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**Datasheet for the decision
of 19 November 2013**

Case Number: T 1998/10 - 3.3.05

Application Number: 05813697.9

Publication Number: 1827663

IPC: B01D71/02, C01B13/02, B01D53/22

Language of the proceedings: EN

Title of invention:
CATALYTIC MEMBRANE REACTOR

Applicant:
L'AIR LIQUIDE, Société Anonyme pour l'Etude
et l'Exploitation des Procédés Georges Claude

Headword:
Layer assembly/L'AIR LIQUIDE, Société Anonyme pour l'Etude
et l'Exploitation des Procédés Georges Claude

Relevant legal provisions:
EPC Art. 84, 111(1), 113(1)
EPC R. 103, 111(2)
Guidelines for examination E-X, 5

Keyword:
Reimbursement of appeal fee - (no) - appealed decision
reasoned (yes)
Remittal to the department of first instance - (yes)

Decisions cited:
T 0571/03

Catchword:



**Beschwerdekammern
Boards of Appeal
Chambres de recours**

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Case Number: T 1998/10 - 3.3.05

D E C I S I O N
of Technical Board of Appeal 3.3.05
of 19 November 2013

Appellant: L'AIR LIQUIDE, Société Anonyme pour l'Etude
(Applicant) et l'Exploitation des Procédés Georges Claude
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Representative: Conan, Philippe Claude
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75321 Paris Cedex 07 (FR)

Decision under appeal: **Decision of the Examining Division of the
European Patent Office posted on 26 May 2010
refusing European patent application No.
05813697.9 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman: G. Raths
Members: H. Engl
D. Prietzel-Funk

Summary of Facts and Submissions

I. This appeal is from the decision of the examining division, posted on 26 May 2010, refusing European patent application EP 05 813 697.9.

II. In the reasons for this "decision according to the state of the file" concerning a main request and two auxiliary requests, reference was made to the communication dated 28 April 2010 in which

"the applicant was informed that the application does not meet the requirements of the European Patent Convention. The applicant was also informed of the reasons therein. The applicant filed no comments or amendments in reply to the latest communication but requested a decision according to the state of the file by a letter received in due time on 05.05.2010."

The application was thus refused.

III. The notice of appeal of the applicant (henceforth: the appellant) was filed by letter dated 7 July 2010. The statement of grounds of appeal was received under cover of a letter dated 1 September 2010 where the appellant not only maintained and defended all rejected requests, but also submitted an additional set of claims constituting a new first auxiliary request.

IV. The board issued a preliminary communication pursuant to Rule 100(2) EPC in which it informed the appellant of its provisional opinion. The board indicated the possibility of remitting the case to the department of first instance for further prosecution on the basis of the claims of the (new) first auxiliary request, which were considered to overcome all reasons for the

refusal.

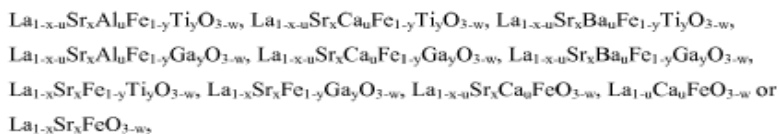
V. In its reply dated 8 November 2013, the appellant made the auxiliary request with the text according to the letter dated 1 September 2010 its new main request. The auxiliary request for oral proceedings was withdrawn.

VI. Claim 1 of the new main request reads as follows:

1. Organized assembly based on superposed layers of materials of similar chemical nature, characterized in that it comprises:

(a) – a dense layer (C_{D1}), with a thickness E_{D1}, the porosity of which does not exceed 5% by volume, the said dense layer (C_{D1}) consisting of a material (A_{D1}) comprising, for 100% of its volume:

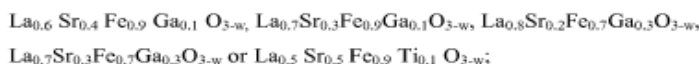
(i) – at least 75% by volume and at most 100% by volume of a compound (C₁) chosen from doped ceramic oxides which, at the use temperature of **between 600°C and 1 100°C**, are in the form of a crystal lattice with oxide ion vacancies of perovskite phase, of formulae:



in which

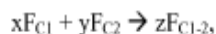
- 0 < x ≤ 0.5;
- 0 ≤ u ≤ 0.5;
- (x + u) ≤ 0.5;
- 0 ≤ y ≤ 0.9; and
- w is such that the structure in question is electrically neutral;

and more particularly those of formulae:



(ii) – optionally up to 25% by volume of a compound (C₂), which differs from compound (C₁), chosen from magnesium oxide (MgO), calcium oxide (CaO), aluminum oxide (Al₂O₃), zirconium oxide (ZrO₂), titanium oxide (TiO₂), strontium-aluminum mixed oxides SrAl₂O₄ or Sr₃Al₂O₆, barium-titanium mixed oxide (BaTiO₃), calcium-titanium mixed oxide (CaTiO₃), La_{0.5}Sr_{0.5}Fe_{0.9}Ti_{0.1}O_{3-w} or La_{0.6}Sr_{0.4}Fe_{0.9}Ga_{0.1}O_{3-w}; and

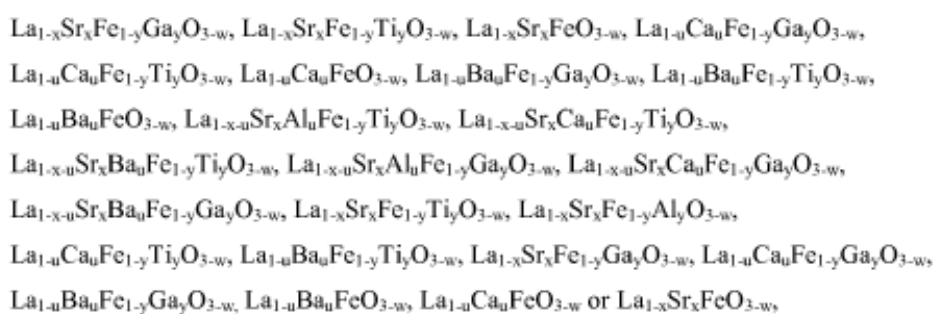
(iii) – optionally up to 2.5% by volume of a compound (C₁₋₂) produced from at least one chemical reaction represented by the equation:



in which equation F_{C1} , F_{C2} and F_{C1-2} represent the respective raw formulae of compounds (C₁), (C₂) and (C₁₋₂) and x, y and z represent rational numbers greater than or equal to 0;

(b) – a porous layer (C_{P1}), with a thickness of E_{P1} , the volume porosity of which is between 20% and 80%, adjacent to the said dense layer (C_{D1}), the said porous layer (C_{P1}) consisting of a material (A_{P1}) comprising, per 100% of its volume:

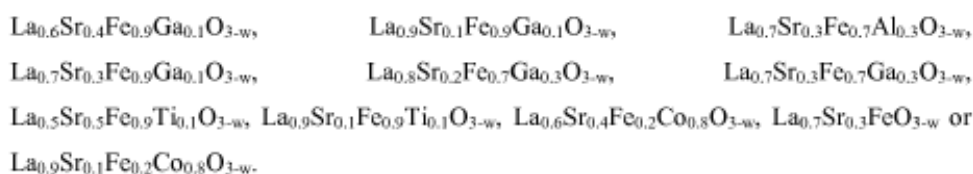
(i) – at least 75% by volume and at most 100% by volume of a compound (C₃) chosen from doped ceramic oxides which, at the use temperature of between 600°C and 1 100°C, are in the form of a crystal lattice having oxide ion vacancies of perovskite phase, of formulae:



in which

- $0 < x \leq 0.5$;
- $0 \leq u \leq 0.5$;
- $(x + u) \leq 0.5$;
- $0 \leq y \leq 0.9$; and
- w is such that the structure in question is electrically neutral;

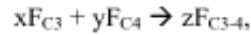
and more particularly those of formulae:



(ii) – optionally up to 25% by volume of a compound (C₄), which differs from compound (C₃), chosen from magnesium oxide (MgO), calcium oxide (CaO), aluminum oxide (Al₂O₃), zirconium oxide (ZrO₂), titanium oxide (TiO₂), strontium-aluminum mixed

oxides SrAl_2O_4 or $\text{Sr}_3\text{Al}_2\text{O}_6$, barium-titanium mixed oxide (BaTiO_3), calcium-titanium mixed oxide (CaTiO_3), $\text{La}_{0.5}\text{Sr}_{0.5}\text{Fe}_{0.9}\text{Ti}_{0.1}\text{O}_{3-0.8}$ or $\text{La}_{0.6}\text{Sr}_{0.4}\text{Fe}_{0.9}\text{Ga}_{0.1}\text{O}_{3-0.7}$; and

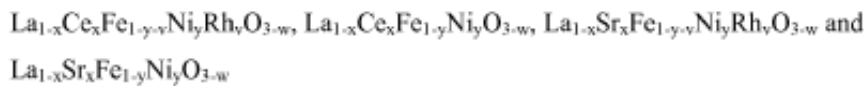
(iii) – optionally, up to 2.5% by volume of a compound (C_{3-4}) produced from at least one chemical reaction represented by the equation:



in which equation F_{C_3} , F_{C_4} and $\text{F}_{\text{C}_{3-4}}$ represent the respective raw formulae of compounds (C_3), (C_4) and (C_{3-4}), and x, y and z represent rational numbers greater than or equal to 0;

(c) and a catalytic layer (C_{C_1}), capable of promoting the reaction of partial oxidation of methane by gaseous oxygen to carbon monoxide and hydrogen, the said catalytic layer (C_{C_1}), of thickness E_{C_1} , having a volume porosity of between 20% and 80%, being adjacent to the said dense layer (C_{D_1}) and consisting of a material (A_{C_1}) comprising, per 100% of its volume:

(i) – at least 10% by volume and at most 100% by volume of a compound (C_5) chosen from doped ceramic oxides which, at the use temperature of between 600°C and 1100°C, are in the form of a crystal lattice having oxide ion vacancies of perovskite phase, of formula (III): , in which compound (C_5) is chosen from compounds of formulae:



in which

$$- 0 < x \leq 0.5;$$

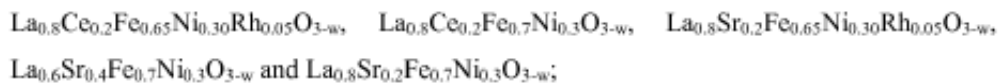
$$- 0 \leq y \leq 0.9;$$

$$- 0 \leq v \leq 0.9;$$

$$- 0 \leq (y + v) \leq 0.9; \text{ and}$$

$$- w \text{ is such that the structure in question is electrically neutral;}$$

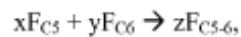
and more particularly those of formulae:



(ii) – optionally up to 90% by volume of a compound (C_6), which differs from compound (C_5), chosen from nickel (Ni), iron (Fe), cobalt (Co), palladium (Pd), platinum (Pt), rhodium (Rh), ruthenium (Ru) or a mixture of these metals, optionally deposited on an oxide or non-oxide ceramic support, in an amount from 0.1% to 60% by weight of the said metal or of the mixture of metals, the said ceramic supports being chosen: either from

boron oxide, aluminum oxide, cerium oxide, silicon oxide, titanium oxide, zirconium oxide, zinc oxide, magnesium oxide or calcium oxide, preferably from magnesium oxide (MgO), calcium oxide (CaO), aluminum oxide (Al₂O₃), zirconium oxide (ZrO₂), titanium oxide (TiO₂) or ceria (CeO₂); mullite (2SiO₂.3Al₂O₃), cordierite (Mg₂Al₄Si₅O₁₈) or the spinel phase MgAl₂O₄; calcium-titanium mixed oxide (CaTiO₃) or calcium-aluminum mixed oxide (CaAl₁₂O₁₉); hydroxylapatite Ca₁₀(PO₄)₆(OH)₂ or tricalcium phosphate Ca₃(PO₄)₂; La_{0.5}Sr_{0.5}Fe_{0.9}Ti_{0.1}O_{3-δ}, La_{0.6}Sr_{0.4}Fe_{0.9}Ga_{0.1}O_{3-δ}, La_{0.5}Sr_{0.5}Fe_{0.9}Ga_{0.1}O_{3-δ} or La_{0.6}Sr_{0.4}Fe_{0.9}Ti_{0.1}O_{3-δ}, or else from materials of the non-oxide type, preferably chosen from silicon carbide (SiC), boron nitride (BN), aluminum nitride (AlN) or silicon nitride (Si₃N₄), sialons (SiAlON);

(iii) – optionally up to 2.5% by volume of a compound (C₅₋₆) produced from at least one chemical reaction represented by the equation:



in which equation F_{C5}, F_{C6} and F_{C5-6}, represent the respective raw formulae of compounds (C₅), (C₆) and (C₅₋₆), and x, y and z represent rational numbers greater than or equal to 0;

so as to constitute an assembly E₁ consisting of three successive layers {(C_{C1}), (C_{D1}), (C_{P1})}, in which:

- at least one of the chemical elements actually present in compound (C₁) is different from one of the chemical elements actually present in compound (C₃).

The dependent claims 2 to 5 define preferred embodiments of the organized assembly of claim 1.

Claims 6 and 7 concern a reactor of non-zero internal volume for the production of synthesis gas, comprising either an organized assembly of tubular form, based on superposed layers of materials as defined in claims 1 to 5, or a combination of several of the said assemblies mounted in parallel.

VII. The arguments of the appellant may be summarized as follows:

The technical problem of the application was to improve the stability of the Catalytic Membrane Reactor (CMR) architecture. In order to solve this problem, the invention proposed an assembly comprising three layers (a dense layer, a porous layer and a catalytic layer) which were characterised in that they all comprised a perovskitic material comprising La and Fe. In this manner a chemical continuity among the layers was assured. Due to said chemical continuity the tendency of fissure and cracks at the interfaces of the layers was reduced and the CMR architecture was stabilized.

The independent claims thus contained the necessary features for solving the underlying technical problem.

A further objection of the examining division, concerning the alleged lack of clarity of the expression "the use temperature", was addressed by amending the claims.

Furthermore, the first instance procedure was flawed by a substantial procedural violation because the decision under appeal was not reasoned. The said decision only made reference to an earlier communication dated 28 April 2010 which in turn referred to the Written Opinion (of the ISA issued in re International application no. PCT/EP2005/056542) of 13 June 2007.

VIII. Requests

The appellant requested that the decision under appeal be set aside and a patent be granted on the basis of the claims filed with letter dated 8 November 2013 as a

main request or, as an auxiliary measure, on the basis of the former main request or on the two auxiliary requests underlying the impugned decision.

Furthermore, the appellant requested that the appeal fee be reimbursed on grounds of a substantial procedural violation.

Reasons for the Decision

1. Article 84 EPC

1.1 The impugned decision of the examining division refers to the division's communication dated 28 April 2010 which in turn refers to the Written Opinion (WO) of the ISA issued *in re* International application no. PCT/EP2005/056542.

1.2 From these documents, it becomes apparent that there were two reasons for the refusal of the application (all then pending requests):

a) An objection under Article 84 EPC (Article 6 PCT in combination with Rule 6.3 a)b)i)ii) PCT), raised in the Written Opinion, *re* item VIII, 4.2. The examining division considered that not all claimed materials - which allegedly represented an almost unlimited number of different perovskite-like compositions - solved the problem posed.

b) An objection under Article 84 EPC, raised in the communication dated 18 April 2010, point 2.2 related to the passage "*...oxides which, at the use temperature, are...*". It was not clear which compounds were embraced by formula II. Also the lack of clarity had an

influence on the claim category. It was not clear whether a product or a process claim was at stake.

1.3 The board considers that the examining division, having taken into account the appellant's arguments in a preceding letter, did not maintain yet another objection under Article 84 EPC (Article 6 PCT) raised in the Written Opinion, *re item VIII, 4.1*, concerning the order of the layers (a), (b) and (c). As the board agrees with the appellant's arguments in this respect, there is no need to take this issue any further.

1.4 As regards the above-mentioned objection a), the appellant correctly pointed out that, in accordance with claim 1 of the new main request, each of the three mandatory layers of the claimed assembly comprises a perovskitic material comprising the elements La and Fe. By virtue of this common presence of a material of similar structure in all the layers, a certain degree of chemical continuity is achieved which limits the risk of delamination and cracks. Said feature is therefore essential for solving the problem of improving the stability of the layer system. In the absence of any counter-evidence, the appellant's argument is persuasive.

Therefore, the objection raised by the examining division in this respect cannot stand.

1.5 Regarding the above-mentioned objection b) relating to the expression "*at the use temperature*", the board concurs with the opinion of the examining division that the claimed material cannot be characterised by reference to a "use temperature" which itself is not defined clearly.

However, the claims as amended in the appeal procedure define a use temperature of between 600°C and 1100°C and are thus considered to overcome this objection.

- 1.6 Therefore, the second objection on which the refusal of the application was based, is also rendered moot by the amended claims.

The decision must therefore be set aside.

2. Alleged procedural violation - reimbursement of the appeal fee

- 2.1 The appellant alleged that the contested decision - using EPO form 2061 for decisions according to the state of the file - was not reasoned within the meaning of Rule 111(2) EPC.

- 2.2 Rule 111(2) EPC stipulates that decisions of the EPO which are open to appeal shall be reasoned. According to the Guidelines for Examination in the EPO, E-X, 5, the reasons given in a decision should be *"complete and independently comprehensible, i.e. generally without references"*.

- 2.3 It follows that in exceptionally plain cases the use of the standard form suggested by the Guidelines for decisions "according to the state of the file" and limiting itself to a reference to an earlier communication could be considered appropriate and sufficient for a decision to be reasoned (see for instance T 0571/03 of 22 March 2006, Reasons, points 13 to 15).

- 2.4 In the present case, the decision under appeal refers to a communication of the examining division dated 28

April 2010 which in turn refers to the section "Item VIII of the Written Opinion (WO) of the ISA" issued in examination of the underlying PCT application (whose text is identical to the corresponding text of the IPER).

Although the board would have preferred an independently fully reasoned, self-consistent decision, it nevertheless concludes that the objections leading to the refusal can be identified and understood from the references in an unambiguous manner and without undue guesswork.

Therefore, in the board's judgment, the reasoning of the decision of the examining division meets the requirements of Rule 111(2) EPC.

2.5 Therefore, a reimbursement of the appeal fee cannot be ordered ((Rule 103(2) EPC).

3. Remittal

Under the circumstances, the board finds it appropriate to exercise its discretion pursuant to Article 111(1) EPC to remit the case to the department of first instance for further prosecution.

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 request for a reimbursement of the appeal fee is refused.

The Registrar:

The Chairman:



C. Vodz

G. Rath

Decision electronically authenticated