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**Datasheet for the decision
of 5 August 2010**

Case Number: T 0121/07 - 3.3.07

Application Number: 01976135.2

Publication Number: 1317319

IPC: B01D 71/02

Language of the proceedings: EN

Title of invention:

Mixed conducting membranes for syngas production

Patent Proprietors:

AIR PRODUCTS AND CHEMICALS, INC.

Opponents:

L'AIR LIQUIDE S.A. A DIRECTOIRE ET CONSEIL DE SURVEILLANCE
POUR L'ETUDE ET L'EXPLOITATION DES PROCEDES GEORGES CLAUDE

Headword:

-

Relevant legal provisions:

-

Relevant legal provisions (EPC 1973):

EPC Art. 56

Keyword:

"Inventive step (yes) - non obvious combination of known
features"

Decisions cited:

-

Catchword:

-



Case Number: T 0121/07 - 3.3.07

DECISION
of the Technical Board of Appeal 3.3.07
of 5 August 2010

Appellants:
(Opponents) L'AIR LIQUIDE S.A. A DIRECTOIRE ET CONSEIL DE
SURVEILLANCE POUR L'ETUDE ET L'EXPLOITATION
DES PROCEDES GEORGES CLAUDE
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Respondents:
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted on
27 November 2006 rejecting the opposition
against the European Patent No. 1317319.

Composition of the Board:

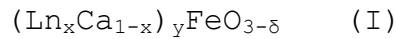
Chairman: S. Perryman
Members: G. Santavicca
B. ter Laan

Summary of Facts and Submissions

I. The appeal by the opponents lies from a decision rejecting their opposition against European patent No. 1 317 319.

II. The patent, granted on European application No. 01976135.2, comprised 22 claims, the independent claims reading as follows:

"1. A metallic oxide represented by Formula (I)



wherein

Ln is La or a mixture of lanthanides comprising La, and wherein

$$1.0 > x > 0.5$$

$$1.1 \geq y > 1.0 \text{ and}$$

δ is a number which renders the metallic oxide charge neutral."

"4. A solid-state membrane which comprises a dense layer formed from a metallic oxide as defined in any one of Claims 1 to 3."

"14. A process for producing a synthesis gas product comprising hydrogen and carbon monoxide which comprises the steps of:

i) providing a reaction zone having an oxidant side and a reactant side which are separated by a solid-state membrane as defined in any one of Claims 4 to 13,

ii) contacting a heated oxygen-containing feed gas with the oxidant side of the said membrane at an oxidant feed temperature and an oxidant gas feed pressure;

iii) contacting a heated methane-containing reactant gas with the reactant side of the said membrane at a reactant gas feed temperature and a reactant gas feed pressure;

whereby oxygen from the oxidant side of the reaction zone permeates through the solid-state membrane to the reactant side of the reaction zone and reacts with the methane-containing reactant gas to form the synthesis gas product;

iv) withdrawing the synthesis gas product from the reactant side of the reaction zone; and

v) withdrawing an oxygen depleted gas stream from the oxidant side of the reaction zone."

III. The patent had been opposed on the ground that its claimed subject-matter lacked an inventive step (Article 100(a) EPC), having regard to the following documents:

D1: US-A-5 306 411;

D2: US-A-4 863 971;

D3: US-A-5 693 212;

D4: D. Kuščer et al., "Some characteristics of Al₂O₃- and CaO-modified LaFeO₃-based cathode materials for solid oxide fuel cells", Journal of Power Sources, 61(1996), pages 161-165;

D5: US-A-6 060 420.

IV. According to the decision under appeal:

- (a) The claimed subject-matter concerned novel nonstoichiometric (A-site rich) ferrites having the defined structure.
- (b) D1, which disclosed ferrite compositions encompassing both stoichiometric and nonstoichiometric compositions, without indicating any effect or desirability for the A-site rich nonstoichiometric ferrites, described the closest prior art.
- (c) Having regard to the compositions of D1, the claimed subject-matter showed an improved creep resistance.
- (d) As to obviousness, D1 required the presence of at least one further "B" metal in addition to Fe. D4 disclosed ferrites comprising the elements as claimed but was silent on nonstoichiometry and did not contain any incentive to remove the further B-site metal B' required by D1. Hence, neither D1 nor its improbable combination with D4 rendered obvious the claimed subject-matter, which thus involved an inventive step.

V. In their statement setting out the grounds of the appeal, the appellants enclosed copies of the following further documents:

- D6: J.W. Stevenson et al., *Effect of A-site cation nonstoichiometry on the properties of doped lanthanum gallate*, Solid State Ionics 113-115 (1998), pages 571-583.
- D7: W.R. Canon et al., *Review Creep of ceramics, Part 2, An examination of flow mechanisms*, Journal of Materials Science 23 (1988), pages 1-20.

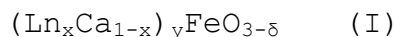
D8: Nina Orlovskaja et al., *Mechanical properties of LaCoO₃ based ceramics*, Journal of the European Ceramic Society, 20 (2000), pages 51-56.

In response to a communication of the Board in preparation for the oral proceedings, the opponents informed the Board that they would not attend the oral proceedings (letter dated 3 June 2010).

VI. The patent proprietors (respondents), by letter dated 16 August 2007, filed observations on the grounds of appeal, and, with a letter dated 30 June 2010 in response to a communication of the Board in preparation for the oral proceedings, submitted four sets of amended claims as First to Fourth Auxiliary Requests.

VII. Oral proceedings were held on 5 August 2010, in the absence of the appellants, pursuant to Rule 115(2) EPC. During the oral proceedings, the respondents submitted a set of amended Claims 1 to 21 as the new Main Request, Claim 1 reading as follows (compared to Claim 1 as granted, the added features are indicated in bold):

"1. A **solid-state membrane which comprises a dense layer formed from a** metallic oxide represented by Formula (I)



wherein

Ln is La or a mixture of lanthanides comprising La, and wherein

$$1.0 > x > 0.5$$

$$1.1 \geq y > 1.0 \text{ and}$$

δ is a number which renders the metallic oxide charge neutral."

VIII. The appellants, in their written submissions, had essentially argued as follows:

- (a) D1 pertained to the same technical field as the patent in suit and disclosed an improved dense membrane suitable for syngas production, so that it described the closest prior art.
- (b) D1 disclosed a perovskite structure of formula $\text{La}_s\text{Ca}_t\text{Fe}_u\text{B}'_v\text{O}_x$, whereby the coefficients s , t , u and v were such to encompass the possibility of an excess of the elements La and Ca. Also the ratio s/t overlapped with the range of values for x as defined in Claim 1 of the opposed patent.
- (c) Therefore, the only distinction between the structure of Claim 1 and that of D1 merely consisted in the absence of element B'.
- (d) Since no effect whatsoever related to the lack of element B' had been demonstrated, the solved problem was to provide alternative oxides to those of D1 being suitable for the conversion of methane, natural gas or other hydrocarbons to syngas.
- (e) For assessing the inventiveness of the claimed structure, the skilled person was a chemist specialized in ceramic materials, *inter alia* having the knowledge described in any of D5 to D8.
- (f) D2 concerned perovskite catalysts for syngas production and disclosed that the perovskite oxide LaFeO_3 was a suitable catalyst for the conversion of methane to syngas. In other words, it taught that a perovskite oxide for syngas production could contain only one transition element such as Fe.
- (g) As the absence of the second transition element resulted in a simpler structure of the oxide, the

skilled person would without hesitation implement it as an alternative to the oxides of D1. Hence, the claimed subject-matter was obvious.

- (h) In case the absence of the second element B' of D1 were not considered obvious or if the problem to be solved were seen as an improvement of the mechanical properties of the oxide material, the skilled person would replace the element Sr in any of the exemplified $(\text{Ln}_x\text{Sr}_{1-x})\text{FeO}_{3-\delta}$ structures of D1 with Ca, as D4 taught that calcium doping was beneficial to the mechanical properties of the perovskite structures containing lanthanum and iron, and would arrive at the claimed subject-matter.
- (i) Therefore, the patent in suit should be revoked.

IX. The respondents argued essentially as follows:

- (a) D1 was the closest prior art document because it disclosed perovskite structures for syngas production that were similar to those as claimed.
- (b) The problem solved over the structures of D1, in particular the lanthanum strontium ferrite, was to provide a membrane for syngas production having sufficiently low creep rate under the applied load and temperature.
- (c) As regards the skilled person and his alleged common general knowledge, D6 and D8 were specialized documents addressing specific perovskite structures that were remote from those defined in Claim 1 of the Main Request, and D7 concerned ceramics in general, not those as claimed. Therefore, D6 to D8 were not relevant and should not be admitted into the proceedings.

- (d) Starting from the perovskite oxides of D1, many choices (in particular of the elements La, Ca, Fe and their coefficients) and modifications (suppression of the further elements B' and B" in the preferred oxides of D1) were necessary in order to arrive at the oxides defined in present Claim 1.
- (e) None of those choices or modifications were suggested in any of D2 and D4. D2 disclosed catalysts, not dense membranes. D4 disclosed perovskite oxides containing La, Ca and Fe. At the priority date of D4, the A/B ratio of those oxides (nonstoichiometry) could be determined with an accuracy of 1/10000. However, D4 concerned porous cathodes for solid oxide fuel cells rather than dense membranes. Hence, D2 and D4 did not hint at a membrane as defined in Claim 1 of the Main Request.
- (f) The claimed subject-matter was thus not obvious.
- (g) Therefore, the amended claims of the Main Request fulfilled the requirements of the EPC.

X. The appellants (opponents) had requested in writing that the decision under appeal be set aside and that the patent be revoked.

XI. The respondents (patent proprietors) requested to set aside the decision under appeal and to maintain the patent on the basis of the claims of the Main Request submitted at the oral proceedings on 5 August 2010.

Reasons for the Decision

1. The appeal is admissible.

Main Request

2. *Amendments*

Claim 1 consists of the combination of the features of Claims 1 and 4 as granted (point II, *supra*).

In Claims 2 and 3 the term "solid-state membrane" replaces the term "metallic oxide" of granted Claim 1.

Claim 3 now refers to Claim 2.

The amendments to claims 4-21 (adaptation of their numbering and references) are a consequence of the deletion of Claim 4 as granted.

The amendments comply with the requirements of Article 123 EPC, paragraphs (2) and (3), and do not introduce any confusion, ambiguity or obscurity (Article 84 EPC). They aim at overcoming a ground of opposition, namely the lack of novelty or of inventive step of the oxides of Claim 1 as granted over the oxides disclosed by D4.

Since the Main Request consists of a more restricted version of the claims as granted as well as of the claims of the First to Fourth Auxiliary Requests submitted with letter dated 30 June 2010, received and not commented upon by the appellants, no surprise arises for the appellants who had decided not to attend the oral proceedings.

Therefore, the amendments are allowable.

3. *Novelty*

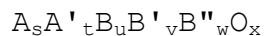
Novelty is not in dispute; the differences between the claimed subject-matter and that of D1 will become apparent from the following.

Inventive step

4. The patent in suit concerns mixed conducting membranes for syngas production, of the type disclosed in D1, acknowledged in the patent in suit as pertaining to the same technical field and chosen by the Opposition Division and all the parties as the closest prior art document. The Board sees no reason to deviate from that point of view.

Closest prior art

5. D1 discloses a solid, gas-impervious, electron-conductive, oxygen ion-conductive, single-phase membrane for use in an electrochemical reactor, said membrane being formed from a perovskite represented by the formula:



wherein A represents a lanthanide, Y, or mixture thereof; A' represents an alkaline earth metal or mixture thereof; B represents Fe; B' represents Cr, Ti, or mixture thereof; and B'' represents Mn, Co, V, Ni, Cu, or mixture thereof and s, t, u, v, w, and x each represent a number such that:
s/t equals from about 0.01 to about 100;
u equals from about 0.01 to about 1;
v equals from about 0.01 to 1;
w equals from zero to about 1;

x equals a number that satisfies the valences of the A, A', B, B' and B'' in the formula; and $0.9 < (s+t)/(u+v+w) < 1.1$ (Claim 15).

- 5.1 The mixed conducting membrane formed from those oxides can be used for oxidizing methane, natural gas or other hydrocarbons to produce synthesis gas (column 4, lines 9-12).
- 5.2 In view of the requirement that "v" be at least 0.01, the oxides of D1 require the presence of at least a second transition element, B', in addition to Fe, and thus necessarily include more elements than those defined in Claim 1 of the Main Request, the subject-matter of which is as a result novel (point 3, *supra*).
- 5.3 B' can be at least chromium (Claim 17), preferably a mixture of chromium and titanium (Claim 18). A' can be at least strontium (Claim 19), in particular in an oxide where B' is at least chromium (Claim 20). The membrane can be formed from a perovskite oxide containing La, Sr and Cr (Claim 21). D1 also mentions lanthanum-calcium-iron chromite as a suitable perovskite oxide (column 11, lines 15-16).
- 5.4 As regards specific perovskites for use in syngas production, D1 (columns 35 to 37) *inter alia* exemplifies the following structures, which have been referred to by the opponents (points VIII(b)(h), *supra*):
La_{0.2}Sr_{0.8}CoO_x (Examples F and H);
La_{0.2}Ca_{0.8}CoO_x (Example G);
La_{0.2}Sr_{0.8}FeO_x (Example J);
La_{0.2}Sr_{0.8}Fe_{0.8}Cr_{0.2}O_x (Example K);
La_{0.2}Sr_{0.8}Fe_{0.8}Cr_{0.1}Co_{0.1}O_x (Example M);

$\text{La}_{0.2}\text{Sr}_{0.8}\text{Fe}_{0.8}\text{Mn}_{0.2}\text{O}_x$ (Example N).

However, the oxides of Examples F, G, H neither contain Fe nor a second transition element B' such as Cr and/or Ti. That of Example J contains Fe but not the required second transition element B' such as Cr and/or Ti. That of Example N contains Mn as a further transition metal B'' instead of a second transition elements B' such as Cr and/or Ti. Hence, the oxides of Examples F, G, H, J and N do not satisfy the definition of Claim 15 of D1 (*supra*), so that they constitute comparative oxide compositions for D1.

Instead, the oxides of Examples K and M satisfy all the requirements of the claims of D1. Membranes made from the above oxides are illustrated in Examples A-11 and A-13 of D1, which can be considered as the closest embodiments for the Main Request.

- 5.5 D1 addresses the problem of facilitating the electrocatalytic conversion of light hydrocarbons to synthesis gas (column 1, lines 38-46), and in particular the problem of increasing the stability of the perovskite structure under the conditions of the catalytic processes carried out, as well as the electron conductivity (column 10, lines 36-42).

Problem solved

6. According to the patent in suit (page 3, [paragraph 0010], lines 22-25), the solid-state membranes for making syngas must have sufficiently high oxygen flux, sufficient chemical stability in the syngas and air environments, sufficiently low creep rate under the

- applied mechanical load, sufficient resistance to demixing of the metal cations and sufficiently low chemical expansion under the membrane operating conditions.
- 6.1 Still according to the patent in suit (page 3, [paragraph 0010], lines 36-38), a solid state membrane formed from A-site rich nonstoichiometric multicomponent metallic oxides as defined in Claim 1 of the Main Request, when used for syngas production, provides a favourable balance of oxygen permeance, resistance to degradation, sintering properties and coefficients of thermal expansion which are compatible with the other materials used.
- 6.2 The examples of the patent in suit concern 8 multicomponent metallic oxides, the compositions of which are summarized in Table 1. Among them, those of Examples 1 and 6 to 8 are according to Claim 1 of the Main Request, the others being comparative.
- 6.3 However, the perovskite compositions summarized in Table 1 of the patent in suit, apart from that of Comparative Example 5*, all contain a high proportion of La (at least 0.8, compared to >0.5 as defined in present Claim 1) and a specific nonstoichiometry (ratio A/B) for the A elements La and Ca, namely 1.01, *versus* $1.1 \geq y > 1.0$ as defined in present Claim 1. Hence, Examples 1 and 6 to 8 of the patent in suit fulfil the definition of present Claim 1 but in fact illustrate only a few specific oxides covering a very narrow area within the broad spectrum of possibilities falling under Formula (I) of present Claim 1.

- 6.4 Moreover, none of the comparative compositions summarized in Table 1 of the patent in suit, including the stoichiometric structure of Comparative Example 5* $[(La_{0.15}Sr_{0.85})_{1.0}Fe_{0.7}Al_{0.3}O_{3-\delta}]$, corresponds to the oxides of D1, as neither Cr nor Ti are present in any of them, although Cr and/or Ti are disclosed by D1 as being necessary for improving the membrane stability.
- 6.5 The same applies to the experimental report submitted with letter dated 18 October 2005, in response to the notice of opposition, which shows the effect of non-stoichiometry (ratio A/B) but only for three specific compositions, none of which represents a composition according to D1. In fact, according to Figure A of the report, it is apparent that if the non-stoichiometry A/B is below about 1.005 no improvement is achieved, even for the specific composition tested. These compositions of low non-stoichiometry are however encompassed by present Claim 1.
- 6.6 It follows from the above that it has not been shown, nor is it plausible in view of the complexity of the defined oxides, that the desired improvements as mentioned in the patent in suit are actually obtained within the whole breadth of Claim 1.
- 6.7 Consequently, the problem solved within the whole breadth of the claims by the claimed features can only be formulated as to provide further membranes for syngas production over those of D1.

Obviousness

7. It remains to be decided whether it was obvious for the skilled person to provide further membranes for syngas production as claimed, starting from D1 as the closest prior art.

7.1 In order to increase the stability of the oxides under the conditions of the process in which they are used, such as syngas production, D1 recommends the inclusion of Cr and/or Ti in the composition of the perovskite oxides for forming the membrane structures (column 10, lines 36-40). According to the definition of Claim 15 of D1, Cr and/or Ti are present in the oxides in addition to Fe, as in the lanthanum-calcium-iron-chromite (mentioned in column 11, lines 15-16, and column 12, lines 29-30). According to D1, the presence of Cr also improves electron conductivity (column 10, lines 40-42). Hence, D1 suggests to supplement Fe with Cr or Ti at the B site of the perovskite structure, possibly also with other transition metals, as encompassed by the symbol B" in the formula given in Claims 1 and 15 of D1.

Instead, the membrane defined in present Claim 1 is formed from a mixed conducting oxide having only Fe as transitional metal "B", i.e. not containing any further transitional metals such as Cr and/or Ti, which are required by D1.

Since the mandatory presence of Cr and/or Ti in addition to Fe in the "B" site of the perovskite structure of D1 is said to be advantageous for the stability and the electron conductivity, the skilled

person would not modify the compositions of D1 in a way that involves the elimination of Cr and/or Ti.

7.2 As regards the arguments of the opponents based on the membranes formed from the oxides of Examples F, G, H, J, K, M and N of D1 (point 5.4, *supra*), if taken as the closest prior art embodiments, the following is noted: only the membranes formed from oxides according to Examples K and M fall under the claims of D1 and can hence serve as a proper starting point; and their proportion of La (i.e. 0.2) does not fulfil the requirement (>0.5) defined in present Claim 1. Hence, at least two modifications would be required to arrive from a membrane made from the oxide of Example K or M, at the membrane of present Claim 1. That conclusion would not change if one nevertheless considered any membrane formed from the other oxides, such as those of comparative Examples G and J, so that those modifications are not suggested by D1.

7.3 Therefore, D1 alone does not suggest the membrane defined in Claim 1 of the new Main Request.

7.4 The other documents cited are less relevant than D1.

7.4.1 D4 discloses a number of electrical and structural characteristics of CaO-modified LaFeO₃-based cathode materials for solid oxide fuel cells (SOFC) (title and Abstract), in particular a La_{1-x}Ca_xFeO₃ system, wherein "x" can be 0, 0.1, 0.3, 0.5, 0.75 and 1 (page 162: point 2, Experimental; Figure 2).

The La_{1-x}Ca_xFeO₃ oxide with x=0.1 and x=0.3 fulfil the requirement for the coefficient of La as defined in

present Claim 1. However, D4 does not mention any non-stoichiometry, let alone A-site rich oxides. Moreover, the purpose of D4 is to investigate alternative cathode materials with mixed conductivities (page 161, right column, first sentence of the last paragraph). As is usual for cathodes, the sintered materials of D4 are porous (abstract, third sentence), not dense membranes.

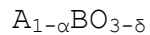
Therefore, D4 pertains to a different field (porous materials for the cathodes of SOFC) and cannot supplement D1 towards a membrane comprising a dense layer for syngas production, despite the closeness of the lanthanum calcium ferrites of D4 to the materials as claimed.

- 7.4.2 D2 concerns a process for synthesis gas conversion comprising reacting synthesis gas comprising hydrogen and carbon monoxide at a pressure in the range of 100 to 20,000 psig and at a temperature in the range of about 200°C to about 400°C in the presence of a perovskite catalyst such that the selectivity for lower oxygenated organic compounds containing one to six carbon atoms is at least about eight mol percent and wherein the perovskite catalyst is *inter alia* selected from lanthanum ferrites such as LaFeMO_3 , $\text{LaMn}_{0.5}\text{Fe}_{0.5}\text{O}_3$, $\text{LaFe}_{0.5}\text{Ru}_{0.5}\text{O}_3$ and $\text{LaFe}_{0.5}\text{Ti}_{0.5}\text{O}_3$ (Claim 1).

All lanthanum ferrites of D2 lack calcium as the "A" element in addition to lanthanum. The only perovskite composition containing two "A" elements disclosed in D2 is $\text{La}_{0.8}\text{Sr}_{0.2}\text{CoO}_3$ (column 8, line 8). In fact, D2 does not mention calcium doping, nor A-site non-stoichiometry.

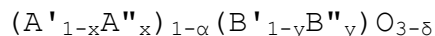
7.4.3 D3 is a continuation application of D1 and does not disclose more than D1.

7.4.4 D5 discloses a catalyst for purifying exhaust gas discharged from an internal combustion engine comprising a composite oxide of A-site defect perovskite structure represented by the following general formula:



wherein A can be an alkaline earth metal or a rare earth element, B can be Fe and $0.12 \leq \alpha \leq 0.15$, $0 < \delta \leq 1$ (Claim 1).

In particular, the perovskite structure can be represented by the following general formula:



wherein A' can be La, A'' can be Ca, B' and B'' are different and one of them can be Fe, and $0.12 \leq \alpha \leq 0.15$, $0 \leq \delta \leq 1$, $0 < x < 1$ and $0 < y < 1$ (Claim 8).

It follows from the foregoing that D5 concerns B-site rich perovskite oxides, suitable for use in catalysts and electrode materials having improved durability and thermal resistance (column 3, lines 35-43). None of the exemplified oxides in D5 concerns a lanthanum-calcium-ferrite. In fact, in D5 lanthanum and calcium are not used together in the same material.

7.4.5 In view of the above, none of the documents D2 to D5 suggests to modify the membranes of D1 in such a way as to arrive at the membranes now being claimed.

7.4.6 As regards the late filed documents D6 to D8, the following can be noted:

- (a) D6 addresses the effect of the A-site cation non-stoichiometry of a perovskite structure such as a doped lanthanum gallate. It mentions that a slight excess A-site cation content for calcium doped lanthanum chromite can substantially enhance densification during the sintering process (page 372, left column, first paragraph, last sentence). However, according to D6, lanthanum manganite and lanthanum chromite having a lanthanum deficiency ($A/B < 1$) are preferred (page 372, left column, second paragraph, first sentence). Hence, D6 concerns the effect of non-stoichiometry on densification of specific oxides.
- (b) D7 concerns a review of the creep in ceramic materials in general. No perovskite ferrite is mentioned.
- (c) D8 discloses a number of mechanical properties of lanthanum cobaltite based ceramics, such as $\text{La}_{0.8}\text{Ca}_{0.2}\text{CoO}_3$, which are suitable for forming membranes for separation processes (page 51, Introduction). The sample illustrated in Table 1 ($\text{La}_{0.922}\text{Ca}_{0.221}\text{CoO}_3$) is A-site rich and presents secondary phases of CaO-CoO. The calcium doped lanthanum cobaltite is said to have good mechanical properties (point 4 conclusions), such as the highest bending strength, which however quickly decreases with increased temperature (Figure 1), as well as fracture toughness and Young's modulus.
- (d) Hence, none of D6 to D8 concerns A-site rich calcium doped lanthanum ferrites as now claimed.

(e) Therefore, those documents are not more relevant than the documents on file and are therefore not admitted into the proceedings.

7.5 It follows from the foregoing considerations that the subject-matter of present Claim 1 is based on a non-obvious combination of known features and hence involves an inventive step (Article 56 EPC). The further claims include or refer to the features of Claim 1 and hence are inventive too.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The matter is remitted to the first instance with the order to maintain the patent on the basis of the claims of the main request submitted at the oral proceedings on 5 August 2010 and a description to be adapted thereto.

The Registrar:

The Chairman:

S. Fabiani

S. Perryman