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D E C I S I O N
of 19 May 2003

Case Number: T 0931/00 - 3.3.5

Application Number: 94119787.3

Publication Number: 0658367

IPC: B01D 53/22

Language of the proceedings: EN

Title of invention:

Integrated high temperature method for oxygen production

Patentee:

AIR PRODUCTS AND CHEMICALS, INC.

Opponent:

Praxair Technology, Inc.

Headword:

Oxygen/AIR PRODUCTS

Relevant legal provisions:

EPC Art. 56, 123(2)

Keyword:

"Unallowable extension (main request and auxiliary requests 1 to 20) - creation of new parameters"

"Inventive step (auxiliary request 21) - yes, no incentive for the claimed solution of the problem in the prior art"

Decisions cited:

T 0201/83, T 1067/97, T 0526/92

Headnote:

Although figures in examples may, under specific conditions, be used to limit a range, which was already present in the original application, they cannot be used to define an entirely new relationship between parameters which were never linked before. Such arbitrary new links between existing parameters introduce new matter, contrary to the requirements of Article 123(2) and 100(c) EPC (points 2.1 and 2.2).



Case Number: T 0931/00 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 19 May 2003

(Opponent)

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(Proprietor of the patent)

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Decision under appeal:

Interlocutory decision of the Opposition
Division of the European Patent Office posted
13 July 2000 concerning maintenance of European
patent No. 0658367 in amended form.

Composition of the Board:

Chairman: R. K. Spangenberg

Members: G. J. Wassenaar

H. Preglau

Summary of Facts and Submissions

I. The appeal is from the decision of the Opposition Division to maintain European patent No 0 658 367 in amended form, with claims 1 to 22 filed with letter of 9 June 2000.

II. Claim 1 thereof was identical to claim 1 as granted and read as follows:

"A process for recovering oxygen from an oxygen-containing gas mixture comprising the steps of:

- (a) compressing said oxygen-containing gas mixture;
- (b) heating the resulting compressed gas mixture of step (a);
- (c) passing the resulting compressed and heated mixture of step (b) into a membrane separation zone comprising one or more oxygen-selective ion transport membranes, and withdrawing therefrom a hot high-purity oxygen permeate stream and a hot oxygen-containing non-permeate stream;
- (d) further heating said non-permeate stream; and
- (e) passing the further heated non-permeate stream through an expansion turbine to generate shaft power and withdrawing therefrom a turbine exhaust stream; wherein the operating temperatures of said membrane separation zone and said expansion turbine are independently maintained by controlling the rate of heat addition in steps (b) and (d), whereby said membrane separation zone and said expansion turbine are thermally delinked for maximum efficiency in recovering said oxygen and the temperature of said zone is higher than that of said exhaust stream."

III. The claims as granted were attacked on the grounds of lack of novelty, lack of inventive step, insufficient disclosure and unallowable extension. In the statement of the grounds of appeal, the appellant maintained its inventive step objection and its objection of unallowable extension within the meaning of Article 123(2) EPC. With respect to the latter it was argued that the feature of claim 1 as granted, that the temperature in the oxygen separation zone is higher than that of the exhaust stream of the expansion turbine, was not disclosed in the application as originally filed. Lack of inventive step was argued on the basis of

D2: JPL Publication D-7790, in combination with

D1: US-A-5 245 110.

Later in the proceedings further reference was made to the Article "Gasturbine" in Meyers Lexikon der Technik und der exakten Naturwissenschaften, and

D3: Advanced Oxygen Separation Membranes, Topical Report by John D. Wright and Robert J. Copeland. Report No. TDA-GRI-90/0303, September 1990.

IV. The respondent (patentee) refuted the arguments of the appellant. With respect to the objection of unallowable extension it was argued that the added feature was implicitly disclosed in the application as originally filed. With respect to inventive step it was argued that the skilled person had no reason to modify the integrated process according to Figure 9-4 of D2 and that it was not obvious to integrate specific features

from D1, presented in a different context, into the process outlined in Figure 9-4 of D2. The process disclosed in Figure 5.12 of D3 clearly showed a direct link between the heating of the oxygen containing gas stream passed to the separation zone with the ceramic membrane and the oxygen lean stream passed to the gas turbine. There was no document on file suggesting a delinking between the heating of these gas streams, let alone how to do it.

With the letter dated 9 May 2003, 21 sets of claims were filed titled main request and auxiliary requests 1 to 20. The claims according to the main request corresponded to the claims as maintained by the Opposition Division. Claim 1 according to auxiliary requests 1 and 2 and 15 to 17 comprised the feature that the temperature of the membrane separation zone was higher than that of the exhaust stream of the expansion turbine. In auxiliary requests 3 to 14 and 18 to 20, said feature was replaced by specified temperature ranges for the membrane separation zone and the exhaust stream. With respect to the temperature ranges in auxiliary requests, which were partly based on examples of the patent in suit, it was argued that such a limitation was in conformity with Article 123(2) EPO. In this respect reference was made to the case law T 201/83 and T 1067/97.

During oral proceedings a further set of claims 1 to 10 (auxiliary request 21) was filed. These claims corresponded to claims 13 to 23 as maintained by the Opposition Division. The set comprised three independent claims 1, 5 and 8, which read as follows:

Claim 1:

"A process for recovering oxygen from an oxygen-containing gas mixture comprising the steps of:

- (a) compressing said oxygen-containing gas mixture;
- (b) dividing the resulting compressed gas mixture of step (a) into a first and a second compressed gas stream;
- (c) heating said first compressed gas stream, heating said second compressed gas stream and combining the resulting heated streams into a combined feed stream;
- (d) passing said combined feed stream into a membrane separation zone comprising one or more oxygen-selective ion transport membranes;
- (e) withdrawing from said membrane separation zone a high-purity oxygen permeate stream and an oxygen-containing non-permeate stream;
- (f) further heating said non-permeate stream;
- (g) cooling the resulted heated non-permeable stream of step (f) by indirect heat exchange with said second compressed gas stream, thereby providing heating for said second compressed gas stream in step (c); and
- (h) heating the resulting cooled non-permeate stream of step (g), passing the resulting heated stream through an expansion turbine to generate shaft power and withdrawing therefrom a turbine exhaust stream;

wherein the operating temperatures of said membrane separation zone and said expansion turbine are independently maintained by controlling the rate of heat addition to each of said first and second compressed gas streams in step (c) and to said resulting cooled non-permeate stream in step (h), whereby said membrane separation zone and said

expansion turbine are thermally delinked for optimum efficiency in recovering said oxygen."

Claim 5:

"A method of operating a process for recovering oxygen from an oxygen-containing gas mixture which comprises:

(a) compressing said oxygen-containing gas mixture;

(b) heating the resulting compressed gas mixture of step (a) by combusting said mixture with a fuel in a first direct-fired burner;

(c) passing the resulting compressed and heated stream of step (b) into a membrane separation zone comprising one or more oxygen-selective ion transport membranes, and withdrawing therefrom a hot high-purity oxygen permeate stream and a hot oxygen-containing non-permeate stream;

(d) measuring the temperature of said hot oxygen-containing non-permeate stream, comparing the measured temperature with a first set point temperature, and utilizing the difference between the measured temperature and the first set point temperature to correct the firing rate of said first direct-fired burner, thereby maintaining said first set point temperature;

(e) further heating said non-permeate stream by combusting said stream with a fuel in a second direct-fired burner;

(f) passing the further heated non-permeate stream of step (e) through an expansion turbine to generate shaft power and withdrawing therefrom a turbine exhaust stream; and

(g) measuring the temperature of said further heated non-permeate stream prior to said expansion turbine,

comparing the measured temperature with a second set point temperature, and utilizing the difference between the measured temperature and the second set point temperature to correct the firing rate of said second direct-fired burner, thereby maintaining said second set point temperature;

whereby the operating temperatures of said membrane separation zone and said expansion turbine are independently maintained, and whereby said membrane separation zone and said expansion turbine are thermally delinked for optimum efficiency in recovering said oxygen."

Claim 8:

"A method of operating a process for recovering oxygen from an oxygen-containing gas mixture which comprises:

- (a) compressing said oxygen-containing gas mixture;
- (b) heating the resulting compressed gas mixture of step (a) by indirect heat exchange with a hot combustion gas stream, yielding a cooled combustion gas stream;
- (c) passing the resulting compressed and heated stream of step (b) into a membrane separation zone comprising one or more oxygen-selective ion transport membranes, and withdrawing therefrom a hot high-purity oxygen permeate stream and a hot oxygen-containing non-permeate stream;
- (d) combusting said hot oxygen-containing non-permeate stream with a fuel in a first direct-fired burner to yield said hot combustion gas stream;
- (e) measuring the temperature of said hot oxygen-containing non-permeate stream, comparing the measured temperature with a first set point temperature, and

utilizing the difference between the measured temperature and the first set point temperature to correct the firing rate of said first direct-fired burner, thereby maintaining said first set point temperature;

(f) heating said cooled combustion gas stream by combusting said stream with a fuel in a second direct-fired burner to yield a high temperature combustion product;

(g) passing said high temperature combustion product through an expansion turbine to generate shaft power and withdrawing therefrom a turbine exhaust stream; and

(h) measuring the temperature of said high temperature combustion product prior to said expansion turbine, comparing the measured temperature with a second set point temperature, and utilizing the difference between the measured temperature and the second set point temperature to correct the firing rate of said second direct-fired burner, thereby maintaining said second set point temperature;

whereby the operating temperatures of said membrane separation zone and said expansion turbine are independently maintained, and whereby said membrane separation zone and said expansion turbine are thermally delinked for optimum efficiency in recovering said oxygen."

- V. The appellant (opponent) requested that the decision under appeal be set aside and European patent No. 0 658 367 be revoked.

The respondent (patentee) requested that the appeal be dismissed and that the patent be maintained as per decision of the Opposition Division (main request) or

that the decision under appeal be set aside and that the patent be maintained according to one of the auxiliary requests 1 to 20 as filed with the letter dated 9 May 2003 or the auxiliary request 21 as filed during the oral proceedings, in numerical order.

Reasons for the Decision

1. The appeal is admissible
2. *Amendments (Article 123(2) EPC)*
 - 2.1 Claim 1 of the main request comprises the feature that the temperature in the oxygen separation zone is higher than that of the exhaust stream of the expansion turbine. This feature is not explicitly disclosed in the application as originally filed. In the summary of the invention and the general part of the detailed description of the invention in the original application nothing is said about a relationship between the temperature ranges for the temperature of the separation zone and the exhaust stream. According to the first embodiment of the invention, as illustrated by Figure 1, air is added to the separation zone at a temperature of 800 to 2000°F, preferably 1000 to 1600°F (page 8, lines 18 to 20) and exhaust gas is withdrawn at a temperature of 200 to 400°F (page 10, lines 23 to 26). For the embodiment illustrated by Figure 2 no temperature ranges are mentioned. For the embodiment illustrated by Figure 3 the same membrane operation temperatures are mentioned as for the embodiment according to Figure 1. The exhaust temperature is however different and is indicated as

being 200 to 1100°F (page 16, first paragraph). Thus in the embodiment according to Figure 3 the exhaust temperature may be higher than the temperature in the separation zone. It is true that in the examples, which are all performed according to the embodiment according to Figure 3, the exhaust temperature is always lower than the temperature in the separation zone. Figures in examples may, under specific conditions, be used to limit a range, which was already present in the original application, but cannot be used to define an entirely new relationship between parameters which were never linked before. Otherwise any new relationship between parameters could be introduced which accidentally is not violated by the examples in the application. In the Board's view such arbitrary new links between existing parameters clearly introduce new matter. The appellant's argument that from the general statement in the summary of the invention that the compressed oxygen-containing gas mixture can be preheated by indirect heat exchange with the hot turbine exhaust gas prior to final heating and flow to the membrane (page 5, lines 24 to 26), as is the case in the embodiment according to Figure 3, it follows that the temperature of the exhaust gas must be lower than the temperature in the separation zone, cannot be accepted. According to Figure 3 only a part of the compressed air is preheated by the exhaust gas. This side stream is reunited with the rest of the compressed air before the combined stream is further heated to the temperature needed in the separation zone. This side stream may be heated to a temperature higher than the temperature in the separation zone, whereas the combined stream may have a temperature lower than the temperature of the separation zone. In the Board's view

there is thus not a general implicit disclosure that the temperature of the exhaust gas should be lower than the temperature of the separation zone. Thus claim 1 of the main request violates the requirements of Article 123(2) and 100(c)EPC. The same applies to claims 1 according to auxiliary requests 1, 2, and 15 to 17 which also comprise that newly introduced relationship between the temperatures of the separation zone and the exhaust gas.

- 2.2 In claim 1 of auxiliary request 3 the said relationship is replaced by the requirements that the temperature of the separation zone is 538°C or higher and the temperature of the exhaust stream is 537°C or lower. The latter temperature requirement is only based on the examples according to the third embodiment of the invention. The temperature of 537°C is the highest temperature of exhaust stream 21 cited in the tables (Table 4). As indicated above, the originally disclosed temperature range for the third embodiment is 200 to 1100°F (93 to 593°C). In the absence of a lower limit a new parameter range has been created, arbitrarily based on a single example. Because of its arbitrary nature the creation of that parameter range introduces new matter in the same way as the creation of arbitrary links between existing parameters as discussed before under point 2.1. Although under specific circumstances an existing parameter range may be limited to values disclosed in an example, see T 201/83 (OJ EPO 1984, 481) and T 1067/97 of 4 October 2000, cited by the respondent, in the Board's view this case law may not be so extended as to allow the creation of a new parameter range by defining its upper limit by selecting a single value from an example. This view is

in agreement with the findings in decision T 526/92 (points 5.3.1 and 5.3.6 and the catchword). Thus the new parameter defined in claim 1 of auxiliary request 3 introduces new matter. The same newly introduced parameter range is present in claim 1 of auxiliary requests 4 and 5.

- 2.3 In claim 1 of auxiliary request 6 the temperature of the separation zone is defined as being from 538 to 1093°C and temperature of the exhaust stream as being from 93 to 537°C. The latter temperature range is a new range construed from the lower value of the range of 200 to 1100°F originally disclosed for the exhaust stream temperature of the third embodiment and the highest value of the temperature of said stream cited in the examples according to the third embodiment. This range is different from the range of 200 to 400°F disclosed for the exhaust stream of embodiment 1. The respondent's allegation during oral proceedings that the range of 200 to 400°F disclosed for the first embodiment was a mistake and should have been equal to that disclosed for the third embodiment might be correct, but his mistake is not apparent from the original disclosure. The original application provides the unambiguous information that for the different embodiments different exhaust stream temperatures are foreseen so that a temperature range for one embodiment may not be generalized to all embodiments. Thus, even if it was allowable to define the new range of 93 to 537°C within the framework of the third embodiment, it is certainly not allowable under Article 123(2) EPC to put it in a claim without the other essential features of the third embodiment as recited in claim 13.

Auxiliary requests 7 and 8, comprise a claim 1 with the same amendment as in claim 1 of auxiliary request 6.

2.4 In claim 1 of auxiliary request 9 the temperature of the exhaust stream is defined as being 492°C or lower. The new upper value is based on a single value from an example according to the third embodiment (Table 5) and creates a new parameter range. This is not allowable for the same reasons as given above under 2.2 with respect to auxiliary request 3. The same applies to auxiliary requests 10 and 11, comprising a claim 1 with the same parameter range.

2.5 In claim 1 of auxiliary requests 12 to 14 the temperature of the exhaust stream is defined as being 93 to 492°C. This new range is not allowable for the same reasons as given above under point 2.3 for the range of 93 to 537°C.

2.6 In claim 1 of auxiliary requests 18 to 20 the temperature of the exhaust stream is defined as being 93 to 204°C. This range is based on the range of 200 to 400°F originally disclosed for the temperature of the exhaust stream in the first embodiment, but is now generalised to apply to all embodiments covered by claim 1. Without the essential further features of said embodiment in claim 1, these requests are rejected for similar reasons as given above under point 2.3 for auxiliary requests 6 to 8.

2.7 For these reasons neither the main request nor the auxiliary requests 1 to 20 meet the requirements of Article 100(c) and 123(2) EPC, so that these requirements must fail.

2.8 The claims of auxiliary request 21 are limited to claims 13 to 23 as maintained by the Opposition Division. No objections are apparent or have been raised under Article 100(c) EPC against these claims. The obviousness objection, however, was maintained against claims 1 to 10 according to auxiliary request 21.

3. *Inventive step (auxiliary request 21)*

3.1 Auxiliary request 21 comprises three independent claims; claims 1, 5 and 8. They all relate to processes for the recovery of high-purity oxygen from a gas mixture by separation through an ion transport membrane. In the Board's opinion D3 represents the closest prior art. D3 is regarded to be a more appropriate starting point for an inventive step analysis than the processes disclosed in paragraph 9.7 of D2 because in the latter the product is only electrical power provided by the combustion of carbon monoxide formed by the gasification of coal by oxygen. Thus in D2 oxygen is only produced as an intermediate product, which is directly and completely used in the gasification process. In the processes according to the patent in suit and D3 high-purity oxygen is produced as a product which is freely available for any further use.

3.2 D3, acknowledged in the patent in suit, discloses a process for the recovery of oxygen from air by pressurizing and separation in a pressure activated oxygen selective ion transport membrane, whereby the non-permeated, oxygen lean stream is used for the combustion of natural gas and whereby the combustion

gases are expanded in a gas turbine to generate power and to drive the compressor for compressing the air (page 55, Figure 5.12). The compressed air is heated to the working temperature of the membrane separation zone by indirect heat exchange with the combustion gases in the combustion chamber. It is contemplated to heat the inlet air to the membrane zone to essentially the same temperature of 1300 K as the inlet temperature of the turbine. In the process as outlined in said Figure 5.12 the heating of the compressed air is directly coupled to the heating of the gas for the gas turbine. In this way the heat distribution to the separation cell and the turbine is fixed and operation temperatures cannot be adapted to changing conditions in the separation zone and/or turbine. In agreement with the explanation of the background of the invention in the patent in suit (page 3, lines 17 to 23), the problem underlying the invention is to be seen in the provision of a more flexible process for the production of oxygen, which maximizes energy utilization. The invention proposes to solve this problem in three different but related ways according to claim 1, claim 5 and claim 8 respectively. The different processes have in common that the operating temperatures of the membrane separation zone and the expansion turbine are independently maintained, whereby the membrane separation zone and the expansion turbine are thermally delinked. This thermal delinking allows both the separation zone and the turbine to be operated at their optimum performance temperature and renders the process more flexible. The Board is therefore satisfied that the claimed processes actually solve the above mentioned problem.

3.3 According to claim 1 said problem is solved by the division of the compressed oxygen containing gas mixture (air) into a first and a second compressed gas stream, which are separately heated and thereafter combined to a feed stream for the separation zone (features b and c), cooling of the heated non-permeate stream by indirect heat exchange with the second compressed gas stream (feature g) and further heating the cooled non-permeate gas stream before it is passed to the expansion turbine. By the splitting of the compressed air and the separate heating thereof a delinking from the heating of the separation zone from the heating of the oxygen lean stream passed to the expansion turbine is obtained.

The splitting of the compressed air into two streams is shown in Figure 2 of D1 but for a completely different purpose in a completely different process. D1 does not relate to the production of high-purity oxygen but to the production of an oxygen enriched gas mixture containing at least 40 mol % nitrogen (abstract and column 14, lines 11 to 34). The split streams are also not united; one is fed to an oxygen separation zone, whereas the other is fed to the burner for the gas turbine.

In the process according to Figure 9-4 of D2 the compressed air is not split and is heated by burning a side stream of the fuel for the gas turbine combustor. The fuel for the combustor is carbon monoxide generated by gasifying coal by the oxygen generated in the air separation unit. The amount of fuel is thus dependent on the amount of oxygen generated in the air separation unit, so that the heating of the gas stream to the gas

turbine is not delinked from the heating of the compressed air fed to the air separation unit. D1 and D2 therefore do not suggest the process according to present claim 1.

The Board does not dispute that a skilled person would operate the membrane and the gas turbine at their optimum temperature, as suggested in D2, annex 10-12 and D1, column 16, lines 19-28 respectively, but cannot accept the appellant's position that features (b), (c) and (g) are obvious measures as a consequence of routine further development by the skilled person trying to obtain that desired result. In the absence of any hints towards said features in the available prior art documents, the appellant's position seems to be not free from hindsight considerations.

- 3.4 According to claim 5 said problem is solved by heating the compressed gas mixture by combusting the mixture with a fuel in a first direct-fired burner, whereby the temperature is regulated by the firing rate of the burner (features b and d) and heating the non-permeate gas stream by combustion in a second direct-fired burner before it is passed to the expansion turbine, whereby the temperature is regulated by the firing rate of the burner (features e, f and g). By the use of two independent burners the heating of the separation zone is delinked from the heating of the oxygen lean stream passed to the expansion turbine.

The use of two direct-fired burners is disclosed in D2, but for the reasons given herein before under point 3.3 their working is not delinked. Moreover, the process according to Figure 9-4 of D2 is a highly integrated

process for the production of electrical power and has no relationship with the above-mentioned problem underlying the invention. In order to solve the problem the skilled person had no reason to select an isolated feature from D2 and to combine that with the process according to Figure 5.12 of D3.

D1 discloses that the compressed air is cooled before it enters the membrane zone comprising preferably a semi-permeable membrane of an organic polymer (column 19, lines 26 to 30 and column 20, lines 3 to 17). In D1 the use of an inorganic membrane is also contemplated, in which case the heated compressed gas may be contacted with the membrane at elevated temperature (column 21, lines 3 to 9). By adiabatic compression the compressed air is automatically heated. D1 does not disclose a further heating, let alone by a direct-fired burner.

The other documents on file do not contain any indication towards using an additional direct-fired burner in the method according to D3 either. Thus the process according to present claim 5 does not follow in an obvious way from the prior art.

- 3.5 According to claim 8 said problem is solved by heating the compressed oxygen containing gas mixture by indirect heat exchange with a hot combustion gas stream yielding a cooled combustion gas stream, combusting the hot non-permeate stream with a fuel in a first direct-fired burner, whereby the temperature of the hot non-permeate stream is regulated by the firing rate of the burner (features b, d and e) and heating the cooled combustion gas stream by combusting with a fuel in a

second direct-fired burner to yield the high temperature combustion product which is passed through the expansion turbine, whereby the temperature of the combustion product is regulated by the firing rate of the burner (features f, g and h). By the use of the two independent burners the heating of the separation zone is delinked from the heating of the oxygen lean stream passed to the expansion turbine. The use of two separate direct-fired burners in the oxygen lean stream, with an indirect heat exchange with the compressed air between the two burners in order to operate the separation zone and the gas turbine at their optimum temperature, is not disclosed or suggested in any of the cited documents. The appellant has in fact not pointed to any specific document with respect to the combination of features according to claim 8, but only argued that the additional features over claim 1 as maintained by the Opposition Division are routine measures of heating and temperature control (page 12, point C, of the grounds of the appeal). The Board does not dispute that the heating and temperature control measures are well known in the art, but holds that in the absence of any guidance in the prior art documents to solve the above-mentioned problem, the combination of heating and temperature control steps according to claim 8 does not follow in an obvious way from the available prior art.

- 3.6 Claims 2 to 4, 6 and 7, and 9 and 10, are dependent upon claims 1, 5 and 8 respectively. The inventive step of their subject-matter follows from this dependency.

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 claims 1 to 10 submitted during the oral proceedings and the description to be adapted, figures as granted.

The Registrar:

The Chairman:

U. Bultmann

R. Spangenberg