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**Datasheet for the decision
of 18 June 2008**

Case Number: T 1333/05 - 3.3.05

Application Number: 98109984.9

Publication Number: 0882670

IPC: C01B 3/36

Language of the proceedings: EN

Title of invention:

Synthesis gas production by ion transport membranes

Patentee:

AIR PRODUCTS AND CHEMICALS, INC.

Opponents:

Praxair Technology, Inc.
L'AIR LIQUIDE S.A.

Headword:

Synthesis gas production by ion transport membranes/AIR
PRODUCTS AND CHEMICAL

Relevant legal provisions:

EPC Art. 56

Relevant legal provisions (EPC 1973):

-

Keyword:

"Request filed during oral proceedings - not admitted"
"Inventive step - no - improvements (accepted) - technical
solution derivable from prior art documents"

Decisions cited:

T 0831/92, T 0270/90

Catchword:

-



Case Number: T 1333/05 - 3.3.05

D E C I S I O N
of the Technical Board of Appeal 3.3.05
of 18 June 2008

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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 9 September 2005
revoking European patent No. 0882670 pursuant
to Article 102(1) EPC 1973.

Composition of the Board:

Chairman: G. Raths
Members: H. Engl
C. Vallet

Summary of Facts and Submissions

I. This appeal lies from the decision of the Opposition Division revoking European patent EP-B-0 882 670. It was held that the claims in accordance with the main, first and second auxiliary requests lacked novelty, and the claims in accordance with the third auxiliary request lacked an inventive step, having regard to document

D3: R. Bredesen *et al*, "*A technical and economic assessment of membrane reactors for hydrogen and syngas production*", United Nations Seminar on Ecological Applications of Innovative Membrane Technology in the Chemical Industry, Cetraro (Italy), 1 - 4 May 1996.

II. The following documents were also cited during the opposition proceedings:

D1: EP-A-0 875 282

D2: EP-A-0 875 284

D4: EP-A-0 673 675

D5: US-A-5 580 497

D6: US-A-5 122 299

D7: EP-A-0 658 366

D8: EP-A-0 739 649

D9: U. Balachandran *et al*, "*Fabrication and Characterization of Dense Ceramic Membranes for Partial Oxidation of Methane*", June 1995, pages 466 to 482, for presentation at the Coal Liquefaction and Gas Conversion Contractors Review Conference, 29 - 31 August 1995, Pittsburgh, Pa., USA

D10: Encyclopedia of Chemical Processing and Design,
Vol. 56, J. J. McKetta (Ed.), Marcel Dekker Inc.,
New York, USA, 1996, pages 188 to 191 and 215 to
229

D12: US-A-5 160 713

D14: EP-A-0 875 281

III. With the letter stating the grounds for appeal the patentee (henceforth: the appellant) filed a main and two auxiliary requests. Further submissions were received with a letter dated 16 May 2008, containing three sets of claims as a first, second and third auxiliary request, respectively, replacing the auxiliary requests previously on file. The appellant's main request was directed at the claims as granted.

The appellant also filed a new document

D16: W. L. Nelson, "*Petroleum Refinery Engineering*",
Fourth Edition, McGraw-Hill Book Company, New York,
1969, pages 650 and 651.

IV. The observations of opponent I (henceforth: respondent I) were received with letters dated 22 June 2006 and 4 June 2008.

V. Opponent II (henceforth: respondent II) did not file observations; it informed the board in a letter dated 22 February 2008 that it did not intend to attend the oral proceedings.

VI. Oral proceedings took place on 18 June 2008 in the absence of respondent II (Rule 115(2) EPC).

At the beginning of the oral proceedings the appellant requested to file, as a new main request, a set of claims based on the second auxiliary request filed by letter dated 16 May 2008, modified in that the features of granted claim 7 were incorporated into claim 1. Respondent I requested that these new claims be rejected as late filed. The board drew attention to the provisions of Articles 12(2) and 13(1) and (3) RPBA. After discussion and deliberation, the board rejected the new claims as late filed.

The appellant then withdrew all pending requests except for the set of claims filed with letter dated 16 May 2008 as the second auxiliary request, which remained the sole, main, request.

VII. Claim 1 in accordance with said main request reads:

"1. A method for the production of a synthesis gas product containing hydrogen and carbon monoxide which comprises:

- (a) providing a reaction zone (37) having an oxidant side (43) and a reactant side (45) which are separated by a solid mixed conducting membrane (47);*
- (b) heating (7, 15) an oxygen-containing oxidant gas feed (5, 11) and introducing the resulting heated oxidant gas feed (17) into the oxidant side (43) of the reaction zone (37) at an oxidant gas feed temperature and an oxidant gas feed pressure;*
- (c) heating (23) a methane-containing reactant gas (19) and introducing the resulting heated reactant gas feed (35) into the reactant side (45) of the reaction zone (37) at a reactant gas feed temperature and a reactant gas feed pressure;*

(d) permeating oxygen from the oxidant side (43) of the reaction zone (37) through the mixed conducting membrane (47) to the reactant side of the reaction zone (37) and reacting the oxygen with the methane-containing reactant gas to form at least hydrogen and carbon monoxide;

(e) withdrawing the synthesis gas product comprising at least hydrogen and carbon monoxide from the reactant side (45) of the reaction zone (37) at a product gas outlet temperature; and

(f) maintaining the reactant gas feed temperature at between 950°F (510°C) and 1400°F (760°C) and the product gas outlet temperature at greater than 1500°F (815°C),

(g) wherein the total pressure at any point in the reactant side (45) of the reaction zone (37) is 13.8 to 22.6 barg (200 - 400 psig) and the total pressure at any point in the oxidant side (43) of the reaction zone (37) is 0.069 - 3.1 barg (1 to 45 psig)."

VIII. The appellant essentially argued as follows:

Claim 1 was novel having regard to D3 which failed to disclose the claimed features (f) and (g) relating to the reactant gas feed temperature and the product gas outlet temperature, and the total pressure on the reactant side and the oxidant side of the membrane, respectively. The lower temperature range as required by feature (f) of the opposed patent (510°C to 760°C) was envisaged in D3 only for a combined steam reforming/autothermal reforming process, not for a stand-alone autothermal reforming process. For both embodiments, *i.e.* a stand-alone autothermal reformer and a combined steam reforming/autothermal reforming

process discussed in D3, the pressure prevailing on the reactant side of the membrane was not disclosed. Pressures of around 40 bar mentioned at page 4, line 2 of D3 related to autothermal reforming, not to a membrane process. Moreover, the pressure ranges recited in feature (g) of claim 1 were not disclosed in D3.

The appellant saw a further difference in feature (c) of the claim, which required the methane-containing reactant gas to be heated prior to introducing it into the reaction zone. In combined reforming, as taught in D3, the gas exiting the prereformer was not heated. By virtue of said feature (c), the claims excluded combined reforming.

As to the other prior art documents, D1 failed to disclose feature (g). A general statement in the introduction of D1 to transport oxygen from a low pressure stream to a high pressure stream related to the background art and should not be read into the process actually disclosed in D1. The entire document D1 was silent on specific pressure ranges.

D2 lacked features (f) and (g) and, additionally, was silent with respect to operating temperatures.

Regarding inventive step, the appellant argued that D3 as the closest prior art provided no motivation to apply high pressures on the reactant gas side of the membrane, as required by feature (g) of the claims. Said feature was also not obvious having regard to other documents relating to the membrane art, such as D4, D5 or D9. In particular, D4 and D5 disclosed atmospheric pressure on both sides of the membrane.

During the oral proceedings, the appellant defined the problem of the invention as providing an improved process of synthesis gas production using a membrane reactor wherein the formation of carbon is prevented or reduced. According to the appellant, the examples of the patent showed that this problem had been solved by the claimed method. The membrane itself operated best at high temperatures. Unfortunately, at high temperatures coke formation occurred, so that a balance was required. This problem was addressed in different ways by the prior art. D3 taught that the preheat temperature should be kept low at 600°C to 630°C and that a portion of the hot gas from the reactor outlet should be recycled. This manner of operating the process reduced reactor productivity. In contrast, the claimed invention taught a different solution consisting of carefully adjusting reactor temperatures and pressures. At high pressures on the reactant side carbon formation increased, therefore this pressure was limited to 27.6 barg, whereas the pressure on the oxidant side was not important for carbon formation. None of the documents addressed this problem and its solution.

Document D10 related exclusively to conventional autothermal reforming. In a conventional reactor, there was only one reactor chamber and hence only one pressure. Therefore, D10 was not combinable with D3.

IX. Respondent I essentially argued as follows:

D3 already disclosed a syngas production process wherein the required oxygen was produced by separation through an oxygen ion permeable membrane.

More specifically, D3 disclosed two specific embodiments, which could be referred to as (1) "*stand-alone autothermal reformer*" and (2) "*combined steam reforming/autothermal reforming process*". The claims of the opposed patent covered both embodiments (1) and (2). At least for embodiment (2) a reactant feed gas temperature of 750°C was disclosed in D3 which fell within the range of claimed feature (f). A reactant gas feed pressure of 40 bar was also disclosed for embodiment (2), it being evident from several statements in paragraph 4.2 (page 12 and page 21) of D3 that the reactant side pressure was considerably higher than the total pressure on the oxidant side (2.5 bar). Said oxidant feed gas pressure of 2.5 bars was disclosed in D3 for embodiments (1) and (2). Hence this feature of the claims was anticipated by D3 and so was the corresponding advantage in terms of power and investment cost savings.

D1 disclosed claimed feature (f) at page 3, lines 13 - 16, a passage found in the "*Background of the Invention*", but clearly also belonging to the invention described in D1. The temperature of the syngas product stream (213) leaving the reactor was disclosed at page 6, lines 31 to 35, as from 500°C to 1200°C, preferably 900°C to 1100°C. The reactant feed temperature was not explicitly mentioned in D1, but it was clear from Figure 2 that oxidant stream (270) and reactant feed

225 had about the same temperature after leaving the heat exchanger (211). As the reactant stream (225) had a preferred temperature of 400°C to 650°C (page 6, lines 7 to 9), claimed feature (f) was also anticipated by D1.

The chemistry of syngas formation in a membrane reactor did not differ from the one in conventional autothermal reforming. The membrane process differed only in the way oxygen was introduced in the reactor chamber. Therefore, it was obvious to apply conventional operating conditions, in particular feed and outlet temperatures and reaction zone pressures.

Such conventional process conditions for autothermal reforming were disclosed in D10 (page 223, Table 4). In fact, all claimed process parameters were the same as in conventional autothermal reforming, except for the oxidant side pressure in the range of 0.069 to 3.1 barg.

According to the opposed patent, the benefit of said feature consisted of power and investment savings due to the fact that compression power only needed to be sufficient to compensate pressure losses. Therefore, the problem to be solved could be considered as the provision of a particularly efficient process.

However, D3 already taught that the extremely low oxygen partial pressure in the reaction zone created a large driving force for oxygen separation through the membrane, so that only compression work to overcome friction losses was needed. Consequently, an air feed pressure of 2.5 bar was considered sufficient in this respect. D12 stated that the pressures in both zones of

a membrane reactor could be subatmospheric, atmospheric or superatmospheric, provided the oxygen partial pressure of zone 1 was greater than the one of zone 2. Hence it was obvious to the skilled person that, for reasons of power saving, the reactant side should be operated at a significantly higher pressure than the oxidant side of the membrane reactor. The ion conducting membranes were known to be able to withstand high pressure differences (D3, page 21 and D9, page 471).

As regards the reactant side pressure, it was known to the skilled person that the reactant feed (natural gas) was usually available at high pressure (paragraph [0057] of the opposed patent). D10 taught conventional reforming for syngas production at elevated pressures.

Moreover, the specific temperature values and pressures claimed in the opposed patent were nothing other than the usual operating parameters of a conventional autothermal reforming process (see D10, page 223, Table 4). Since the chemical reactions were exactly the same in a corresponding autothermal or combined reforming process conducted within a membrane reactor, these features of claim 1 were obvious for this reason alone.

Regarding the problem of carbon formation, the patent did not contain comparative experiments supporting this alleged technical effect. It was not shown that the claimed ranges or limits were critical to avoid or reduce carbon formation. Rather, it was stated in the patent that natural gas was available at pressures of 500 to 1200 psig and it appeared that the claimed

reactant side pressure was chosen merely to match said pressures. The problem of the patent under appeal related thus merely to economic aspects.

The claims therefore lacked an inventive step.

X. Requests of the parties

The appellant requested that the decision under appeal be set aside and the patent be maintained on the basis of the set of claims 1 to 15 filed at the oral proceedings.

Respondent I requested that the appeal be dismissed.

Respondent II did not file any requests.

Reasons for the Decision

1. *Admissibility of the claims filed during oral proceedings*

1.1 Pursuant to Article 13(3) RPBA, amendments sought to be made after oral proceedings have been arranged shall not be admitted if they raise issues which the board or the other party or parties cannot reasonably be expected to deal with without adjournment of the oral proceedings.

1.2 It is established case law of the Boards of Appeal that late filed claims are not to be admitted if they are not found to be clearly allowable and if there is no proper justification for their being late filed. See

T 455/03 (of 5 July 2005; Reasons point 2.1) and the decisions cited there.

According to T 831/92 (of 13 December 1994; Reasons point 3),

"[A]s a matter of principle, the filing of an auxiliary request in opposition proceedings during oral proceedings before a Board of Appeal is contrary to procedural fairness. [...] it is difficult for an Opponent to deal properly with a request not presented in good time before oral proceedings, and an adjournment, even to another day in order to allow further searches, might be appropriate if the request was to be admitted."

- 1.3 In the present case, the appellant asked to restrict claim 1 of the pending second auxiliary request by adding the following feature:

"wherein the methane-containing reactant gas contains water and wherein the water to carbon molar ratio is between 0.5 and 5, the water to carbon molar ratio being defined as the number of molecules of water in the reactant gas divided by the total number of carbon atoms present in the hydrocarbon and carbon monoxide in the reactant gas."

- 1.4 Adding this feature to claim 1 does not clearly answer any question or objection raised by the board or another party in the proceedings. No reason was given as to why it would have been impossible to file this submission earlier. No arguments had been brought forward before as to the technical significance of this

particular feature. It thus confronted the board and the respondent(s) with a substantially changed situation. The proposed claims are therefore to be rejected as inadmissible.

2. *Amendments*

- 2.1 Claim 1 is based on claim 1 as originally filed; the additional features concerning the pressure ranges of 200 to 400 psig; 13.8 - 22.6 barg and 1 to 45 psig; 0.069 - 3.1 barg are disclosed on page 9, paragraph [0056], of the granted patent (page 17, line 2 and page 16, line 10; and page 24, lines 22 to 25 of the application as originally filed).
- 2.2 Incidentally, the board notes that the value of 22.6 barg quoted on page 24, line 23, of the application as originally filed is miscalculated from the value of 400 psig, the correct value of 27.6 being quoted on page 17, line 2. The incorrect value also appears in the claims of the main request. The question of correction of this error is moot since the patent cannot be maintained for the reasons given further below.
- 2.3 With the above proviso concerning the value of 22.6 barg, the requirements of Article 123(2) and (3) EPC are met.

3. *Novelty*

- 3.1 There were no novelty objections raised by the respondents against the claims as amended.

3.2 The board also considers that none of the documents on file discloses all the features of claim 1 in combination, for the following reasons:

3.2.1 Document D3 is an article assessing the potential technical and economic usefulness of membrane reactors for two purposes, namely hydrogen and synthesis gas (syngas) production (see sections 2.1 and 2.2). Section 2.2 (page 3 to page 4) in particular deals with the production of syngas by autothermal reforming:

"Autothermal reforming is a well established synthesis gas production technology where the heat required to maintain the endothermic steam reforming reaction is added by internal partial combustion of a portion of the gas feed with oxygen or air. The autothermal reformer consists of a burner and a fixed catalyst bed for equilibration of the synthesis gas product at temperatures typically in the range of 900 - 1000°C and pressures typically around 40 bar. [...] The autothermal reformer is also used as secondary reformer in combination where the primary reformer is a fired steam reformer converting a portion of the natural gas to synthesis gas."

The author then goes on to state that autothermal reforming, although attractive compared with steam reforming, suffers from high oxygen cost due to the very high investment necessary for a cryogenic oxygen plant. In view of this, he suggests at page 4, fourth paragraph, that

"a cryogenic oxygen plant may be replaced by a high temperature membrane that can separate oxygen from air

and supply it directly into the reactor for converting CH₄ to CO + H₂. The extremely low partial pressure of oxygen in the reaction zone of an autothermal reformer (mole fraction of the order of 10⁻¹⁶ - 10⁻¹⁸), creates a large driving force for oxygen separation in oxygen ion conducting membranes. Using such membranes, only compression work needed to overcome the friction losses in the system would be required.[...]"

Various types of oxygen and hydrogen separating membranes (for instance zeolites, Pd-based membranes, oxygen conducting solid oxide electrolytes, proton conducting solid oxide membranes, and membranes of mixed conducting oxides (perovskites)) are examined below with respect to their usefulness in such an autothermal reactor.

- 3.2.2 Chapter 4 (page 16) investigates the economic feasibility of membrane reactors, whereas chapter 4.2. concentrates on syngas production:

"4.2. Economic assessment of syngas production

Estimations have been performed in order to give an indication of the technical and economic potential of the autothermal and combined reforming processes applying oxygen ion conducting membranes. [...] The reformer feed temperature has been set to 800°C and 750°C for the autothermal and combined reforming processes, respectively. [...] Furthermore, an air delivery pressure of 2.5 bara has been chosen."

- 3.2.3 In the opinion of the board, this economic assessment concerns autothermal or combined reforming processes as

generally described on pages 3 and 4, modified in that oxygen is produced by membrane permeation and directly supplied to the reactor, rather than in a cryogenic oxygen separating plant. D3 thus discloses a process for the production of syngas containing hydrogen and carbon monoxide in a membrane reactor.

Also disclosed on page 20, fourth full paragraph, "*hot synthesis gas product from the reformer outlet ha[s] a temperature of around 950°C*", is the second part of feature (f), concerning the product gas outlet temperature of greater than 1500°F (815°C).

The pressure of 40 bars mentioned at page 4, line 2, refers in the opinion of the board to the typical pressure in a fixed catalyst bed reactor for equilibration of the synthesis gas at 900°C to 1000°C. It cannot automatically be equated to the pressure on the fuel side of a membrane reactor which, thus, remains unknown. Even if the latter is in practice possibly higher than 2.5 bars (the pressure on the oxidant feed side reported on page 16, third paragraph), this cannot replace a positive disclosure of the said feature in D3. The value of 40 bars mentioned at page 4, line 2, of D3 clearly refers to a different embodiment (syngas production in autothermal reformers, not in membrane reactors) and cannot, thus, be combined with the membrane reforming processes described on page 16, as far as novelty is concerned. In any event, D3 fails to disclose the specific pressure ranges of 13.8 to 27.6 barg (see point 2.2 above) on the reactant side and of 0.069 to 3.1 barg on the oxidant side of the reaction zone, as recited in feature (g) of claim 1.

Moreover, whereas a reactant feed temperature of 750°C, falling within the temperature range claimed in feature (f) of the opposed patent, is reported for a **combined** reforming process, the temperature of the autothermal ("**stand-alone**") reformer is 800°C, outside feature (f) of the claims. Respondent I argued that the opposed patent did not exclude combined reforming processes and that, consequently, feature (f) was satisfied. However, the board does not find this argument convincing, because there is no hint in the description or the claims of a combined reforming process. Moreover, feature (c) (heating of the methane-containing reactant gas prior to its introduction into the reaction zone) would not be present or necessary in combined reforming, as convincingly pointed out by the appellant.

For these reasons, the claimed subject matter is novel having regard to D3.

3.3 Document D1 does not disclose a reactant feed temperature of stream 225 in the range of 510°C to 760°C and the pressure ranges claimed in feature (g) of claim 1. Therefore, the claims are novel having regard to D1.

3.4 D2 has been cited as novelty-destroying in opposition proceedings against claims then on file. However, respondent I did not rely any more on this document in its submissions during the appeal procedure.

Document D2 discloses at page 8, line 43 to page 9, line 2 and Figure 1 an embodiment of a membrane reactor for the production of syngas, wherein the pressure of the oxidant side (first zone 40) is 1 to 40, preferably

1 to 10 atm, and the pressure on the fuel side (second zone 41) is typically 1 to 100, preferably 1 to 40 atm. In the board's view, an unambiguous disclosure of feature (g) as claimed in claim 1 of the patent in suit cannot be derived from these broad ranges.

Therefore, novelty over D2 is also acknowledged.

- 3.5 Other documents relevant for novelty have not been cited and are not apparent to the board.

D4 concentrates on the manufacture of the perovskitic oxygen permeable membrane, but lacks disclosure of reactant feed temperatures and pressures.

D5 discloses a Sr-Fe-Co-Perovskite composition for the manufacture of an oxygen permeable membrane and a reactor comprising the same. Reactor temperatures of from approximately 400°C to 900°C were investigated and methane conversions very near to 100% observed. The document is however silent on feed and reactant gas temperatures in syngas production.

Documents D7 and D8 concern the integrated production of oxygen and electric power and are without particular interest for the claimed subject matter.

D14 and D15 disclose an integrated solid electrolyte ionic conductor separator-cooler and a solid electrolyte ionic conductor reactor, respectively, comprising oxygen ion transporting membranes. According to D15, a reactant gas stream may be contacted and reacted with the oxygen transported through the membrane. Syngas production is briefly mentioned in D14

(page 4, lines 30; page 10, lines 27 to 36), but specific process conditions are not disclosed, except for the operating range of about 500°C to 1100°C.

3.6 The requirements of Article 54(1)(2) EPC are thus met.

4. *Inventive step*

4.1 The priority date of 6 June 1997 can be acknowledged. Consequently, documents D1, D2, D14 and D15 (all published on 4 November 1998) belong to the prior art under the provisions of Article 54(3) EPC and are not taken into account for inventive step.

4.2 In accordance with the appellant and respondent I, the board considers D3 to represent the closest prior art because it deals - *inter alia* - with the production of syngas in a membrane reactor. The relevant contents of the document have already been analysed in detail above (see point 3.2.1).

4.3 During oral proceedings the appellant defined the problem as providing an improved process of syngas production using a solid mixed conducting membrane which reduces or prevents coke formation.

4.4 As a solution to this problem, the patent in suit proposes a method for the production of a synthesis gas which is characterised by features (f) and (g).

4.5 According to the description of the patent under appeal and the submissions of the appellant, the following effects and improvements are achieved by these features:

- 4.5.1 Paragraph [0084] of the patent in suit states that the pressure on the reaction side should be higher than on the oxidant side (feature (g)) in order to save power and capital cost, because the compression work on the oxidant side is minimised and a blower may be used instead of a compressor (cf. also paragraphs [0056] to [0060]).
- 4.5.2 Regarding feature (f), the patent in suit discloses that maintaining the reactant feed temperature below the threshold temperature (of 760°C) prevents carbon deposition and allows the use of unlined metal piping and manifolding at the reactor inlet (paragraph [0083], last lines thereof).
- 4.6 The board considers that a saving of power and capital cost by reducing the pressure on the oxidant side is already achieved by the prior art according to D3. As mentioned above, said document discloses on page 16 a membrane process for syngas production wherein the air delivery pressure is as low as 2.5 bara (= 1.5 barg). The oxygen pressure can be kept low because the extremely low partial pressure of O₂ in the reaction zone of an autothermal reactor creates a large driving force for oxygen separation in the oxygen ion conducting membrane (see D3, page 4, second full paragraph, second sentence). Consequently, there is no need to increase oxidant feed pressure beyond what is required to overcome pressure drop in the device. A low oxygen feed pressure allows savings on compression equipment investment and compression power cost. It is also expressly stated at page 4 (second full paragraph, third sentence) that "*using such membranes, only compression work needed to overcome the friction losses*

in the system would be required". This is a clear indication that, for instance, a power saving blower can be used instead of a compressor.

This particular effect of the claimed invention is therefore not to be taken into account for formulating the technical problem.

- 4.7 As to the question whether this problem has actually been solved, the appellant referred to the examples of the patent in suit. By balancing the temperature requirement of the membrane (which operated best at high temperatures) with sufficiently low reactant gas feed temperatures the formation of coke was prevented.
- 4.8 Although an experimental comparison has not been made, the board assumes in favour of the appellant that the technical problem of reducing or preventing soot formation has been solved and an improvement been achieved, compared with an autothermal reforming process using a membrane reactor operated under the process conditions suggested in D3.
- 4.9 It remains to be decided whether or not the proposed solution to that objective problem, namely the method according to claim 1, was obvious to the skilled person in view of the state of the art.
- 4.9.1 Firstly, the board notes that no statement in the description or in the appellant's argumentation links the problem of coke formation to the claimed pressure range on the oxidant side (0.069 to 3.1 barg). It has also not been shown that the claimed product gas outlet temperature of greater than 815°C contributed to the

solution of the problem. Respondent I argued that the temperature of the reactants and of hot nitrogen leaving the membrane reactor would inevitably be higher than the feed temperature. Since a reactant feed temperature of 750°C was disclosed in D3, respondent I concluded that the reformer exit temperature would inevitably be higher than this temperature. The board finds this argument convincing.

The board has still to investigate whether or not it was obvious to select the claimed reactant gas feed temperature of between 510°C and 760°C and the claimed reactant side pressure of 13.8 to 27.6 barg (cf. point 2, second paragraph above) in order to solve the problem posed.

4.9.2 According to the submission of the appellant, the prior art of D3 taught that the preheating temperature should be kept below 600°C to 630°C to avoid hydrocarbon cracking (page 20, fourth full paragraph). Partial recycling of the hot syngas product from the reactor was recommended in D3 to raise the feed temperature above that preheating temperature. Such recycling inevitably reduced the productivity of the reactor. In contrast, the invention solved these problems by adjusting the temperatures to the claimed ranges.

4.9.3 The main argument of respondent I was that the chemistry of the process of syngas formation was the same, no matter whether it was carried out in a conventional autothermal reformer (ATR) or in a membrane reactor. Said chemistry was characterised by the well-known chemical reaction equilibria (1) to (9) as shown on page 8 of the opposed patent. For the

desired conversion and reaction of a hydrocarbon feed to syngas it would have been obvious to choose the same pressure and temperature conditions as in a conventional ATR.

4.9.4 Conventional ATR processes are described in some detail in D10 (pages 216 to 223). Typical operating conditions, compared with claim 1 of the opposed patent, are (page 223, Table 4):

	D10 Table 4	patent claim 1, feature f
HC feed	200-650°C	510-760°C
O2 feed	150-600°C	-
exit temp	850-1100°C	>815°C
pressure, reactant side	20-70 bar	13.8 to 27.6 barg*
pressure, oxidant side	-	0.069-3.1 barg

4.9.5 These conventional operating conditions satisfy feature (f) insofar as the product gas outlet temperature is greater than 815°C and the reactant gas feed temperature overlaps with the claimed range. The reactant side pressure of 20 to 70 bar also overlaps with the range given in D10.

Respondent I also referred to D3, page 20, where it is said that *"in ATR the natural gas/steam mixture cannot be heated to a higher temperature than 600 to 630°C (when a pre-reformer is used) due to risk of carbon formation mainly because of hydrocarbon cracking."* Since according to feature (f) of the opposed patent the hydrocarbon feed temperature may be as high as 760°C, respondent I argued that thermal cracking and decomposition would be unavoidable such that the

* see point 2, second paragraph above

claimed benefit could not be achieved over the full range of the claim.

This question can be left aside, however, because it is sufficient for the board to note that the claimed reactant gas feed temperature of from 510°C to 760°C is by and large consistent with the conventional range of from 200°C to 650°C disclosed in D10. Further to the problem of soot or carbon formation, D10 also teaches at page 223, second paragraph, that "*low pressures (< 12 bar) may not be applied due to soot formation which cannot be eliminated through steam addition or burner design.*" This condition is also observed in claim 1, feature (g), of the opposed patent.

- 4.9.6 The appellant argued that documents D3 and D10 were not obviously combinable, because the latter related to autothermal reforming without membrane and thus to a completely different technique.

The board does not find this argument convincing. A membrane reactor for syngas production differs only in the way oxygen is supplied into the reactor, the membrane serving a dual purpose of separating oxygen from air and providing a support for the catalyst. There is no substantial difference in the underlying chemistry of the process, the reactants and the chemical reactions in both reactors being the same. The skilled person would therefore have considered D10 as being pertinent to syngas processes carried out in a membrane reactor as taught in D3.

4.9.7 The board concludes that the process parameters appearing in claim 1 are either known from D3 or suggested by the prior art autothermal reforming processes in order to prevent or reduce coke (carbon, soot) formation.

The subject-matter of claim 1 is therefore derivable in an obvious manner from D3 and D10.

4.10 To summarise, the method according to claim 1 does not involve an inventive step (Article 56 EPC).

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar

The Chairman

C. Vodz

G. Rath