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D E C I S I O N
of 3 August 2004

Case Number: T 0932/99 - 3.3.7

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

Patentee:
AIR PRODUCTS AND CHEMICALS, INC.

Opponent:
Praxair Technology, Inc.

Headword:
-

Relevant legal provisions:
EPC Art. 69, 108, 111, 114
EPC R. 65

Keyword:
"Appeal - admissible (yes)"
"Late filed evidence - admitted (yes)"
"Decision re appeals - remittal (yes)"
"Costs - apportionment of future costs (no) - apportionment of part of costs (yes)"

Decisions cited:
T 0156/84, T 0252/95, T 0611/90, T 0389/95, T 0416/87, T 0758/99, T 0048/00, T 0380/00, T 0715/95, T 1208/97, T 0890/00

Catchword:
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Case Number: T 0932/99 - 3.3.7

D E C I S I O N
of the Technical Board of Appeal 3.3.7
of 3 August 2004

Appellants: (Opponents)	Praxair Technology, Inc. 39 Old Ridgebury Road Danbury, CT 06810-5113 (US)
Representative:	Schwan, Gerhard, Dipl.-Ing. Schwan Schwan Schorer Patentanwälte Bauerstrasse 22 D-80796 München (DE)
Respondents: (Proprietors of the patent)	AIR PRODUCTS AND CHEMICALS, INC. 7201 Hamilton Boulevard Allentown, PA 18195-1501 (US)
Representative:	Sandmair, Kurt, Dr. Dr. Patentanwälte Schwabe, Sandmair, Marx Stuntzstrasse 16 D-81677 München (DE)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted 15 July 1999 rejecting the opposition filed against European patent No. 0592809 pursuant to Article 102(2) EPC.**

Composition of the Board:

Chairman: R. E. Teschemacher
Members: G. Santavicca
B. J. M. Struif

Summary of Facts and Submissions

I. The grant of European patent 0 592 809, in respect of European patent application 93 114 421.6, filed on 8 September 1993 and claiming a right of priority in the USA of 15 September 1992 (US 945320), was published on 12 March 1997. The patent as granted contained the following independent claims:

"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 porous layer as function of distance away from the dense layer, the porous layer 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 about 500°C."

II. A notice of opposition was filed on 10 December 1997, in which revocation of the patent was requested on the grounds of Article 100, paragraphs (a) and (b), EPC, that the claimed subject-matter lacked novelty and inventive step and that the patent did not disclose the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art. The following documents were *inter alia* mentioned:

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;

D7: EP-A-0 438 902.

III. In a decision notified in writing on 15 July 1999, which was based on the claims as granted, the Opposition Division rejected the opposition. In its decision, the Opposition Division held that:

(a) The disclosure in the opposed patent met the requirements of Article 83 EPC;

(b) D1 was the closest prior art document for the membrane of Claim 8 and D7 did not supply any information filling the gap between D1 and the opposed patent. This conclusion applied *a fortiori* to the membranes of Claims 1 and 16;

(c) as regards the alleged similarities between the field of solid oxide fuel cells (SOFC) and that of oxygen ion transport membranes (ITM), even if any similarities were accepted, they would not give a hint towards the findings on which the opposed patent was based;

(d) therefore, the claimed subject-matter had not been rendered obvious by the prior art cited.

Furthermore, according to the minutes of the oral proceedings held before the Opposition Division, the opponents had sought to introduce into the proceedings the following late-filed documents:

D9: T. Kenjo et al., "High Temperature Air Cathodes Containing Ion Conductive Oxides", J. Electrochem. Soc., Vol. 138, N°2, February 1991, pages 349-355;

D10: I.V. Murygin, "Steady Polarization of Distributed Gas Electrodes in Cells with Solid Electrolyte. The Method of Effective Coefficients", Elektrokhimiya, Vol. 23, N°6, June 1987, pages 740-747, referred to as the English translation published by Plenum Publishing Corporation, 1987, pages 697-704; and

D11: H.P. Hsieh et al., "Microporous Alumina Membranes", Journal of Membrane Science, 39 (1988), pages 221-241.

IV. On 22 September 1999, the opponents (appellants) lodged an appeal against that decision; the fee for appeal was paid on the same day. In their statement setting out the grounds of appeal, received on 25 November 1999, the appellants enclosed a number of further documents as well as three declarations of qualified experts in the technical fields concerned.

V. In reply, the proprietors (respondents) submitted a table with a new numbering of all of the documents cited, including the declarations of the experts, i.e. D1 to D47 (letter dated 6 June 2000), to which reference is made in the present decision; furthermore, they filed a first auxiliary request, in which an amended Claim 8 replaced Claim 8 as granted (letter dated 24 June 2004).

VI. Oral proceedings were held on 3 August 2004, in which the relevance of the following documents, in particular, was discussed:

D12: H. L. Tuller et al., "Doped Ceria as a Solid Oxide Electrolyte", Journal of The Electrochemical Society, February 1975, pages 255-259,

D33: J. D. Wright et al., "Advanced Oxygen Separation Membranes", Report N° TDA-GRI-90/0303, Gas Research Institute, September 1990,

D36: US-A-5 114 803,

D45: US-A-4 330 633.

VII. The appellants argued essentially as follows:

Admissibility of the appeal

(a) Although Claims 1, 8 and 16 as granted defined ion transport membranes (ITM), they encompassed similar structures used in solid oxide fuel cells (SOFC). Since the Opposition Division did not acknowledge the similarity between the fields of ITM and SOFC, the appellants had thus tried to show that it was general knowledge that those two fields were closely related. If the close interconnection of the SOFC and ITM arts were taken into consideration, and the further documents were introduced into the proceedings, the impugned decision was not tenable. The present appeal was based on one of the grounds referred to in the notice of opposition. Also, the facts, evidence and arguments presented in the statement setting out the grounds of appeal did not constitute an entirely fresh factual framework with respect

to the debate before the Opposition Division, but a mere continuation of that debate without any change of the framework. Further, the appeal had been substantiated in detail and it was not necessary that the facts argued were cogent. In the case law, one could find much more extreme cases in which the appeal was held to be admissible. In fact, even if the case was a fresh one, the appeal would still be admissible.

Inventive step

(b) As to the subject-matter of Claim 1, the closest prior art document was D1, which described an asymmetric membrane structure of a dense thin film of a perovskite-type oxide on a porous substrate. The membrane, which should be as thin as possible to increase the permeation of oxygen, as well as the porous support were in particular made of a multi-component metal oxide, which was capable of conducting both electrons and oxygen ions at temperatures greater than about 500°C. The pore radius of the porous support was about 10 to 15 micrometers. According to D1, the physical diffusion of oxygen gas through the porous body was not rate-determining. In order to further increase the permeation of oxygen it was not only necessary to reduce changes in the surface composition but also to increase the porosity of the porous support layer. More particularly, according to D1 it was effective to control the open pores in the substrate at finer size and

to increase the effective distribution concentration of fine pores. This essentially corresponded to what was defined in Claim 1 in suit. Nevertheless, D1 neither disclosed the upper limit of 10 micrometers for the pore radius nor the graduation of the pore size of the porous structure as a function of the distance away from the dense thin film.

(c) The problem underlying the patent in suit was to provide solid state membranes which exhibited superior oxygen flux. The patent in suit offered three alternative solutions to that problem, as delineated in independent claims 1, 8 and 16.

(d) However, the features defined in Claims 1, 8 and 16 did not reflect the core of the alleged invention as argued by the proprietors:

(i) Claim 1 merely required that the structure should be capable of separating oxygen from an oxygen-containing gaseous mixture, not that the separated oxygen was recovered. Since separation as such always preceded the consumption of the separated product and since the membrane structures used in SOFC applications also separated oxygen from oxygen-containing gases, Claim 1 in suit encompassed SOFC structures. Further, Claim 1 did neither define that the dense membrane had a limited thickness nor that the mixed conducting layer was contiguous to that thin dense membrane.

(ii) As to Claim 8, it did not require that the dense membrane and the contiguous porous layer should be mixed conducting. Instead, the multi-component metallic oxides forming the dense and the contiguous porous layer should conduct oxygen ions, like the solid electrolytes used in the dense layer of SOFC. According to the description of the patent in suit, the mentioned multi-component metallic oxides could also be either ion or electron conducting.

(iii) As regards Claim 16, it failed to define that each porous layer contiguous to the dense membrane had an average pore radius of less than about 10 micrometers.

(iv) Therefore, since the claimed structures did not include limitations to improve the oxygen flux, the problem underlying the patent in suit had not been solved.

(e) As regards obviousness, the claimed subject-matter merely represented an alternative to asymmetric structures comprising a thin dense membrane on a porous support, which were indeed well known before the priority date of the patent in suit. If, on the other hand, the problem could be formulated more ambitiously than providing a mere alternative, then the closely related fields of oxygen ion transport membranes (ITM) and solid oxide fuel cells (SOFC) would be considered by the skilled person when developing oxygen ion transport membranes, since he was fully aware of the developments in the field of solid oxide fuel

cells as well. Hence, the teaching of D1 could be supplemented by e.g. D36, which *inter alia* described a structure to be used as an air electrode of a solid oxide fuel cell (SOFC). That structure comprised a dense thin film of a solid oxide, which was ion conducting, and a porous layer, the pore size of which was gradually changed as a function of the distance away from the thin film. Among the materials used for the porous support, D36 *inter alia* mentioned $LaCoO_3$, which was a mixed conducting multi-component metallic oxide, as taught by e.g. D7 and D33. In the experiments described in D36, the porous layers or at least some of them had a pore size within the range as defined in Claim 1 in suit. The object of D36 was to increase the oxygen flux, like the patent in suit. Further, D36 taught that a porous support having a pore diameter that continuously changed in the direction away from the dense layer permitted the formation of a thinner dense membrane and opposed a lower resistance to diffusion of oxygen from the space where the oxygen-containing gas was present, like the patent in suit. The fact that the structure of D36 comprised a dense layer of a solid electrolyte, which was only ion conducting, did not diminish the relevance of D36. In fact, D12 showed that there was no difference whether a mixed conductor or a solid oxide was used in a fuel cell. Further, D45 described that a solid oxide film having both ion and electron conductivity, supported on a porous support, could be used either as an

oxygen membrane or as an electrode material of solid oxide fuel cells.

(f) Claim 8 in suit merely required a ion conducting thin dense layer. Since the dual transport mechanism was only related to the porous structure, it was only necessary to compare the porous structure of D1 with the porous structure of the electrode in D36, which comparison only required consideration of general physical laws that applied in the same way to both supports. Hence, not the mechanism of conduction behind the structures was important, but whether or not the porous structures described in D1 and D36 could be combined according to the problem solution approach. In order to increase the oxygen flux as suggested by D1, the skilled person would have divided the support layer of D1 in more layers, as shown in D36, so that the claimed solution was obvious. This conclusion would also apply if the contiguousness of the dense and the porous layers was additionally defined in Claim 1.

(g) In view of the closeness of the ITM and SOFC fields, as illustrated in D33, even a further restriction to the production of oxygen would not be sufficient to remove the objection of obviousness. Finally, the same conclusion also applied to Claim 8. Therefore, at least the subject-matter of independent claims 1 and 8 was rendered obvious by the combination of D1 and D36.

Apportionment of costs

(h) Since the opponents' group of experts was small, they relied on searches carried out by external sources, which produced documents D1 to D8 within the nine-month period for opposition. Further experts' opinions ordered by the opponents led to further documents filed before the oral proceedings of the first instance. After the unexpected decision of the opposition division, according to which ITM and SOFC fields were not closely related, the opponents, while trying to fill the gaps in the arguments not followed by the opposition division, became aware of further documents, e.g. D36, which was more important than D9. Many of those documents were periodicals, notoriously difficult to find, or only recently published documents. In the present case, the filing of further documents in reaction to the impugned decision did not amount to an abuse of the proceedings. Therefore, the filing of the new documents at the appeal stage was justified, and an apportionment of costs was not equitable.

VIII. The respondents argued essentially as follows:

Admissibility of the appeal

(a) The impugned decision was based on documents D1 to D8, submitted with the notice of opposition, although further documents were submitted in preparation of the oral proceedings before the

Opposition Division. In their statement setting out the grounds of appeal, the appellants still pursued original grounds of opposition but they now presented a new case based on new evidence. Since the appellants had failed to attack the legal and factual reasons of the decision, they had in effect agreed that the decision of the first instance was correct. Hence, they were now attempting to lodge a second opposition, guised in the form of an appeal, which however violated the principle of fair and expedient proceedings. According to the Case Law of the Boards of Appeal of the EPO (4th edition, 2001, VII.D.7.5.1 and 7.5.2(c)), in five relevant decisions, two rejected the appeal, because they considered that the case was a fresh one, and three admitted the appeals. In two admitted appeals, the opponents-appellants had referred to a prior use, where information was found after the first instance decision was issued. In the first case, the Board disregarded the evidence relied upon (T 389/95 of 15 October 1997). In the other case (T 252/95 of 21 August 1998), the Board admitted the new evidence but remitted the case to the first instance. In the present case, no prior use had been invoked but a set of 24 new documents, which could have been found in a database before the decision was issued, was presented. This in fact constituted a new opposition based on new evidence. Hence, admission of the appeal would open the door to successive oppositions on the same grounds invoked in the notice of

opposition, i.e. a tactical abuse. Therefore, the appeal was not admissible.

Inventive step

(b) D1, which disclosed a dense mixed conducting membrane on a porous support, both of the same material, was the closest prior art document. However, according to D1, the area accessible to oxygen at the interface between porous and dense layers would be at maximum without any support. Hence, D1 taught that porous layers reduced the accessible area, thus blocking oxygen transport. The suggestion given in the conclusions of D1, namely to disperse open pores into finer pores and to increase the effective surface area of pores, in fact went against the addition of further porous layers. Hence, the technical effects of the present invention were not disclosed in D1.

(c) The problem underlying the patent in suit was to provide membranes exhibiting superior oxygen flux without sacrificing physical compatibility and mechanical stability.

(d) The subject-matter as delineated in Claims 1, 8 and 16, which concerned a mixed conducting membrane for separating oxygen from oxygen-containing gases to obtain pure oxygen as the desired product, overcame the flux limitations which had been observed when using known membranes. In fact, the improved flux of gas through the membrane had been achieved by a

structure comprising a thin dense membrane supported on porous mixed conducting supports, which structure was able to withstand the pressure gradient applied. The proprietors had indeed realized that, in order to overcome the flux limitations, in particular the surface kinetic limitations as well as the bulk limitations, not only the dense membrane should be made thinner but the contiguous layers should be mixed conducting and have a gradually rising pore size as a function of distance away from the membrane, which pores size should however be less than 10 micrometers. This permitted the permeation of the oxygen ions also through the bulk of the contiguous porous layers. The fact that the dense layer was thin was implied by the term membrane.

(e) On the proper interpretation of Claims 1, 8 and 16 on the basis of the description, those allegedly missing features mentioned by the appellants should be read into the claims. Hence, in Claim 1, the porous layer was contiguous to the dense layer; in Claim 8, the multi-component metallic oxides were mixed conducting; and in Claim 16, both porous layers had an average pore radius of less than 10 micrometers. Further, from the description, it was apparent that the separated oxygen was the desired product. Since the claimed structures surprisingly showed better fluxes than membranes having a single porous layer, whether mixed conducting or not, the problem had been solved.

(f) None of the freshly submitted documents was prejudicial to the maintenance of the patent in suit. Therefore, all belatedly filed documents should be disregarded. In particular, the art of solid oxide fuel cells (SOFC), relied upon by the appellants, did not neighbour on the art of ion transport membranes (ITM). In fact, the combinations of documents used by the appellants to attack the subject-matter of Claims 1, 8 and 16 were essentially based on hindsight, and in any case failed to render obvious the claimed membranes.

(g) More particularly, the membrane problems mentioned in D1 could not be mixed with the electrode problems mentioned in D36. It was known that a solid oxide fuel cell required a dense layer of ion conducting solid oxide as well as electrodes on both sides of the ion conducting oxide, to conduct the electrons. In contrast thereto, since in an ion transporting membrane the conduction of the electrons was within the membrane itself, the membrane did not require any electrode. Therefore, the structures were not similar. Nor did D33 or D45 support any alleged similarity between the fields of SOFC and ITM. The reason why a mixed conducting membrane was used for some electrodes was that they provided electrons to reduce oxygen gas, but only at the free face boundary where the oxygen ion was formed and transported. This was not the case in D36, wherein oxygen flux limitations occurred to

electrodes at the (solid oxide-electrode-gas) three-phase interface. No three-phase interface was however present in D1.

(h) Further, the problem defined in D36 was how to enhance the diffusion of oxygen, which was favoured by large pores, while increasing the surface contact density of the three-phase interface, which was favoured by small pores. This problem did not exist in ITM membranes.

(i) Therefore, without hindsight, the skilled person had no motivation to combine an ITM structure of D1 with a SOFC electrode structure of D36, or of any other new evidence.

Apportionment of costs

(j) In their notice of opposition, the appellants had only cited documents D1 to D8. Before the oral proceedings, they had then sought to introduce some belated documents into the proceedings. In the statement setting out the grounds of appeal, further documents were cited. However, the further documents sought to be introduced at the appeal stage did not relate to something which could not have been established before. Hence, in the present case, no mitigating circumstances were present which could justify the lateness of the submission. The introduction of the new documents at the appeal stage amounted to a tactical abuse of the proceedings. If, however, an abuse was not acknowledged, and if any documents, such as D36,

D33 or D45, was found to be sufficiently relevant to prejudice maintenance of the patent as granted, then the case should be remitted to the first instance. Since the lateness of filing new documents was not justified, at least the costs of the appeal level and the costs of the second first instance proceedings should be charged to the appellants, as decided in T 611/90 (OJ 1993, 50) and T 416/87 (OJ 1990, 415).

IX. The appellants (opponents) requested that the decision be set aside and that the European patent be revoked. In addition they requested to admit into the proceedings at least documents D9, D10 and D12 already cited during the first instance proceedings as well as freshly submitted documents D29, D33 and D36; auxiliarily, to remit the case to the first instance for further prosecution. Finally, they requested to reject the respondents' request for apportionment of costs.

X. The respondents (proprietors) requested that the appeal be rejected as inadmissible; auxiliarily, that the appeal be dismissed and that the patent be maintained as granted, or, alternatively on the basis of the auxiliary request submitted with letter dated 24 June 2004. In addition, they requested to reject the newly submitted references as filed belatedly, or, should one of the new references be considered as sufficiently relevant, to remit the case to the first instance and to apportion the respondents' costs incurred by these appeal proceedings.

Reasons for the Decision

1. *Admissibility of the appeal*

1.1 The respondents have argued that the present appeal was based on new, belatedly filed evidence, which was available before the impugned decision was issued, as well as on new facts based on that new evidence. Hence, it constituted a fresh case which was inadmissible.

1.2 From the statement setting out the grounds of the present appeal, it is apparent that:

1.2.1 the decision is alleged to be incorrect because an argument of crucial importance, i.e. that the solid oxide fuel cell (SOFC) art be taken into account when assessing inventive step, was not followed by the Opposition Division (statement, point 1.1);

1.2.2 further, it is the appellants' position that the claimed membranes were rendered obvious by the known art (counterarguments referred to as points 2(a), 2(b) and 2(c) on pages 8 and 9 of that statement); and,

1.2.3 finally, the facts on which the appellants based their arguments are detailed extensively on the basis of specific combinations of documents on pages 11 to 46, whereby each attack is concluded by a graphic presentation showing which feature is known from the document taken as prior art and which feature is to be found in the other document referred to.

1.2.4 Therefore, the present statement of grounds of appeal sets out an arguable case as required in the established practice (Case Law, s*upra*, VII.D.7.5.1).

1.3 As to the lateness of the new evidence, the question whether late submissions are disregarded under Article 114(2) EPC cannot as a rule be answered without starting the substantive examination of an appeal. This presupposes that the admissibility of the appeal has been accepted before. Therefore, the possibility that facts and evidence submitted for the first time with the grounds of appeal may be disregarded does not affect the admissibility of the appeal.

1.4 It follows from the above that the present appeal is admissible.

Main request

2. *Sufficiency of the disclosure*

The appellants have not maintained their grounds of insufficient disclosure during the appeal proceedings. According to the decision under appeal the opposed patent met the requirements of Article 83 EPC. The Board has no reason to take a different position.

3. *Late filed facts and evidence*

3.1 In view of the requests presented during the oral proceedings, the only point dealt with was whether or not any of the documents filed for the first time at the appeal stage was relevant, in particular D36, although also D12, D33 and D45 were discussed.

3.2 All these documents could have been submitted before, i.e. during the nine-month period for opposition, or before the oral proceedings for consideration by the Opposition Division. Therefore, they are late filed.

3.3 According to the Case Law, *supra*, VI.F.2, in particular landmark decision T 156/84 (OJ 1988, 372), the admissibility of late filed documents in the proceedings is, unless there is an abuse of the proceedings, in particular decided with respect to their relevance.

4. As regards the reasons for the lateness, the Board has considered that:

(a) The appellants provided plausible reasons for submitting the material so late, in particular that the new evidence was submitted in reaction to the impugned decision, in which the arguments of the opponents concerning the similarity between the ITM and SOFC fields, which similarity was held to be crucial, had not been followed.

(b) The respondents could not be surprised by the submission of new documents relating to an argument discussed during the opposition proceedings, which however had not persuaded the Opposition Division.

4.1 Therefore, the submission of the new evidence cannot be considered as a tactical abuse of the proceedings. Consequently, the new evidence cannot be disregarded without considering its relevance.

4.2 As regards the relevance, the main criterion for deciding on the admissibility of a late-filed document is its evidential weight in relation to other documents already under consideration in the case so that it may change the outcome of the case (Case Law, *supra*, VI.F.3.1.1). The new documents have *inter alia* been filed to supplement the disclosure of D1 when arguing on inventive step. Before considering their relevance, it is therefore necessary to assess the content of independent Claims 1, 8 and 16 in suit as well as the disclosure of D1.

4.3 Since there has been considerable divergence of opinion between the parties on the content of Claims 1, 8 and 16, the meaning of these claims has to be made clear:

4.3.1 Claim 1 concerns a membrane capable of separating oxygen from an oxygen-containing gaseous mixture. Thus, the claimed subject-matter is directed to a product per se, i.e. a structure in form of a membrane, a wall having a thin structure. Claim 1 is silent as to any recovery of the separated oxygen. The membrane as such cannot separate any gas, unless it is installed in an apparatus comprising compartments for the oxygen-containing gas and for the separated gas, which are tightly separated by that membrane, under conditions appropriate for the separation. Claim 1 does not define any such compartments, let alone any apparatus for gas separation comprising those compartments and means for applying the necessary conditions, such as temperature and pressure gradient. In fact, Claim 1 only defines the structure of the membrane as such, independently from its installation in any apparatus. Therefore, the

indication in Claim 1, i.e. "capable of separating oxygen from an oxygen-containing gaseous mixture", merely serves the purpose of defining a capability of the claimed membrane, without imparting any limitations on any actual use of the structure claimed, such as recovery of pure oxygen.

Further, although it is not contested that a membrane has a thin structure, Claim 1 nevertheless does not define the thinness of the structure, nor does it define that the porous layer having the smallest pore size is contiguous to the dense layer. Hence, those missing features cannot be read into Claim 1.

Also, since Claim 1 mentions that the dense and the porous layer are "independently" formed from a mixed-conducting multi-component oxide, it follows that in the structure of Claim 1 the dense and the porous layers may be present as two distinct phases. Consequently, the presence of a three-phase interface (dense layer-porous layer-gas) is not excluded by the wording of Claim 1.

4.3.2 The respondents have argued that if Claim 1 was interpreted in the light of the description, those limitations would be apparent, in particular that the porous layer having the smallest pore size was contiguous to the dense layer.

4.3.3 However, a distinction should be drawn between, on the one hand, the fact that it might be necessary to take into account any explicit definition as given in the description for interpreting a claim's term and, on the other hand, the tentative to use Article 69 EPC as a

basis for reading limitations derived from the description into claims in order to avoid objections based on lack of novelty or inventive step. The latter approach to claim interpretation by the respondents, whereby features mentioned only in the description are read into Claim 1 as necessary limitations is incompatible with the EPC (T 1208/97 of 3 November 2000, not published in OJ EPO; Reasons, point 4).

It follows from the above that the limitations invoked by the respondents (oxygen gas is the recovered product of the separation; since the membrane is thin per definition, the dense layer is much thinner; and, the porous layer with the smallest pore size is contiguous to the dense layer) cannot be read into Claim 1.

4.3.4 As to Claim 8, in addition to the comments on Claim 1 which are applicable mutatis mutandis, attention is drawn to the following:

(a) The second porous layer, which is defined to be "not a mixed conducting oxide", may be only ionic conducting, or only electron conducting, or inert;

(b) furthermore, the multi-component metallic oxides are required to be capable of conducting oxygen ions at the specified temperature. Consequently, those multi-component metallic oxides need not be mixed conducting either. Therefore, the dense layer may be only ion conducting.

4.3.5 With respect to Claim 16, in addition to the comments on Claim 1 which are applicable mutatis mutandis, the question arises whether or not the limitation recited in Claim 16 "having an average pore radius of less than about 10 micrometers" applies only to the second porous

layer, as argued by the appellants, or to both porous layers, as argued by the respondents.

In this respect the following is noted: The term radius is singular; no comma is present after "second porous layer"; no word like "each" or "any" is present before "having ... 10μm"; the limitation "having ...10μm" plainly follows the definition of the second porous layer; the claimed construction is such that the two porous layers are separated by the membrane, such that they might have different porosities. Therefore, the above limitation applies only to the second porous layer.

4.4 The decision under appeal and the parties have considered D1 as suitable starting point for assessing inventive step.

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

According to D1, when a dense film of mixed conductive perovskite type oxides is in the form of a membrane and different oxygen partial pressures are applied on either sides, oxygen permeates from the high oxygen partial pressure side to the low pressure side at temperature higher than 500°C. Oxygen molecules are ionized on the high oxygen partial pressure 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 authors of D1 thus considered that an asymmetric structure with a thin film formed on a porous substrate which contains through holes (Figure 1) was a potential thin film-type oxygen permeation device, which had good mechanical strength and was suitable for the manufacture of large surface area products (page 523, right column, first full paragraph, first sentence).

To develop such an oxygen semipermeable membrane having an asymmetric structure using mixed conductive perovskite-type oxides, the preparation of a dense $La_{0.6}Sr_{0.4}CoO_3$ (LSCO) film on a porous LSCO substrate was studied by means of *inter alia* spray deposition-techniques (Abstract). The porous LSCO substrate had open pores in size of 20 to 30 μm, i.e. a radius of 10 to 15 μm (page 524, paragraph 3-1).

Using the samples with a thin dense film layer (about 15 μm thick) formed by the spray deposition technique and comparative samples of a dense sintered disk (about 2 mm thick; page 524, point 2.1), the rate of oxygen permeation was measured (Figure 9 of D1).

4.4.2 Although the rate of oxygen permeation of the thin film element was more than twice as high as that of the sintered disk sample, it nevertheless was around 1/5 of an expected value which had been estimated from an equation (1) in D1 (page 528). 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.

4.4.3 One of the reasons for the much lower rate of oxygen permeation was found to be the change in surface composition of the samples caused by firing at high temperature (sintering), which caused a deterioration in oxygen adsorption and desorption, which in turn reduced the rate of oxygen permeation (page 528, right column, penultimate paragraph).

4.4.4 Further, in order to increase the rate of oxygen permeation of the thin film element, it was found necessary to prepare a thinner dense film as well as to increase the porosity of the substrate (the porosity corresponded to "p" in equation (1) of D1).

4.4.5 To increase the porosity of the substrate, enlargement of the pore size was not recommended, because it made it more difficult to form dense thin films on it. Instead it was effective to make the open pores of the substrate finer as well as to increase the distribution concentration of the fine pores (paragraph bridging pages 528 and 529).

4.4.6 Therefore, in order to further increase the rate of oxygen permeation through a thin film element supported on a porous substrate, D1 suggests, *inter alia*, to make the open pores of the substrate finer as well as to increase the effective surface area of the pores (conclusion (4) on page 529).

4.4.7 However, any concrete realisation of that suggestion is missing in D1.

5. The subject-matter of Claim 1 of the patent in suit is distinguished from the asymmetric membrane disclosed in

D1 by the following features: the upper limit of 10 μm for the average pore radius; the graduation of the pore size in the porous layers as a function of distance away from the dense layer.

6. According to the decision under appeal, none of the documents cited during the opposition proceedings could fill the gap between D1 and the claimed subject-matter. Hence, the question arises whether or not any of the documents discussed during the oral proceedings before the Board is relevant in that respect, in particular D36, which was the subject of a long discussion.

6.1 D36 *inter alia* concerns 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, a diameter of 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 a thickness of the electrode from the side of interface to the side opposite to the interface (Claim 12).

Further particulars of said porous electrode are specified in Claims 13 to 18 of D36.

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. It is apparent from the above, that the pore radius of all the layers of the support of Experiment II as well as that of a number of layers of the support of Experiment III, is less than 10 μm.
Compared to the porous support of Experiment I, with uniform average pore diameter, the electrical resistance of a porous support with graduated pores was reduced (column 7, lines 5 to 48; table 1). Thus, the porous support of D36 appears to improve the conduction.

According to D36, the use of a porous support made of a material having ionic and electron conductivity as an air electrode in SOFC was known (column 1, lines 31 to 37). On that porous material a thin, dense solid electrolyte layer was formed.

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

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

In that respect it was known that, although large diameter pores in the porous electrode material would be beneficial to the diffusion of the gas, the solid electrolyte film formed thereon would not have a large contact surface density at the interface solid electrolyte-electrode-gas; on the other hand, in porous materials with 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 the following solution: the pore diameter of the porous electrode on the side of one surface is smaller than that of the porous electrode on the side of the other surface (column 2, lines 24 to 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 to 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 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, compared 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).

As regards the materials used for making the porous support, D36 mentions $LaMnO_3$, $CaMnO_3$, $LaNiO_3$, $LaCoO_3$ and $LaCrO_3$, doped or not. All of them have a perovskite structure (i.e. a structure ABO_3, where A and B are metal atoms, i.e. cations). It has not been contested by the respondents during the oral proceedings that at least $LaCoO_3$ was mixed-conductive, which fact resulted from other documents like D7 and D33. This fact, would indeed be in line with the statement in D36 that the air electrode, i.e. the porous support of the solid electrolyte film, was ion and electron conducting (column 1, lines 33 to 34).

6.2 The Opposition Division had not admitted into the proceedings the late filed documents relating to the SOFC art, e.g. D9 to D11 (Minutes of the oral proceedings before the Opposition Division, page 5, third last paragraph). The respondents have argued that the fields of ITM and SOFC are not related, such that the skilled person would not have considered the disclosure of D36, which concerned an air electrode, to improve a membrane according to D1. The following documents relating or also relating to the SOFC art have been discussed during the oral proceedings before the Board:

6.2.1 D45 concerns a solid electrolyte having high electron conductivity and high oxide ion conductivity, which is composed of a sintered body consisting substantially of (a) 5 to 85 mole% of an oxide of cobalt, (b) 2 to 70 mole% of an oxide of at least one metal selected

from strontium and lanthanum, and (c) 13 to 80 mole% of an oxide of at least one metal selected from bismuth and cerium (Claim 1). That solid electrolyte preferably has an electron conductivity of at least 10^{-2} $ohm^{-1}xcm^{-1}$ (Claim 2) and an oxide ion conductivity of at least 10^{-4} $ohm^{-1}xcm^{-1}$ (Claim 4).

The above solid electrolyte, which has high electron conductivity and oxide ion conductivity and very good oxygen semipermeability, is suitable as a material for an oxygen perm-selective membrane and, for that purpose, it is advantageously used as a thin film having a thickness of generally 10^{-3} to 10^{+4} μm (column 9, lines 10 to 12 and 29 to 35).

The solid electrolyte of D45 can not only be used for selective separation of oxygen from the air, but also as an electrode material such as an electrode on the air pole side of high-temperature solid electrolyte fuel cells (column 10, lines 34 to 39).

While conventional solid electrolytes have oxide ion conductivity alone and for use in oxygen separation, electron conductivity must be imparted thereto by, for example, providing an electrode and an external circuit on both sides of the solid electrolyte, the solid electrolyte membrane of D45 has the advantage that it has both electron and oxide ion conductivity, does not particularly require an electrode nor an external circuit and can be used as such as an oxygen separating membrane (column 10, lines 40 to 49).

6.2.2 D33 concerns advanced oxygen separation membranes (title). In particular, D33 compares the structures of

electrically-driven and pressure-driven solid electrolyte oxygen separation membranes (Figure 5.1), whereby the electrically driven membrane is said to be essentially a high-temperature, solid oxide fuel cell in reverse (page 34, second paragraph), and also deals with the theory of oxygen-ion conduction (points 5.1) and the membrane properties (points 5.2). In particular, D33 mentions that perovskites having the general formula ABO_3, where A and B are metal atoms (cations), such as $LaCoO_3$, are mixed-conductive (paragraph bridging pages 34 and 35; page 37, last paragraph). This appears to confirm the disclosure of D7 (e.g. page 10, line 26).

Several statements concerning similarity between solid oxide fuel cells and ionic oxygen separation membranes can be found in D33, in particular in respect to similarity of materials and fabrication techniques used in both fields (page 40, point 5.3, first paragraph, third sentence; page 41; last sentence of the first paragraph and third sentence of the second paragraph; page 44, second full paragraph, first sentence, and last paragraph, first sentence; page 62, point 5.4.4). In particular, D33 mentions that "solid electrolyte oxygen separation will benefit greatly from research on solid oxide, electrolyte fuel cells" (page 45, first full paragraph, first sentence).

As regards the flux through pressure driven composite membranes, D33 discloses that as the conductivity of the electrolyte is decreased, mass transfer through the electrode and the support tube rapidly becomes dominant (page 51, third full paragraph, second sentence). The use of mixed conductors eliminates the need of an electrode, thus simplifying fabrication and eliminating

the mass-transfer resistance of the air-side electrode. Nevertheless, the models for mixed conductors and ionic conductors are identical (page 54, first paragraph).

6.3 As regards the material and structural similarities between ion transport membranes and air cathodes of SOFC, the respondents have declared that: "As far as mixed conducting multi-component metallic oxides are employed in the SOFC art, they are employed as cathode materials and not as electrolyte materials", (response to the notice of appeal, dated 6 June 2000, point 7.3.1, page 19, first full paragraph, last sentence).

6.4 Taking into account the above facts, and without wishing to bind the Opposition Division in the assessment of the case, the Board however considers the following points to be of relevance for the further prosecution:

6.4.1 whether or not the structure of the porous support described in D36 essentially corresponds to the structural features of the support which distinguish the membrane of Claim 1 from the disclosure of D1;

6.4.2 whether or not the skilled person would fill that gap by technical information according to D36, after considering:

(a) whether or not similarity between ITM and SOFC fields, if any, is apparent from D33;

(b) whether or not, from the analysis of D33, the skilled person working on pressure driven membranes would consider the ongoing development in the field of SOFC, in particular of the air

cathode, to benefit from them (page 62, last paragraph, first sentence);

(c) whether or not a structural and material similarity between electrical- or pressure-driven oxygen separation membranes and the membrane structures used in SOFC, as discussed above, in particular for the air cathodes, provides an incentive to combine the teachings of documents D1 and D36, in order to put into practice the suggestion given in D1;

(d) whether or not D45 gives weight to the argument (see point 6.1, seventh paragraph, *supra*), on the one hand, that the air electrode of a SOFC can be mixed-conducting and, on the other hand, that oxygen-ion transport membranes have structural and material similarities with an air electrode for a solid oxide fuel cell.

6.4.3 In that respect, the Opposition Division should also consider:

(a) whether or not the problem to be solved in the porous structure of D1 and the problem solved by the porous structure of D36 are similar;

(b) whether or not the problem underlying the patent in suit has been solved by the features in Claims 1, 8 and 16;

(c) whether or not the modification of the porous structure of the membrane of D1 along the structure of the porous support of the cathode of D36 is in contradiction with the teachings in D1 and D36 and whether or not that combination plainly follows from the problem solution approach;

(d) whether or not the solution of the problem underlying the claimed subject-matter is obvious

in view of the teachings of D1 and D36 when applying general principles, applicable to both fields, governing, on the one hand, the flux of gas through porous media and the mass-transfer function of the porous media (such as the ease of the access of the gas to the dense layer and the ease of diffusion of the ions to the dense layer), and, on the other hand, safeguarding the possibility of making a thin dense layer and the mechanical stability of the structure.

6.5 From the above if follows that a number of new elements give weight to the argument that the field of SOFC neighbour on the field of ITM, such that the documents discussed during the oral proceedings, i.e. D12, D33, D36 and D45, particularly D36, are more relevant than the documents considered in the impugned decision, in order to fill the gap between D1 and the claimed subject-matter.

6.6 Therefore, D36 is admitted into the proceedings because it might lead to revocation or limitation of the patent in suit (Case Law, *supra*, VI.3.1.1). The same applies to the further documents which have been discussed during the oral proceedings before the Board. The admission of any of the further late-filed documents is left to the discretion of the Opposition Division, upon consideration of the relevance thereof.

6.7 All of the documents discussed during the oral proceedings before the Board, apart from D1, D7 and D9, have been submitted at the appeal stage. Both parties have requested to remit the case to the first instance. The Board finds it appropriate that the assessment of

inventive step based on new evidence is carried out by the Opposition Division. Therefore, the Board has come to the conclusion:

(a) On the one hand, to take into account the late filed documents submitted at the appeal stage and discussed during the oral proceedings, on the basis of its discretionary power under Article 114(1) EPC, which ensures that the proceedings be conducted in the interests of the parties, the public and the EPO (Article 11(3) of the Rule of Procedure of the Boards of Appeal); and,

(b) on the other hand, to remit the case to the department of first instance, on the basis of its discretionary power under Article 111(1) EPC.

7. *Apportionment of costs*

7.1 As said above (point 4), the late filing of evidence was not an abuse of the procedure. Nevertheless, evidence has been submitted for the first time in the opposition appeal proceedings which could have been filed during the first instance proceedings. If that evidence had been filed early, the Opposition Division would have considered it.

7.2 Further, in view of the number of items of evidence submitted belatedly, the examination of the further evidence had to be concentrated on the most relevant items, i.e. only few documents have been considered during the oral proceedings before the Board. Hence, it remains to be examined whether or not other items of evidence not yet considered are relevant as well.

7.3 Since relevant evidence was admitted into the proceedings, and since both parties have requested to remit the case to the first instance, if a document was relevant, the Board considered the remittal appropriate.

7.4 Since all of the relevant late-filed documents were admitted at the appeal stage, cost of that appeal stage could have been avoided, if the documents had been filed in time.

7.5 Therefore, it is equitable in the present case to apportion part of the costs incurred by the respondents, such that the appellants bear the costs as defined in the order (Case Law, *supra*, VII.C.12.3, in particular T 416/87).

7.6 The Board did not consider it appropriate to apportion the costs of the future proceedings before the Opposition Division, as requested by the respondents. Future costs depend on the course of the future proceedings, in particular on the course of action by the parties. An award of future costs is open-ended, and its consequences are unpredictable. As any discretionary decision, the apportionment of costs requires the consideration of all relevant circumstances. In the present situation, the most relevant factors, i.e. the dimension of the costs and whether they have been incurred in an appropriate manner, are not yet known.

7.7 This decision is in agreement with T 758/99 of 25 January 2001 (point 5 of the reasons), T 48/00 of 12 June 2002 (point 14 of the reasons) and T 890/00 of 28 October 2002 (point 5 of the reasons), all deviating

from the older judgements given in T 611/90 (point 5 of the reasons) and T 715/95 (commented in Case Law, *supra*, VII.C.12.4).

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 for further prosecution.

3. The costs shall be apportioned so that the appellants shall pay the respondents (a) the costs charged by the respondents' European professional representative to the respondents in connection with the present appeal proceedings; and (b) the expenses (travelling, accommodation) for the two participants of the respondents at the oral proceedings before the Board.

The Registrar: The Chairman:

C. Eickhoff R. Teschemacher