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D E C I S I O N
of 28 May 2004

Case Number: T 0848/01 - 3.3.5

Application Number: 96103979.9

Publication Number: 0732306

IPC: C01G 51/00

Language of the proceedings: EN

Title of invention:

Novel composition capable of operating under high carbon dioxide partial pressure for use in solid-state oxygen procuding devices

Patentee:

AIR PRODUCTS AND CHEMICALS, INC.

Opponent:

Praxair, Inc.

Headword:

Multicomponent metallic oxides/AIR PRODUCTS

Relevant legal provisions:

EPC Art. 54, 56

Keyword:

"Novelty - yes"

"Inventive step - yes, non obvious solution"

Decisions cited:

T 0198/84

Catchword:

-



Case Number: T 0848/01 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 28 May 2004

Appellant: AIR PRODUCTS AND CHEMICALS, INC.
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Decision under appeal: Interlocutory decision of the Opposition
Division of the European Patent Office posted
23 May 2001 concerning maintenance of European
patent No. 0732306 in amended form.

Composition of the Board:

Chairman: M. M. Eberhard
Members: E. O. Wäckerlin
S. U. Hoffmann

Summary of Facts and Submissions

I. The appeal is from the decision of the opposition division dated 23 May 2001 concerning the maintenance of the European patent No. 0 732 306 in amended form according to the set of claims of the auxiliary request submitted on 2 March 2001.

II. The two independent claims as granted read as follows:

"1. A composition of matter represented by the formula $\text{Ln}_x\text{A}'_x\text{A}''_x\text{B}_y\text{B}'_y\text{B}''_y\text{O}_{3-z}$, wherein Ln is an element selected from the f block lanthanides, A' is selected from Group 2, A'' is selected from Groups 1, 2 and 3 and the f block lanthanides, and B, B', B'' are independently selected from the d block transition metals, excluding titanium and chromium, wherein $0 \leq x < 1$, $0 < x' < 1$, $0 \leq x'' < 1$, $0 < y < 1.1$, $0 < y' < 1.1$, $0 < y'' < 1.1$, $x + x' + x'' = 1.0$, $1.1 > y + y' + y'' > 1.0$ and z is a number which renders the compound charge neutral wherein such elements are represented according to the Periodic Table of Elements adopted by IUPAC."

"3. A composition of matter represented by the formula $\text{La}_x\text{A}'_x\text{A}''_x\text{Co}_y\text{Fe}_y\text{Cu}_y\text{O}_{3-z}$, wherein A' is selected from strontium, barium, calcium or magnesium, A'' is selected from Groups 1, 2 and 3 and the f block lanthanides, wherein $0 \leq x < 1$, $0 < x' < 1$, $0 \leq x'' < 1$, $0 < y < 1.1$, $0 < y' < 1.1$, $0 < y'' < 1.1$, $x + x' + x'' = 1.0$, $1.1 > y + y' + y'' > 1.0$ and z is a number which renders the compound charge neutral."

III. During the opposition proceedings, *inter alia* the following prior art documents have been cited:

- D1: N. Q. Minh, "*Ceramic fuel cells*", Journal of the American Ceramic Society 76 [3], 1993, p. 563-587.
- D2: E. Bergsmark et al., "*On the stability of $La_{0.8}Sr_{0.2}MnO_3$* ", In: Proc. of the 2nd int. symposium on solid oxide fuel cells, Athens 1991, EUR 13564, p. 473-478.
- D4: US-A-5 306 411
- D5: WO 94/24065
- D6: J. Mizusaki et al., "*Nonstoichiometry and defect structure of the perovskite-type oxides $La_{1-x}Sr_xFeO_{3-\delta}$* ", Journal of Solid State Chemistry 58, 1985, p. 257-266.
- D7: US-A-5 273 628
- D8: English translation of: H. Yokogawa et al., "*Simultaneous thermogravimetry and mass spectrometry on perovskite-type oxides for solid oxide fuel cells*", 30th Annual Meeting of the Japan Society for Calorimetry and Thermal Analysis, No. 1A13 (D-8), October 1994, p. 44-45.
- D9: US-A-4 748 143
- D14: US-A-5 240 480

In the impugned decision the opposition division stated that the composition according to claim 1 as granted was novel in respect of the prior art, in particular with regard to documents D4 and D9, respectively. The opposition division held, however, that this composition was not based on an inventive step, in contrast to the composition according to claim 1 according to the auxiliary request, which claim is identical to claim 3 as granted.

Therefore the opposition division decided that the patent could be maintained on the basis of the claims in accordance with the auxiliary request.

IV. With his notice of appeal the appellant (proprietor) sought the maintenance of the patent as granted as a main request and submitted a set of amended claims as an auxiliary request. This request was withdrawn at the oral proceedings which were held on 28 May 2004. The appellant stated that the claims as granted had to be construed as requiring the presence of three different B-site transition metals. Any other construction of the claims was technically not meaningful. Concerning inventive step, he argued that, although the examples contained in the description related to a specific combination of B-site elements, namely Co, Fe and Cu, the technical teaching of the patent was more general and the scope of claim 1 was a reasonable generalization of the examples. The respondent had not presented any evidence that other combinations of elements did not lead to the stability improvement which results from the B-site richness and the presence of three different B-site elements. The stability of the claimed compositions under operating conditions, viz. at temperatures of at least 600°C and high partial pressures of carbon dioxide and water, was a surprising improvement. None of the documents D14, D4, D1 and D6 suggested the use of an over-stoichiometry of the B-site elements in order to solve the stability problem in oxygen producing devices.

V. The respondent (opponent) contested the appellant's argumentation. He submitted, *in essence*, the following:

The wording of claim 1 as granted is unclear because of the expression "are independently selected". Claim 1 does not unambiguously call for three different B-site elements B, B' and B", respectively. In conformity with certain passages contained in the description the presence of two different metals or a mixture of two different metal oxides appeared to be sufficient.

Moreover claim 1 is too broad in scope. It embraces a large number of compositions having different combinations of $\text{Ln}_x\text{A}'_x\text{A}''_x$ and $\text{B}_y\text{B}'_y\text{B}''_y$. However, only one single combination of B-elements, namely CoFeCu, in connection with only three different combinations of $\text{Ln}_x\text{A}'_x\text{A}''_x$, namely LaBa, LaSr and Sr, are exemplified in the description, and a technical advantage has been shown only for the specific combination of CoFeCu. In the respondent's view it is not credible to assume that all variants covered by claim 1 exhibit the improvement. It was the appellant's task to show that the appealed decision is not correct in this respect.

An objection of lack of novelty arises against claim 1 having regard to documents D1, D2, D4, D5, D6, D7 and D9, respectively.

In particular D4 destroys the novelty of the subject-matter of claim 1, even if claim 1 is interpreted in the narrow sense, viz. as embracing exclusively compositions containing three different B-site elements. D4 describes mixed metal oxides comprising two A-site and three B-site elements. As far as the ratio of the B-site to the A-site elements is concerned, D4 discloses a range of 0.9 to 1.1, which includes the

entire B-site rich sub-range as defined in claim 1, i.e. $1.1 > y + y' + y'' > 1.0$. In view of the overlap with the known range the claimed sub-range cannot be regarded as being novel. The criteria developed in decision T 0198/84 for selection inventions are not all fulfilled.

Claim 1 also lacks novelty in respect of D9, where a composition represented by the formula $AA'BO_3$ is disclosed. According to D9 the transition metal B may be iron, cobalt and nickel, **or mixtures thereof**. Therefore the combination of iron, cobalt and nickel for B, B' and B'', respectively, is encompassed by D9. The surface atomic ratio of the B-site to the A-site elements is in the range from 1.0 : 1.0 to 1.1 : 1.0.

Irrespective of the question of novelty, the respondent submitted that claim 1 lacks an inventive step in respect of D4 alone, or D14 in combination with any of the documents D1, D6 or D8.

D4 reveals materials comprising three different B-site elements. Moreover the examples contained in D4, although stoichiometric at first sight, extend to the non-stoichiometric range if the contents of the B-site elements are expressed using two decimals (as in the patent in suit) instead of one decimal in D4.

Taking D14 as the starting point, and confronted with the problem of increasing the stability of the perovskite material under operating conditions, the skilled person would have found a pointer to the solution in document D1 which discloses that "lanthanum deficiency" of the material, in other words "B-site richness", increases the stability when used in solid-

oxide fuel cells, because it avoids undesirable hydration. As in the case of D4 the seemingly stoichiometric materials extend in reality to the non-stoichiometric range when two decimals are used to express the contents of the B-site elements.

D6 provides an incentive to use an over-stoichiometric amount of B-site elements, since according to D6 a slight excess of iron in sintered oxides of the formula $\text{LaFeO}_{3-\delta}$, i.e. B-site richness, gives rise to increased stability. As iron usually contains a certain amount of Ni and Co as impurities, the skilled person would arrive at a compound having three different B-site elements.

D8 reveals that a B-site rich material, namely $(\text{La}_{0.7}\text{Sr}_{0.3})_{0.9}\text{MnO}_3$, is more stable at temperatures as high as 1500°C than an A-site rich material, i.e. $(\text{La}_{0.7}\text{Ca}_{0.32})\text{CrO}_3$.

Thus all three documents D1, D6 and D8 demonstrate that B-site rich materials have improved stability over stoichiometric and A-site rich materials. The skilled person would take this teaching into consideration and, thus, arrive at the compositions according to claim 1.

VI. The appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of Claims 1 to 9 as granted; pages 2, 3, 5 to 8, 10 of the description and figures 1 to 9 as granted; and pages 4, 9, 11 and 12 of the description as amended during the oral proceedings.

The respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

Construction of claim 1 - number of distinct "B-site elements"

2. The number of different B-site elements is not expressly stated in claim 1. However the formula contained in claim 1 gives a clear indication how claim 1 must be construed properly. In fact all three coefficients y , y' and y'' of the B-site elements B, B' and B'' are greater than zero. This implies that the elements B, B' and B'' are mandatory components of the claimed compositions, and in order to be distinguishable from each other, they have to be different.

- 2.1 The Board cannot agree with the argument submitted by the respondent, according to which the scope of claim 1 extends to embodiments where the number of distinct B-site elements is only two in view of the description. Even if it is stated in claim 1 that the elements B, B' and B'' are "*independently selected*", this does not mean that one and the same "B-site element" may be selected more than once, as the respondent contends. In the Board's view and as indicated by the appellant the expression "*independently selected*" has to be construed as meaning that, once a specific element B has been selected, any **other** d block transition metal, except titanium and chromium, may be selected as element B'. Likewise, any transition metal **other** than those chosen as B and B' may be selected as element B'' (except Ti

and Cr). In other words no specific combination of B-site elements is required. Any combination of d block transition metals, with the exception of titanium and chromium, is possible, provided that the number of different B-site elements is three, as set out in the formula contained in claim 1. This construction of claim 1 is in agreement with the description of the invention in the patent in suit (see column 4, line 10 to column 5, line 12; column 6, line 38 to column 14, line 31) and with the examples thereof which all disclose compositions containing three different transition metals Fe, Co and Cu. As pointed out by the respondent, in the description reference is also made to *"layers of the membranes ... comprising an oxide of **at least two** different metals or a mixture of **at least two** different metal oxides"* (see column 15, lines 40 - 46; emphasis added). At first sight this appears to be inconsistent with the meaning of claim 1, which requires the presence of **three** different B-site elements. At the oral proceedings the appellant has explained, however, that this statement forms part of a summary of the contents of the application from which the present patent claims priority and which is set out in the description from column 14, line 32 to column 15, line 54. It does not relate to the invention as set out in the patent. These explanations are consistent with the statements in column 14, lines 32 - 34 and column 14, lines 48 - 50 of the patent in suit. From these statements it is clear that the disclosure beginning at column 14, line 32 and continuing on column 15, which refers to oxides compositions containing three transition metals or less (y' and y'' may be equal to zero, see column 14, lines 42 - 43), relates to compositions described in the priority

document. In these circumstances it is questionable whether the skilled person reading the passage in column 15, lines 40 - 46, would consider this teaching as concerning the compositions as defined in the granted claims, since the latter are clearly defined in the rest of the patent in suit, in the examples and in granted claim 1 as including three different transition metals. In any case to avoid any misconstructions the statement in column 15, lines 40 - 46 has been deleted from the description. Therefore, no inconsistency exists between claim 1 and the description as far as the number of distinct B-site elements is concerned.

3. The amendments in the description, in particular the deletion of the passage in column 15, lines 40 - 46, meet the requirements of Article 123 EPC. This was not in dispute.

Novelty of the claimed compositions

4. The respondent has contested the novelty of the claimed compositions of the formula $\text{Ln}_x\text{A}'_x\text{A}''_x\text{B}_y\text{B}'_y\text{B}''_y\text{O}_{3-z}$ set out in claim 1 in respect of each of the prior documents D1, D2, D4, D5, D6, D7 and D9, respectively.
 - 4.1 D1 discloses various oxides of the perovskite type including, in particular, LaMnO_3 , $\text{La}_{0.99}\text{MnO}_3$, $\text{La}_{0.94}\text{Sr}_{0.05}\text{MnO}_3$, $\text{La}_{0.89}\text{Sr}_{0.10}\text{MnO}_3$, $\text{La}_{0.79}\text{Sr}_{0.20}\text{MnO}_3$, $\text{La}_{0.69}\text{Sr}_{0.30}\text{MnO}_3$ and LaCrO_3 (D1, p. 568, left-hand column, paragraph 4; p. 569, table II; p. 570, paragraph (C)). All compositions specifically disclosed in D1 contain only one or two different B-site elements, as opposed to three different B-site elements according to claim 1 of the patent.

- 4.2 The respondent has referred to a statement on p. 570, paragraph (C), third paragraph of D1, according to which *"LaCrO₃ can be substituted with a cation on either the lanthanum or chromium sites. Examples of the substituents or dopants include strontium and calcium (lanthanum site) and magnesium, cobalt, zinc, copper, nickel, iron, aluminium, and titanium (chromium site)."* Based on that statement the respondent argued that mixed oxides comprising three different B-site elements form part of the disclosure of D1, since the skilled person would have realised that "multiple substitution of chromium", in particular the substitution of chromium by three other elements, was an option. The Board is not convinced by this argument, because D1 does not reveal any details regarding the number of elements which are used to substitute chromium. Whether the skilled person would contemplate compositions with three different B-site elements is a question of inventive step, not novelty. The subject-matter of claim 1 is therefore novel with regard to D1.
- 4.3 Document D2 discloses a material of the formula $\text{La}_{0.79}\text{Sr}_{0.2}\text{MnO}_3$ (p. 473, last paragraph of point 1). This does not affect the novelty of the compositions according to claim 1, since there is only one B-site element, viz. Mn.
- 4.4 Document D4 discloses various mixed metal oxides of the formula ABO_3 having a perovskite structure and containing various metal ions at the A- and B-sites, respectively (column 9, lines 23 - 47). It is stated in D4 that in general any combination of metals which satisfies the requirement of a perovskite may be used,

e.g. lanthanides, metals of groups Ia and IIa, transition metals Al, Ga, Ge, etc. (column 9, lines 63 - 66). Preferred B-site elements include Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu and Zn, whereby Co, Mn, Fe and Cr are particularly preferred (column 10, lines 11 - 15). Moreover D4 discloses that a wide variety of *"multiple cation substitutions"* at both the A and B sites is possible, and that *"perovskite materials containing more than two metals besides oxygen are preferred"* (column 10, lines 16 - 22). According to D4 the presence of chromium and/or titanium in the B-sites gives rise to increased stability of the perovskite structure. With chromium an increase of the electron conductivity is reported (column 10, lines 36 - 42). Mixed metal oxides represented by the formula $A_sA'_tB_uB'_vB''_wO_x$ (formula II), are preferred, wherein u is from 0.01 to about 1, v is from 0.01 to about 1, w is from 0 to about 1, and $0.9 < (s+t)/(u+v+w) < 1.1$ (column 10, lines 43 - 63). In the formula A represents a lanthanide, Y or a mixture thereof, A' represents an alkaline earth metal or mixtures thereof, B represents Fe, B' represents Cr or Ti or a mixture thereof, and/or B'' represents Mn, Co, V, Ni or Cu or a mixture thereof (claim 1 and column 10, lines 64 - 68). All specific examples given in D4 of mixed metal oxides containing three different B-site elements contain chromium (column 11, lines 12 - 14, 16 - 18, 20 - 22; column 12, lines 28 - 29, 30 - 32, 34 - 36; column 36, example M; column 39, table I, example A-13). Moreover all examples of D4 relate to stoichiometric materials. In other words the ratio between the A-site elements and the B-site elements is 1.0. Therefore a compound containing three different B-site elements selected from the transition metals, excluding Ti and Cr,

whereby the total amount of B-site elements is over-stoichiometric ($1.1 > y + y' + y'' > 1.0$) is not directly and unambiguously derivable from D4.

4.5 The respondent has contested the novelty of the compositions according to claim 1 on the basis of the following arguments: The general teaching of D4 does not require that chromium or titanium be present as a B-site element (column 12, lines 23 - 24). Titanium may even be excluded from the preferred embodiments (column 12, lines 56 - 58). If chromium and titanium are deselected from the preferred B-site elements Co, Mn, Fe and Cr (column 10, lines 14 - 15), the remaining elements correspond to the patent in suit. That the number of different B-site elements may be three is disclosed in column 10, lines 48 - 60. As far as the ratio of A-site elements to B-site elements is concerned, it is stated in column 10, lines 62 - 63 that the range is from 0.9 to 1.1, the preferred range being from 0.99 to 1.01. Thus, the feature of "B-site richness" forms part of the disclosure of D4. Although the sub-range of "B-site richness" characterised in granted claim 1 by the relation $1.1 > y + y' + y'' > 1.0$ is not disclosed as such in D4, it covers nearly 50% of the whole range of from 0.9 to 1.1 and cannot be regarded as being novel if the normal criteria for the selection of ranges are applied.

4.6 The Board is not convinced by this argumentation. The consistent case law of the Boards of Appeal is that for an invention to lack novelty its subject-matter must be directly and unambiguously derivable from the prior art. In the present case, starting from formula II in column 10 of D4, four steps have to be carried out in

order to arrive at the compositions according to claim 1 when using D4 as a basis: Firstly the special case of three different B-site elements has to be selected. Secondly the amounts of the three B-site elements must be such that the relation $1.1 > y + y' + y'' > 1.0$ is satisfied. Thirdly chromium and titanium have to be excluded from the list of B-site elements. And fourthly a metal of group 2 has to be selected as an A-site element. Nowhere in D4 is there a clear teaching, let alone a specific example of such a combination of four steps. To arrive at his conclusion the respondent has in fact combined different passages or features of D4 belonging to different preferred embodiments, although such a combination is not specifically suggested in D4. Decision T 0198/84 (OJ 1985, 209) does not concern such a situation and is therefore not directly applicable to the present case. Therefore D4 is not novelty-destroying.

4.7 D5 is concerned with mixed metal oxide compositions containing at least strontium, cobalt, iron and oxygen (page 1, lines 10 - 12). The compositions have a non-perovskite structure (see abstract). There is no disclosure of a composition containing three different B-site elements. For this reason alone D5 does not destroy the novelty of the compositions according to the patent in suit.

4.8 D6 relates to Perovskite-type oxides of the formula $\text{La}_{1-x}\text{Sr}_x\text{FeO}_{3-\delta}$ containing a single B-site element, viz. Fe. According to page 263 (right hand column, second paragraph) it is probable that the samples had a slight excess of iron oxide. However this assumption is not confirmed by any data. There is no disclosure of

compositions having three different B-site elements. The respondent's allegation at the oral proceedings according to which the samples contained also Co and Ni, since iron includes Co and Ni as impurities, was not supported by any evidence.. This allegation cannot be accepted in the absence of evidence showing that the preparation of the samples as described on page 258 of D6 (point 2, "Experimental") inevitably leads to final products containing measurable quantities of Co and Ni.

- 4.9 In D7 a mixed oxide composition of the formula $\text{La}_{0.89}\text{Sr}_{0.1}\text{MnO}_{3-\delta}$ is disclosed (column 4, lines 45 - 46). This material is slightly A-site deficient or, in other words, slightly over-stoichiometric with regard to the (single) B-site element Mn. Again there is no disclosure of a material with three different B-site elements, however.
- 4.10 D9 discloses compositions represented by the formula $\text{AA}'\text{BO}_3$, wherein A represents a mixture of rare earth elements, for example La, Pr, Nd. A' represents at least one element selected from alkali metals, alkaline earth metals and rare earth elements other than those contained in the mixture, and B represents a transition metal (column 3, lines 21 - 28; column 5, lines 1 - 6). According to column 3, lines 64 - 66, the B-site metal may be Co, Ni, Fe or mixtures thereof, whereby Co is preferred. Further it is stated in column 5, lines 7 - 8, that *"the transition metals may be iron, cobalt, nickel or mixtures thereof"*. The latter case is exemplified by the combinations of Co with Fe and Co with Ni, respectively (column 5, lines 13 - 17). In dependent claim 10 the expression "at least one transition metal" is said to be a mixture of two

transition metals, and according to claim 11 the mixture is a mixture of Co and Fe or Ni. None of the examples discloses a composition containing three different B-site elements. D9 further teaches that the surface atomic ratio of the transition metal and the rare earth elements of the A-site is in the range of from 1.0 : 1.0 to 1.1 : 1.0 (see claims 1 and 8). Thus, D9 refers to a different ratio from the one stated in granted claim 1 which takes into account the total amount of all A-site elements (i.e. A, A', A'', A' being mandatory). In column 4, lines 16 - 26 of D9 it is further disclosed that when at least one element is used as A'-site element, the surface atomic ratio of the transition metal and the rare earth elements is controlled to be nearly stoichiometric. It is therefore not directly and unambiguously derivable from the teaching of D9 that the oxidation catalyst disclosed therein contains an over-stoichiometric amount of the B-site elements in respect of the total amount of the A-site elements (A + A') in combination with three different B-site elements (Fe, Co and Ni), and with A' being an alkaline earth metal.

- 4.11 The Board concludes, therefore, that none of the documents D1, D2, D4, D5, D6, D7 and D9 discloses the compositions according to claim 1 of the patent in suit. The same applies to the remaining documents referred to by the respondent. Thus, the subject-matter of claim 1 is novel.

Inventive step of the compositions according to claim 1

5. At the oral proceedings the parties agreed that either D14 or, alternatively, D4 represents the closest prior art. The Board can accept this approach.
- 5.1 D14 discloses multicomponent metallic oxides represented by the formula $A_xA'_x A''_x B_y B'_y B''_y O_{3-z}$, wherein A, A' and A'' are selected from groups 1, 2, 3 and the f block lanthanides, B, B' and B'' are selected from the d block transition metals, and $0 < x \leq 1$, $0 \leq x' \leq 1$, $0 \leq x'' \leq 1$, $0 < y \leq 1$, $0 \leq y' \leq 1$, $0 \leq y'' \leq 1$, $x + x' + x'' = 1$, $y + y' + y'' = 1$, $z =$ a number which renders the compound charge neutral (column 3, lines 45 - 54). More specifically a composition comprising three different B-site elements is disclosed in D14, namely $La_{0.2}Ba_{0.8}Co_{0.6}Cu_{0.2}Fe_{0.2}O_{3-x}$ (column 6, line 34; claims 7, 14, 21 and 29). The compositions according to D14 are distinguished from the compositions according to claim 1 in that the ratio of B-site to A-site elements is 1.0 : 1.0. The respondent has argued that when applying the common understanding of decimal numbers to the coefficients y, y' and y'' of the B-site elements, the formula $La_{0.2}Ba_{0.8}Co_{0.6}Cu_{0.2}Fe_{0.2}O_{3-x}$ of D14 has to be construed to mean in reality $La_{0.2}Ba_{0.8}Co_{0.55 \text{ to } 0.66}Cu_{0.15 \text{ to } 0.24}Fe_{0.15 \text{ to } 0.24}O_{3-x}$. Accordingly the total amount of B-site elements is from 0.85 to 1.14 which includes non-stoichiometric relationships and, in particular, "B-site richness". The Board is not convinced by this argumentation. D14 is clearly concerned with stoichiometric compositions, and there is no evidence that non-stoichiometric embodiments are also included in the technical teaching of D14. Furthermore the respondent has not substantiated that his assumptions, which are based on

general considerations as to how decimal digits are commonly interpreted, reflect the reality of the present case. The compositions of D14 are suitable for producing membranes which are capable of separating oxygen from oxygen containing gaseous mixtures at elevated temperatures of, typically, greater than about 500°C (column 3, line 61 to column 4, line 8).

- 5.2 Starting from D14 the technical problem underlying the compositions as set out in claim 1 of the patent in suit can be seen in providing multicomponent metallic oxides compositions which are in particular suitable for use in solid-state oxygen-producing devices, and which have an improved resistance to degradation when subjected to high carbon dioxide and water partial pressures during operation. (column 4, lines 1 - 6 of the patent in suit).
- 5.3 According to claim 1 of the patent the appellant proposes to solve that problem by increasing the total amount of the B-site elements, so that the coefficients meet the relations $x+x'+x''=1.0$ and $1.1 > y+y'+y'' > 1.0$. It follows from example 10 of the patent in suit that the oxygen flux through such a "B-site rich" membrane prepared in accordance with example 2 substantially increases with increasing partial pressure of water, oxygen and carbon dioxide. Under the same operating conditions with a similar but "A-site rich" membrane, prepared in accordance with example 8, the oxygen flux did not change (column 20, line 50 to column 21, line 46; Figure 9). For a corresponding membrane having a "stoichiometric" composition the oxygen flux did not change (see experimental report dated 02.03.2001, page 15 and Figure A, resubmitted during the appeal

procedure with letter dated 11.09.2001, page 8). Thus, both stoichiometric and "A-site rich" materials show a deterioration of oxygen flux performance if exposed to elevated partial pressures of carbon dioxide and water at the operating temperature. In contrast, "B-site rich" materials in accordance with claim 1 of the patent exhibit a marked increase of the oxygen flux, which means that they have improved resistance to degradation (i.e. improved stability) under operating conditions involving heating the membrane to 850°C (see column 21, lines 2 - 6 of the patent in suit). In view of the comparative tests submitted by the appellant, where a temperature of 850°C has been used, the Board is satisfied that the compositions according to claim 1 actually solve the technical problem.

- 5.4 The respondent has not contested the experimental results submitted by the appellant, but he has argued that only one single combination of elements B, B' and B'', viz. Co, Fe and Cu, has been investigated in the patent in suit. In his view the findings are not necessarily representative for the whole scope of claim 1. This argument must fail for the following reasons. The patent in suit and the additional comparative tests show a significant improvement in resistance to degradation for a combination of the transition metals of the first row of the d-block transition metals, i.e. Co, Fe, Cu (3d transition metals). There is no information in the patent in suit from which it could be derived that the stability improvement is obtained only with compositions of claim 3 (limited to Co, Fe, Cu as B-site elements). The respondent has not substantiated why this effect should not be achieved with other combinations of transition

metals in the formula as defined in claim 1. The appealed decision likewise contains no technical reasons in this respect. Under these circumstances the respondent's allegation that the improvement is not obtained with other combinations, which was contested by the appellant, cannot be accepted by the board, taking into account that the burden of proof rests with the respondent in this respect. The sole fact that the same unsubstantiated allegation was made in the appealed decision is not sufficient to reverse the burden of proof. The number of d-block transition metals and of possible combinations with the A-site elements is indeed large; however, this is not sufficient to support the respondent's allegation in the present case where the stability improvement with respect to the closest prior art D14 has been shown to result from an over-stoichiometry of the B-site elements. In the absence of any evidence to the contrary, it is plausible that the improvement is also achieved with other combinations of transition metals.

5.5 Further the respondent has raised the criticism that the experimental data relate to a specific application of the claimed compositions, namely their use as oxygen transport membranes, whereas these compositions are intended for use in a broad field of various other applications including the separation of hydrogen (column 8, lines 49 - 50), the production of synthesis gas (column 8, lines 55 - 56) and other uses. Again the argument must fail. The appellant has demonstrated that there exists a technical advantage in a specific area of application, namely the use as membrane material in oxygen producing devices. This is sufficient for the acknowledgment of an inventive step of the claimed

product if the combination of features which leads to the said technical advantage involves an inventive step in view of the prior art. No necessity exists to demonstrate that the claimed compositions offer the same or other technical advantages when used in different applications.

5.6 The question arises whether the skilled person would have combined D14 with D1 in order to arrive at the claimed compositions. D1 deals with ceramic fuel cells based on oxygen-ion conductors. It points out that each component of a fuel cell must have the proper stability (chemical, phase, morphological, and dimensional) in oxidizing and/or reducing environments (page 564, left hand column, section II, first paragraph). Further it states that LaMnO_3 , which may be used as a cathode material, can have lanthanum deficiency or excess. In order to prevent undesired hydration when La is in excess and in view of the difficulties to prepare a stoichiometric material, it is recommended to use LaMnO_3 with a lanthanum deficiency (page 569, left hand column, second paragraph; table II). Similar to LaMnO_3 , it is said that LaCrO_3 with a lanthanum excess tends to undergo hydroxyde formation (page 570, right hand column, lines 28 - 30). There is no teaching in D1, however, that the stabilizing effect is achieved with oxides other than LaMnO_3 and LaCrO_3 , let alone with more complex compositions according to D14 under operating conditions. Thus, D1 cannot suggest how to solve the technical problem mentioned above in connection with compositions such as those of D14.

5.7 The question of the stability of sintered $\text{LaFeO}_{3-\delta}$ is briefly mentioned in D6, which states that, contrary to

the expectations, no breakdown of the structure had been observed, probably because of "a slight excess of iron oxide" (page 263, lines 8 - 15). As in the case of D1 there is no teaching in D6 that "B-site richness" leads to better stability of mixed metal oxides in general, when they are used in solid-state oxygen-producing devices and subjected to elevated carbon dioxide and water partial pressures during operation.

- 5.8 D8 is a scientific study dealing with the application of simultaneous thermogravimetry and mass spectrometry in the temperature range from 298 to 1773 K to two perovskite-type samples, namely lanthanum strontium manganite of the formula $(\text{La}_{0.7}\text{Sr}_{0.3})_{0.9}\text{MnO}_3$ and lanthanum calcium chromite of the formula $\text{La}_{0.7}\text{Ca}_{0.32}\text{CrO}_3$. It follows from the formulae, that the first sample has an excess of the B-site element (manganese), whereas the second sample has an excess of A-site elements (lanthanum and calcium). According to D8 the amounts of carbon dioxide and water that evolve from the lanthanum calcium chromite sample are larger than the amounts evolving from the lanthanum strontium manganite sample (page 3 of the English translation of D8, Fig. 1, Fig. 2, Table). On the basis of this teaching the respondent has argued that the skilled person would have recognised that B-site rich materials are more stable than A-site rich materials. The board is not convinced by this argument, however. D8 is concerned with the behaviour of the two specific sample materials under measurement conditions, not with the relationship between "B-site richness" and the stability of multicomponent metallic oxides comprising three different B-site elements, let alone with the stability of such compositions under operating conditions in

oxygen-producing devices. For this reason the experimental data provided by D8 cannot be regarded as being conclusive in respect of the technical problem underlying the patent in suit. Moreover, in view of the small number of experiments, as well as the fact that the two materials which have been investigated comprise different metals, it is questionable whether anything can be derived at all from D8 regarding the effects of "B-site richness". Therefore, in the board's view D8 gives no hint to the skilled person how the present technical problem can be solved.

5.9 In the light of the foregoing the Board concludes that the skilled person did not have an incentive to combine the teaching of D14 with D1 or, alternatively, with D6 or D8.

5.10 The respondent has argued that the skilled person would have arrived in an obvious manner at the compositions according to claim 1 on the basis of D4 alone. He submitted that the skilled person, starting from D4, would have taken formula II (column 10, line 46) as a basis. When exploring the preferred options within the framework of formula II, he would have found a first indication in D4, namely to exclude Ti (column 12, lines 56 - 57). A second indication would have been to replace Cr in formula II by Fe, Co and Mn (column 12, lines 23 - 24). Having this in mind, the skilled person would then have selected a suitable ratio of the B- and A-site elements. Here the selection of the sub-range of $1.1 > y + y' + y'' > 1.0$ would have been obvious, since this was not less than nearly 50 % of the entire range of $1.1 > y + y' + y'' > 0.9$ disclosed in D4. When following the various indications contained in D4, the skilled person

would have arrived in an obvious manner at the claimed compositions.

5.11 The Board does not share the respondent's views regarding D4. In fact no explanation has been given by the respondent why the skilled person would have picked out the specific elements referred to above, in order to combine them in the expectation of being able to solve the technical problem. The problem of increasing the stability of the perovskite structure at operating conditions is dealt with in D4. In order to solve this problem D4 teaches to use Cr and/or Ti in the B-sites of the perovskite ionic lattice (see column 9, lines 55 - 60 and column 10, lines 36 - 42). The solution proposed in D4 is thus completely different and cannot suggest the claimed over-stoichiometry of the B-site elements. The skilled person would not have combined the passages picked out by the respondent since he could not expect such a combination of features to lead to a stability improvement. In the Board's view such a combination could only be made with hindsight. Therefore the argumentation of the respondent is not convincing.

5.12 D9 concerns an oxydation catalyst of the perovskite-type compound oxide (see point 4.10 above for the composition). It deals with the problem of providing catalysts which have a high oxidation activity and are highly resistant to heat (see column 2, lines 1-6). According to D9, these catalysts have a good resistance to heat when determined at a temperature of 900°C, and they can be used under high temperature conditions, contrary to the platinum group metals (column 7, lines 46 - 51). The problem of instability of metallic oxides

of the perovskite-type when used in oxygen-producing devices under high carbon dioxide and water partial pressures during operation at high temperatures is not dealt with in D9. The respondent's argument that it was obvious to provide compositions containing all three preferred transition metals, namely Co, Fe **and** Ni is not convincing, since D9 does not contain any indication that mixed metal oxides containing all three of the transition metals and an over-stoichiometric amount of the B-site elements would improve the stability of the material under operating conditions in oxygen-producing devices. Thus, the skilled person could not expect that D9 would make any contribution to the solution of the present technical problem.

- 5.13 The respondent did not rely on the other documents in connection with the issue of inventive step. The Board is also convinced that these documents are of less relevance than the ones discussed above, and that they do not provide an incentive for the claimed solution of the above-mentioned technical problem. Therefore the compositions according to claim 1 involve an inventive step within the meaning of Article 56 EPC.
- 5.14 Claim 2 is dependent on claim 1. The inventive step of its subject-matter follows from that dependency.
- 5.15 Claims 3 to 9 as granted correspond to claims 1 to 7 of the first auxiliary request which were considered to meet the requirements of the EPC in the interlocutory decision of the opposition division. The respondent did not object to these claims during the appeal proceedings, and the board sees no reason to deviate from the decision of the opposition division as regards

the patentability of these claims under the requirements of the EPC.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the first instance with the order to maintain the patent with the following documents:
 - Claim 1 to 9 as granted,
 - description pages 2, 3, 5 to 8, 10 and figures 1 to 9 as granted,
 - description pages 4, 9, 11 and 12 as filed during the oral proceedings.

The Registrar:

The Chairman:

A. Wallrodt

M. Eberhard