as the Main Request during the oral proceedings held on 7 November 2006 before the Opposition Division, of which claim 1 read as follows:

"1. Use of a membrane
comprising a dense layer having no connected through porosity and a plurality of porous layers having an average pore radius of less than about 10 micrometers wherein the average pore radius of each respective porous layer is larger than the average pore radius of the preceding layer as function of distance away from the dense layer, the porous and the dense layer which are contiguous and independently formed from a multicomponent metallic oxide capable of conducting electrons and oxygen ions at temperatures greater than about 500°C to separate oxygen from an oxygen-containing gaseous mixture by delivering the oxygen-containing gaseous mixture into a first gas compartment which is separated from a second gas compartment by said membrane, establishing a positive oxygen partial pressure difference between the first and the second gas department, contacting the oxygen-containing gaseous mixture with the membrane at a temperature greater than 500°C, thereby separating the oxygen containing gaseous mixture into an oxygen permeate and an oxygen-depleted stream."

V. According to the decision under appeal:

(a) The Main Request fulfilled the requirements of Article 123, paragraphs (2) and (3), EPC.

(b) The change of category in the independent claims, from a product to the use of that product, was in compliance with G 2/88 (OJ 1990, 93).

(c) Independent Claims 1, 8 and 16 were clear and did not lack essential features of the alleged invention such as the recovery of the separated oxygen, the thinness of the dense layer and the pressure difference between the two sides of the membrane, which was the driving force.

(d) Novelty was not contested. In particular, D1 [Y. Teraoka & al., "*Development of Oxygen Semipermeable Membrane Using Mixed Conductive Perovskite-Type Oxides*" (Part 2), J. Ceramic Soc. Jpn. Inter. Ed., Vol. 97, 1989, pages 523-529] did not disclose an average pore radius less than 10 µm and an increase of the pore size in the porous layers as a function of distance away from the dense layer.

(e) As to inventive step, D1 described the closest prior art. The problem to be solved was an increase of the oxygen flux through the membrane. That problem had been solved by the small pore structure as claimed, which mitigated the surface kinetic rate limitations for gas-solid oxygen exchange, which were responsible for the overall oxygen flux in composite membrane with thin dense layer. D1 hinted at a higher porosity of the porous layers, which, for stability of the dense layer, should be obtained with finer pores. According to D33 [J. D. Wright et al., "*Advanced Oxygen Separation Membranes*", Report N° TDA-

GRI-90/0303, Gas Research Institute, September 1990], there were similarities between the fields of solid oxide fuel cell and ion transport membrane devices but also important differences such as that the membrane of a solid oxide fuel cell must not be mixed conducting. Also, the similarities rather concerned the devices and not their uses. D36 [US-A-5 114 803] addressed the problem of increasing diffusion of gases through the porous electrodes of a solid oxide fuel cell and proposed to solve that problem with features as those distinguishing the claimed subject-matter from the disclosure of D1. Nevertheless, the skilled person looking at an improved process for separating oxygen would not look at a solid oxide fuel cell as disclosed by D36. Even if he did look at D36, he would not find any hint on how to improve oxygen flux in a separation process as that of D1. Therefore the subject-matter of Claims 1 and 8 were not rendered obvious by D1 and D36. Nor was the subject-matter of Claim 16 rendered obvious by D1 and D29 [US-A-4 957 673], because D29 did not concern pressure driven separation of oxygen as D1.

(f) Therefore, the claimed subject-matter met the requirements of Article 56 EPC and the Main Request fulfilled the requirements of the EPC.

VI. In their statement setting out the grounds of the appeal as well as in the response to the communication of the Board in preparation for the oral proceedings, the opponents (appellants) maintained that the subject-matter of each of the independent claims extended beyond the content of the application as filed

(Article 123(2) EPC), lacked essential features of the alleged invention (which was considered as a contravention of the requirements laid down in Article 84 EPC and/or Article 56 EPC), and were obvious (Article 56 EPC).

VII. By letter dated 12 November 2007, the patent proprietors (respondents) filed observations on the grounds of appeal and enclosed amended claims as 1st and 2nd Auxiliary Requests. Then, with letter dated 31 October 2008, in response to a communication of the Board in preparation for the oral proceedings, the respondents ***inter alia*** submitted a new Main Request, and new 1st to 21st Auxiliary Requests.

VIII. Oral proceedings were held on 4 December 2008. The respondents withdrew what had been filed as the Main Request and the 1st, 11th and 12th Auxiliary Requests submitted with letter dated 31 October 2008. What had been filed as 2nd Auxiliary Request thus became the Main Request, with what had been filed as 3rd to 10th and 13th to 21st Auxiliary Requests being maintained as seventeen auxiliary requests. The claims 1 of the maintained requests were as follows:

Main request (filed as 2nd Auxiliary Request)

"1. Use of a membrane comprising a dense layer having no connected through porosity and a plurality of porous layers having an average pore radius of less than about 10 micrometers wherein the average pore radius of each respective porous layer is larger than the average pore radius of the preceding layer as function of distance away from the dense layer, the porous and the dense

layer which are contiguous and independently formed from a multicomponent metallic oxide capable of conducting electrons and oxygen ions at temperatures greater than 500°C, demonstrating an oxygen ionic conductivity ranging from 0.01 $ohm^{-1}cm^{-1}$ to 100 $ohm^{-1}cm^{-1}$ and an electronic conductivity ranging from about 1 $ohm^{-1}cm^{-1}$ to 100 $ohm^{-1}cm^{-1}$ and wherein the dense layer has a thickness ranging from 0.01 micrometer to about 500 micrometers and the porous layer has a thickness ranging from 1 micrometer to about 2 millimeters.(sic) to separate oxygen from an oxygen-containing gaseous mixture

by delivering the oxygen-containing gaseous mixture into a first gas compartment which is separated from a second gas compartment by said membrane, establishing a positive oxygen partial pressure difference between the first and the second gas compartment, contacting the oxygen-containing gaseous mixture with the membrane at a temperature greater than 500°C, thereby separating the oxygen-containing gaseous mixture into an oxygen permeate and an oxygen-depleted gaseous stream."

Auxiliary requests

Compared to claim 1 of the main request, claim 1 of the first auxiliary request (originally filed as 3rd Auxiliary Request) further restricts the average pore radius ("of 0.1 to less than about 10 micrometers") and the thickness of the dense layer ("of less than about 100 micrometers"), but omits the requirements concerning the oxygen ionic and the electronic conductivities.

Claim 1 of the second auxiliary request (originally filed as 4th Auxiliary Request) compared to claim 1 of the main request contains the said further amendments to the average pore radius ("of 0.1 to less than about 10 micrometers") and the thickness of the dense layer ("of less than about 100 micrometers").

Claim 1 of the third auxiliary request (originally filed as 5th Auxiliary Request) compared to claim 1 of the main request contains the amendments "to recover oxygen" and "and recovering the oxygen permeate stream", but has no limitation whatsoever as regards the thickness of the dense and the porous layers, nor of the oxygen ionic and electronic conductivities.

Claim 1 of the fourth auxiliary request (originally filed as 6th Auxiliary Request) compared to claim 1 of the main request contains the amendments "to recover oxygen", "and recovering the oxygen permeate stream", and refers to an average pore radius "of 0.1 to less than about 10 micrometers", but has no limitations whatsoever as regards the thickness of the dense and the porous layers, and of the oxygen ionic and electronic conductivities.

Claim 1 of the fifth auxiliary request (originally filed as 7th Auxiliary Request) compared to claim 1 of the main request contains the further limitations "to recover oxygen" and "and recovering the oxygen permeate stream".

Claim 1 of the sixth auxiliary request (originally filed as 8th Auxiliary Request) compared to claim 1 of the main request contains the additional features "an

average pore radius of 0.1 to less than about 10 micrometers", "to recover oxygen", "and recovering the oxygen permeate stream" and "wherein the dense layer has a thickness of less than about 100 micrometers", but has no limitations as regards the oxygen ionic and electronic conductivities.

Claim 1 of the seventh auxiliary request (originally filed as 9th Auxiliary Request) compared to claim 1 of the main request contains the additional features "an average pore radius of 0.1 to less than about 10 micrometers", "to recover oxygen", "and recovering the oxygen permeate stream" and "wherein the dense layer has a thickness of less than about 100 micrometers".

Claim 1 of the eighth auxiliary request (originally filed as 10th Auxiliary Request) differs from claim 1 of the main request in the additional features "to recover oxygen", "and recovering the oxygen permeate stream" and "wherein the dense layer has a thickness of less than about 100 micrometers".

The claims 1 of the eighth to fifteenth auxiliary requests (originally filed as 13th to 21st Auxiliary Requests) correspond to the respective claims 1 of the main request and the first to seventh auxiliary requests but with the insertion of the term "about" before the mention of the value of "500°C".

IX. In view of the extensive amendments made to the claims as granted the arguments of the respondents (patent proprietors) are given first, who essentially argued as follows:

Main request

(a) The amendments in claim 1 and the change of category from product to use were made to meet the objections made as to inventive step, and thus were allowable under Rule 80 EPC.

(b) The amended claims were based on the use of the membranes defined in claim 1 as granted, while the use steps concerned the separation of oxygen. The change of category (from a membrane to the use of that membrane) aimed at distancing from fuel cell membrane technology and was in accordance with G 2/88 (OJ EPO *supra*). Since the mention "recovery of permeated oxygen" in the application as filed meant "separation of oxygen", it was not necessary to specify the step "recovery of the permeated oxygen" in the amended claims. The claims thus met the requirements of Article 123(2) EPC. The term "about" had been cancelled for consistency. The claim was also clear.

(c) Novelty of the claimed subject-matter was not in dispute.

(d) As to inventive step, the closest prior art was described in D1, which, however, did not disclose an average pore size and a plurality of porous layers arranged and having the pore structure as defined in Claim 1.

(e) The problem to be solved was to increase the interfacial area of the gas and solid phases without causing any significant pressure drop or resistance

to mass transfer, in order to achieve an increased oxygen flux.

(f) Since the solid oxide fuel cell art was not neighbouring on the ion transport membrane art, e.g. the mechanisms were different, there were differences in the aims, in the materials, in the structure of the dense layers used, so that documents relating to this art should not be taken into account when assessing inventive step.

(g) In fact, D12, D33, D36 and D45 addressed problems which differed from the problem addressed in the patent in suit. In particular, D36 aimed at improving the output power of the electrode and gave no hint at the solution of the problem underlying the patent in suit. Also, D36 was so contradictory (e.g. the material of the dense layer did not possess enough electronic conductivity and Experiment III (using a pore radius greater than 10 µm) gave better output than Experiment II (using a pore radius smaller than 10 µm)) that it taught away from the claimed solution. Hence, the subject-matter of Claim 1 was not obvious.

Auxiliary requests

The further amendments to the claims in the auxiliary requests were based on the application as filed and, respectively, addressed the alleged lack of clarity ("about") and/or definition of the essential features in the claims, in particular having regard to the question whether the problem underlying the patent in suit had been solved.

X. The appellants essentially argued as follows:

Main request - claim 1

(a) Whilst claim 1 as granted concerned a membrane for separating oxygen, claim 1 now concerned the use of that membrane for separating oxygen. However, the claimed use did not include the step "recovering the oxygen permeate stream", which was the final step of the only use of the membrane disclosed in the application as filed. Consequently, the use of a solid oxide fuel cell, a device in which oxygen was separated but not recovered, was now encompassed by the terms of the amended claims albeit it was not disclosed in the application as filed. Therefore, the subject-matter of the application as filed had been extended, which was contrary to Article 123(2) EPC.

(b) No objection under Article 123(3) EPC was raised.

(c) Even if the fact that claim 1 lacked essential features for achieving any improvement such as requirements as to the thickness of the dense layer or as to the total pressure difference across the membrane, should be found not to be objectionable under Article 84 EPC, this should be taken into account when considering inventive step, as without such requirements the problem stated to be underlying the patent in suit could not be regarded as solved.

(d) Novelty was not disputed, as D1 did not show a plurality of porous layers though it showed the use

of a membrane made up of mixed (ionic and electrical) conducting materials, namely a 15 µm thick dense layer which had been deposited on a 2 mm thick porous support having average pore size ranging from 20 to 30 µm (i.e. an average pore radius from 10 to 15 µm).

(e) D1 described the closest prior art. If the problem to be solved were stated as being to increase the oxygen flux it was not plausibly solved since claim 1 stated no requirements as to how thin the dense layer had to be. Even according to the theoretical examples of the patent in suit, which exemplified a thickness as thin as 10 or 5 µm, an improved oxygen flux could not be attained with a thickness of the dense layer above 100 µm. The problem to be solved compared to D1 was the mere provision of a further use.

(f) As regards the obviousness of that further use, D1 already hinted at using finer pores for increasing the permeated oxygen flux, so that the mere reduction of the pore size from 30 to 20 µm to below 20 µm was obvious. Ways of achieving this could be derived from the other documents in particular D36, in which the same arrangement of layers and pores as now claimed was used to improve diffusion of the gases through the electrodes, in order to increase the oxygen flux.

(g) A skilled person faced with improving oxygen flux would consider solutions adopted for this purpose in neighbouring fields. In particular D33 showed the common general knowledge of the skilled person

and disclosed that the fields of solid oxide fuel cell and ion transport membrane had many similarities and used similar materials and fabrication techniques.

(h) D45 [US-A-4 330 633] showed that mixed conducting electrodes as disclosed in D36 could also be used as membranes for oxygen separation. D12 [H. L. Tuller et al., "Doped Ceria as a Solid Oxide Electrolyte", Journal of The Electrochemical Society, February 1975, pages 255-259] disclosed that solid oxides could have mixed conducting properties, thus being suitable for both solid oxide fuel cells and ion transport membranes. Therefore, the use of D1 being of mixed conduction would be no reason for the skilled person not to look at the neighbouring field of solid oxide fuel cell.

(i) It was a fact that the combination of the distinguishing features of the claimed subject-matter over D1 was known from D36, in which the same arrangement of layers and pores was used to improve diffusion of the gases through the electrodes, in order to increase the oxygen flux. Thus, D36 also addressed the same aim as the patent in suit. The arguments of the respondents that the teaching of D36 was anyhow contradictory were not convincing. Experiment III of D36 (Table 1) fulfilled the conditions set out in Claim 1, because five layers of a mixed conducting material, of decreasing size, had been deposited on the air electrode tube, in which the average pore radius increased from 0,65 to 13,4 µm, so that a number of

layers below the dense layer had an average pore radius less than 10 µm.

(j) Therefore, the skilled person would have combined D1 and D36, and would have obviously arrived at the claimed subject-matter.

Further Auxiliary Requests

(k) The further modifications to the claims 1 of the auxiliary requests, neither changed the problem to be solved nor the fact that the problem as stated in the patent was not solved within the whole breadth of the claims. Therefore, the ground of opposition invoked, lack of an inventive step, also prejudiced the maintenance of the patent in suit in the version according to any of the further Auxiliary Requests on the same line of argument as for claim 1 of the main request.

Conclusion

(l) Therefore the patent should be revoked.

XI. The appellants have requested that the decision under appeal be set aside and that the patent be revoked.

XII. The respondents have requested that the decision under appeal be set aside and that the patent be maintained on the basis of the claims of one of the requests filed as 2nd to 10th or 13th to 21st Auxiliary Requests on 31 October 2008.

Reasons for the Decision

1. The appeal is admissible.

Claim 1 of main request (filed as 2nd Auxiliary Request)

2. *Amendments (Articles 84 and 123(2) EPC)*

The appellants raised objections under Articles 84 and 123(2) EPC to the basis and the clarity of the changes made compared to claim 1 as granted. The Board does not consider these objections made out under these articles, though the objections have some relevance to what problem can be regarded as solved. Since the issues under these articles are not decisive for the outcome of the appeal, the Board will not elaborate on them.

3. *Novelty*

Novelty was not in dispute. In particular, it was not contested that the membrane whose use forms the subject-matter of Claim 1 of this request is distinguished from the membrane used as disclosed in D1, at least by the requirement of a plurality of porous layers and that the average pore radius of each respective porous layer is larger than the average pore radius of the preceding layer as function of distance away from the dense layer.

Inventive step

Closest prior art

4. The patent in suit concerns the use of composite mixed conductor membranes for producing oxygen as does D1, which is acknowledged in the patent in suit as the closest prior art. The decision under appeal and both parties on appeal treated D1 as describing the closest prior art for assessing inventive step and the Board also agrees that it is appropriate to treat D1 as describing the closest prior art.

4.1 D1 concerns the use of oxygen semipermeable membranes using mixed-conductive perovskite-type oxides (title).

According to D1, when a dense film of mixed conductive perovskite type oxides in the form of a membrane is subjected to different oxygen partial pressures applied on either sides, at a temperature higher than 500°C, oxygen permeates from the high oxygen partial pressure side to the low oxygen partial pressure side. Oxygen molecules are ionized on the high oxygen partial pressure side and transit through the membrane in the form of oxygen ions, which are then discharged and released on the low oxygen partial pressure side, while electrons (or holes) required for this discharge quickly transit through the membrane (Point 1., Introduction, first two paragraphs). The use of the membrane disclosed in D1 and that described in the patent in suit rely on the same mechanisms and conditions (high temperature and driving force).

4.2 The oxygen semipermeable membrane used in D1 has an asymmetric structure and is made of mixed conductive perovskite-type oxides. In particular, a dense film of $La_{0.6}Sr_{0.4}CoO_3$ (LSCO) is spray deposited on a porous LSCO substrate that has a thickness of about 2 mm (page 524, Point 2-1, line 7) and open pores with size of 20 to 30µm, i.e. a radius of 10 to 15µm (page 524, paragraph 3-1). The dense film has a thickness of 15 µm (Abstract; page 527, left column, line 10). It is not contested that $La_{0.6}Sr_{0.4}CoO_3$ (LSCO) possesses oxygen ionic and electronic conductivities as required by claim 1.

As regards the performance, asymmetric structures (samples) with a thin dense film layer (about 15 µm thick) were compared with structures consisting of dense sintered disks (about 1.5 mm thick; page 528, right column, first paragraph, first five lines), in order to measure the rate of oxygen permeation (Figure 9 of D1).

4.3 Although the rate of oxygen permeation of the structure with the thin film element was about twice as high a rate of oxygen permeation as that of the sintered disk sample of 1.5 mm (Page 529, Point (3) of the conclusions), it nevertheless was around 1/5 of an expected value which had been estimated by calculation from Equation (1) in D1 (page 528, in particular right column, first paragraph, last line)). That estimation had been made ***inter alia*** under conditions which were such that the physical diffusion of oxygen gas through a porous body did not determine the rate of oxygen permeation.

Problem and solution

5. According to the patent in suit, the use of the asymmetric membrane described in D1 had not attained the expected oxygen flux increase based upon considerations limited to the dense layer thickness (page 2, lines 54 to 57).

5.1 Having regard to the prior art as embodied in D1, the patent in suit formulated the problem to be solved as being the use of a thin, supported solid state ionically conductive membranes for attaining superior oxygen flux without sacrificing the mechanical and physical compatibility of the composite membrane (patent, page 2, last two lines).

5.2 According to the patent in suit, the claimed solution consists in the use of an arrangement of the layers providing superior oxygen flux because of an increased interfacial area suitable to counteract the kinetic limitations associated with oxygen transport, without causing any significant pressure drop or resistance to mass transfer (page 3, lines 14 and 15; page 5, lines 56 to 59).

6. Hence, it has to be assessed whether or not the problem stated in the patent in suit can be considered as solved, in particular within the whole breadth of the claims, by the features as claimed.

6.1 The patent in suit contains examples and counter examples illustrating the claimed uses (page 8, lines 3-4). The examples are based on "a constructive reduction to practice by a mathematical computer

simulation" (page 14, last two lines). That mathematical model is detailed on pages 8 to 13. From page 14, it can be gathered that the behaviour of a specific mixed conducting perovskite oxide ($La_{0.2}Ba_{0.8}Co_{0.8}Fe_{0.2}O_{3-\delta}$), having the physical parameters given in Table 2, some of which are literature data and others have been adjusted to give an improved least squares fit, has been simulated at a temperature of 850°C. Throughout all of the examples, apart from the different arrangement or size of the porous layers, the same model, material and physical parameters have been used to calculate the performance exemplified, i.e. the oxygen flux. In summary, the performance data of the exemplified uses is the result of computer simulations on specific membranes at a specific temperature, and not the result of any practical verification of the model or the calculated results.

6.2 From the results of the computer simulation shown in Tables 3 to 8, the following picture can be gathered.

(a) Since the exemplified fluxes are calculated, for specific conditions, and no experimental comparison is available, the reliability or efficacy of the model is not apparent. Reliability or efficacy cannot simply be assumed, as D1 itself shows that calculated expectations may well not be fulfilled in practice.

(b) The patent in suit does not provide any direct comparison with a membrane as described in D1 (e.g. dense layer of 15 µm and porous active layer of 1.5 or 2 mm), so there is no evidence for any actual improvement over D1.

(c) As regards the role played by the thickness of the dense film, which can be as high as 500 µm in the claimed uses, Table 4 shows that the best performing membranes (runs 29-40) comprise a plurality of active layers situated on the membrane permeate side, wherein the pores are either funnel shaped (pore radius ranging from 0.5 to 10 µm) (runs 29-32) or distributed in two different layers (runs 33-40), the first having a pore radius of 0.5 µm and the second of 10 µm, respectively, wherein however the dense film is 10 µm and the first porous layer, supporting the dense film, is only 50 µm thick. There is no evidence that with thicker dense films, e.g. up to 500 µm, any improvement could still be achieved. On the contrary, it is apparent from Table 4 that under the same pressure differential, the use of a membrane having a dense layer of 200 µm on a mixed conducting porous layer of 2 mm having pore radius of 0.5 µm (run 21 of Table 4) results in an oxygen flux which is less than half that of a membrane having a dense film of 10 µm (run 17 of Table 4) (i.e. the obtainable flux decreases with the thickness of the dense film) and even tend to approach the flux obtainable with a membrane having an inert porous layer and a dense layer of 10 µm (run 9 of Table 4).

(d) Hence, it is not plausible that thick dense films, which are yet part of the subject matter of claim 1, would necessarily attain any improvement in the oxygen flux compared to D1.

(e) Table 5, while not concerning the use of a membrane as claimed, nevertheless shows that also the operating conditions influence the oxygen flux. A membrane with a dense layer of 10 µm on a 2 mm inert layer (run 45) can perform better than a membrane with a dense layer of 10 µm on a 2 mm active layer (run 50). The claimed uses are not restricted to any specific operating conditions, so it is a completely open question whether for all the temperatures falling within the claim, above about 500°C, an improved oxygen flux could even theoretically be obtained.

(f) Table 6 concerns sandwich structures but only shows dense layers of 10 µm, sandwiched between porous layers, the best oxygen fluxes being obtained when the thickness of the porous layers is 0.5 mm.

(g) Table 7 shows the effect of the average pore radius on oxygen flux, however for membranes comprising a dense layer and a porous layer, both of mixing conducting materials, hence, not in line with the membranes defined in the claims. The Board notes, ***inter alia***, (runs 75-76) that the performance of a membrane similar to that described in D1 (runs 74 and 76, wherein the only distinguishing feature from D1 is the thickness of the dense layer = 10 µm) can be comparable to that of a membrane having an average pore radius of 0.5 µm (run 64).

(h) Table 8 shows that in a composite membrane having a dense film of 5 µm, oxygen flux increases if the thickness of porous layers contiguous to the dense

layer and the average pore radius are all kept very small. This is not reflected in the claims.

6.3 In summary, the examples rely on calculated performance based on very specific assumptions or limitations, in particular very thin dense films, not reflected in claim 1. According to the claim, the dense layer can be as thick as 500 µm (compared to 15 µm used in D1), the average pore radius might be slightly lower than 10 µm, the first active porous layer may have any thickness and the use may be carried out under any operating conditions.

6.4 Further the claims while referring to pore radius, are silent as to the distribution concentration of the pores, a feature mentioned in D1 (see discussion point 7.2 below) as of importance.

6.5 Therefore, it has not been shown and it is not plausible either that the problem stated in the patent is suit has been solved within the whole breadth of Claim 1.

6.6 It follows from the above that the problem to be solved and actually solved by the claimed subject-matter, can only be regarded as being to provide a further use over that described in D1.

Obviousness

7. It remains to decide whether the provision of a further use as claimed was obvious for the skilled person using common general knowledge having regard to D1 as the closest prior art.

7.1 In order to further increase the rate of oxygen permeation of the thin film element, D1 (conclusions, (4), page 529) states "...it is necessary not only to disperse the open pores in the porous substrate into finer pores and to increase the effective surface area of pores, but also to develop techniques to prevent any variation in the surface composition of dense thin films."

7.2 In order to increase the porosity of the substrate, D1 does not recommend an enlargement of the pore size, because this makes it more difficult to form dense thin films on it, but to make the open pores of the substrate finer and to increase their distribution concentration (paragraph bridging pages 528 and 529), that is to increase the effective surface area of the pores (conclusions, (4), page 529). Hence, D1 directly points the skilled person towards reducing the pore size of the porous supporting layer.

7.3 The arguments used by the appellants, also taken up in the decision under appeal, regarding the differences between the art of solid oxide fuel cells and that of ion transport membranes, are valid only to the extent that a document concerning a solid oxide fuel cell is not appropriate as the starting point in the closest prior art. Here it is not disputed that D1 is in the same field of ion transport membranes as the subject matter of claim 1. A skilled person seeking to implement the suggestion in D1 to make finer pores is not concerned with whether the solid metallic oxide used to form a porous substrate is also capable of conducting electrons, as in an ion transport membrane

or not capable of conducting electrons as is usual for a solid oxide fuel cells. The methods of forming a porous substrate from solid oxides are shared by the two fields, and a skilled person following the suggestion of D1 will also pay attention to the formation of porous substrates in the solid oxide fuel cell field (see also decision T 0932/99 of 3 August 2004 on the first appeal relating to this patent).

7.4 D36 ***inter alia*** concerns the formation of a porous electrode for a solid oxide fuel cell, said porous electrode having one surface on which a solid electrolyte film having an ionic conductivity is to be formed, wherein a pore diameter of the porous electrode on the side of said one surface is smaller than that of the porous electrode on the other surface (Claim 10).

In that porous electrode, the diameter of the particles of that portion of said porous electrode which is in contact with the solid electrolyte film is smaller than that of the surface portion on the side opposite to the interface (Claim 11).

In particular, the pore diameter of the porous electrode is stepwise increased in the direction of the thickness of the electrode from the side of interface to the side opposite to the interface (Claim 12).

According to the examples of D36, the multilayer porous support of Experiments II and III had an average pore diameter ranging from 0.7 µm (solid electrolyte side) to 8.5 µm (gas side) or 1.3 to 26.8 µm, respectively. Hence, the pore radius of all the layers of the support of Experiment II as well as that of a number of (the

five) layers of the support of Experiment III, is less than 10 µm.

Compared to the porous support of Experiment I, having uniform average pore diameter, the porous supports having graduated pores reduced the electrical resistance (column 7, lines 5 to 48, particularly 46 to 65; table 1), so that the porous supports of D36 improve the oxygen conduction.

According to D36, it was known to use a porous support made of a material having ionic and electron conductivity as an air electrode in solid oxide fuel cell (column 1, lines 31-37), on which a thin, dense solid electrolyte layer was formed.

In order to increase the generated power density of a solid oxide fuel cell comprising such an air electrode, it was however necessary:

(a) to enhance the diffusion of the gas in the pores of the support material;
(b) to elevate the surface contact density at the interface between solid electrolyte, electrode and gas;
(c) to lower the resistance to ion conductivity of the solid electrolyte and electron conductivity of the electrode film (column 1, lines 47-57).

In that respect it was also known that large diameter pores in the porous electrode material would be beneficial to the diffusion of the gas but the solid electrolyte film formed thereon would not have a large contact surface density at the interface solid electrolyte-electrode-gas. Also, in porous materials of

small pores, which would produce a large contact surface density at the three-phase-interface, the diffusion of the gas into the porous electrode became large (paragraph bridging columns 1 and 2).

Therefore, to accomplish its object, D36 proposes that the pore diameter of the porous electrode on the side of one surface be smaller than that of the porous electrode on the side of the other surface (column 2, lines 24-31). In other words, the pores in the porous support are so distributed that the diameter of the pores gradually changes, for example continuously or stepwise in the direction of the thickness of the porous support (column 3, lines 20-34).

By making relatively small pores at the interface between porous support and dense layer, e.g. by using fine particles of the material for the porous support, it is possible to increase the contact surface density at the interface dense layer-porous layer-gas as well as to make the dense layer thinner; further, thanks to the relatively large pores on the other side of the porous support, i.e. the gas side in use, it is possible to lower the resistance to diffusion of gas into the support; furthermore, the mechanical strength is increased and, owing to the increased bound areas of the particles, with respect to a support with uniform pores, also the electrical resistance of the porous support is decreased (column 3, line 35 to column 4, line 6).

7.5 The arrangement of the porous layers described in D36 is designed, on the one hand, for supporting a thin dense film, and, on the other hand, for facilitating

access of the gas through the pores to the interface. That arrangement is suitable for any asymmetric membrane comprising a dense film, thus also for an ion transport membrane made of mixed conducting materials as disclosed in D1. The arrangement of layers proposed by D36 is suitable for implementing the suggestions given in D1, since it permits reduction of the pore size at the interface while distributing the pores so that gas flows unimpeded to a larger interface.

7.6 As regards the materials of porous and dense layers defined in the present claims, they do not constitute any difference from those used in D1. Even the suggestion given in D1 does not affect the materials but simply their arrangement to obtain the desired porosity and surface. Any difference in materials can play no role in the question whether the solution as claimed is obvious over D1 combined with D36.

7.7 Therefore, the skilled person aiming at providing a further use of mixed conducting membranes as described in D1, who as apparent from D33 also considers ongoing developments in the neighbouring field of solid oxide fuel cells, would be told by D36 how to implement this in an obvious way and arrive at something included in the subject-matter of claim 1 of the main request.

7.8 As claim 1 thus lacks an inventive step, the main request as a whole must be refused. For the sake of courtesy in view of the issues which were subject of debate at the oral proceedings, the Board would mention that independent claims corresponding to claims 8 and 16 as granted were also part of this main request, amended in a way analogous to claim 1 of this request.

The conclusion of the Board was that these claims too lacked inventive step on reasoning analogous to that for claim 1.

Auxiliary requests

8. The amendments to the independent claims of all of the auxiliary requests (see Point V, ***supra***) are merely formal ("about"), or aim at specifying the step of oxygen recovery to overcome the objection under Article 123(2) EPC, or concern further limitations of average pore radius and thickness of the dense layer. However, the dense layer can still have a thickness of 500 µm or of about 100 µm and the first porous layer can still be about 2 mm thick. The skilled person starting from D1, having the common general knowledge described in D33, and having regard to D36 would still arrive in an obvious manner at the subject matter of claim 1 of any of these requests, on reasoning analogous to that set out for the subject matter of claim 1 of the main request.

9. Therefore, a ground of opposition, lack of an inventive step, prejudices the maintenance of the patent as amended on the basis any of the requests now put forward.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The patent is revoked.

The Registrar: The Chairman:

S. Fabiani S. Perryman