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**Datasheet for the decision
of 20 June 2007**

Case Number: T 0073/05 - 3.3.05

Application Number: 96119473.5

Publication Number: 0778069

IPC: B01D 53/32

Language of the proceedings: EN

Title of invention:

Reactive purge for solid electrolyte membrane gas separation

Patentee:

PRAXAIR TECHNOLOGY, INC.

Opponent:

AIR PRODUCTS AND CHEMICALS, INC.

Headword:

Reactive purge/PRAXAIR

Relevant legal provisions:

EPC Art. 54, 56

Keyword:

"Novelty - yes"
"Inventive step - yes"

Decisions cited:

-

Catchword:

-



Case Number: T 0073/05 - 3.3.05

D E C I S I O N
of the Technical Board of Appeal 3.3.05
of 20 June 2007

Appellant: AIR PRODUCTS AND CHEMICALS, INC.
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Respondent: PRAXAIR TECHNOLOGY, INC.
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Decision under appeal: Interlocutory decision of the Opposition
Division of the European Patent Office posted
6 December 2004 concerning maintenance of the
European Patent No. 0778069 in amended form.

Composition of the Board:

Chairman: M. Eberhard
Members: B. Czech
H. Preglau

Summary of Facts and Submissions

- I. The appeal is from the decision of the opposition division concerning the maintenance of European patent No. 0 778 069 in amended form.
- II. In the contested decision, the opposition division held that the amended patent comprising the claims according to the auxiliary request filed with letter dated 28 May 2003 met the requirements of the EPC. Independent claims 1 and 4 of this request read as follows (features amended during the opposition proceedings highlighted by the board):

"1. A process for producing a high-purity product from a feed stream containing elemental oxygen, comprising:

applying said feed stream to a first separator including a first feed zone and a first permeate zone separated by a first solid electrolyte membrane capable of transporting oxygen ions;

*driving a first portion of oxygen contained in said feed stream from said first feed zone to said first permeate zone through said first membrane by applying a reactive purge stream to said first permeate zone, said reactive purge stream including a reactive gas that **reacts** with oxygen to establish a lower partial pressure of oxygen in said first permeate zone; and*

obtaining an oxygen-depleted product stream after the first portion of oxygen has been driven from said first feed zone, wherein said oxygen-depleted product stream is a high-purity retentate stream which contains less than five percent by volume of elemental oxygen and wherein said reactive purge

stream includes a portion of output from said first feed zone".

"4. A process for producing a high-purity product from a feed stream containing elemental oxygen, comprising:

applying said feed stream to a first separator including a first feed zone and a first permeate zone separated by a first solid electrolyte membrane capable of transporting oxygen ions;

driving a first portion of oxygen contained in said feed stream from said first feed zone to said first permeate zone through said first membrane by applying a reactive purge stream to said first permeate zone, said reactive purge stream including a reactive gas that **reacts** with oxygen to establish a lower partial pressure of oxygen in said first permeate zone; and

obtaining an oxygen-depleted product stream after the first portion of oxygen has been driven from said first feed zone, wherein said oxygen-depleted product stream is a high-purity retentate stream which contains less than five percent by volume of elemental oxygen,

said process further including positioning said first separator as a second stage and

initially applying said feed stream to a second feed zone of at least a second separator, said second separator being positioned as a first stage and having a second permeate zone separated from said second feed zone by a second solid electrolyte membrane,

driving a second portion of oxygen, which is contained in said feed stream, from said second feed zone to said second permeate zone through said second membrane; and

directing an oxygen-depleted feed stream, obtained after the second portion of oxygen has been driven from said second feed zone, to said first feed zone of said first separator,

wherein the step of driving oxygen for at least one of said first and second separators includes diverting a portion of output from at least one of said first and second feed zones to purge the permeate zone of that separator".

III. The prior art cited by the opponent includes the following documents:

D1: US 5 160 173 A

D3: US 5 035 726 A

D5: US 5 205 842 A

D7: WO 94/24065 A1

IV. In its statement of grounds of appeal, the appellant (opponent) relied on these four documents and on two dictionary excerpts. More particularly, it argued that the subject-matter of claim 1 as upheld by the opposition division lacked novelty over D1. In case the subject-matter of claim 1 was to be considered as novel over D1, it was obvious in view of this document taken alone. Based on the view that the term "*portion*" as used in claim 1 could also be considered to designate a portion "*by ingredient*", it held that the process of claim 1 was also obvious in view of a combination of D1 with D3. Moreover, the process of claim 1 lacked an inventive step in view of combinations of D1 and D5, D7

and D5, and D7 and D3, the latter combination also rendering obvious the process of claim 4.

- V. With its reply, the respondent (proprietor of the patent) refuted the arguments of the appellant and filed two amended sets of claims as first and second auxiliary requests.
- VI. With a further letter the appellant filed three further dictionary excerpts.
- VII. During oral proceedings held on 20 June 2007, the respondent filed another set of claims 1 to 8 as new main request, replacing all the requests previously on file. The set of claims according to this new main request differs from the one underlying the contested decision only in that the term "**diverted**" is inserted into claim 1 between "*said reactive purge stream includes a*" and "*portion of output from said first feed zone*".
- VIII. The arguments of the parties concerning the sole remaining (main) request of the respondent can be summarised as follows:

At the oral proceedings the appellant did not raise objections concerning the allowability of the amendments made to the claims. In its view, D1 disclosed or at least suggested a process with all the features of claim 1 as allowed by the opposition division. More particularly, it held that the skilled person would understand that the passage of D1 concerning the composition of the reactant gas and mentioning the possibility to use a "recycled product

stream" (column 8, lines 5 to 13) applied to both of the purposes of the process described in D1, i.e. to the extraction of oxygen from a feed gas to obtain an oxidised product (D1, column 3, lines 33 to 38) and to the removal or complete elimination of oxygen from a feed gas in order to purify the latter (D1, column 3, lines 53 to 62). In the first case, the reaction products leaving the permeate zone could be considered as "product" gas, whereas in the second case, the purified retentate gas could be considered as "product". This view was corroborated further by D7 (page 15, line 27 to page 16, line 31), wherein a similar process was described and wherein both the retentate and the permeate were designated as "product". D1 thus inter alia disclosed a process leading to the complete elimination of oxygen from a feed gas, i.e. to a high purity product with a residual concentration of less than 5 percent by volume, which process comprising flowing through the permeate zone of the membrane separator a reactant gas diluted with part of the recycled product gas, i.e. retentate. Even if this was not accepted, the use of a part of the retentate as "recycled product stream" for diluting the reactant gas was obvious since it was taught by D1 itself. The only combination of documents the appellant invoked at the oral proceedings against claim 1 of the new main request was the combination of D1 with D5. D5 related to a two-stage membrane gas separation. Starting from D1 and looking for an alternative source for the reactant purge gas fed to the permeate side, the skilled person would consider D5 and its teaching concerning the way the process streams were conducted. D5 disclosed the "well-known" recycling of retentate by means of a valve for lowering the partial pressure of

the permeating component on the permeate side. In this connection, the appellant emphasised that the inventor of D5 was also one of the inventors of the present patent. In view of the teaching of D5, the skilled person would thus obviously consider modifying the process of D1 by diverting a portion of the retentate and leading it back to the permeate side.

The respondent pointed out that D1 did not only disclose processes with a reactive purge, and that in the passage of D1 relating to the "complete elimination" of oxygen (column 3, lines 53 to 62), nothing was said about the use of a reactant gas. The passage concerning the composition of the reactant gas (column 8, lines 5 to 13) was thus not related to the former passage. The expression "**a** recycled product stream" (emphasis added by the board) appeared only once in D1 and did not say anything about the nature or origin of the said stream. On the other hand, the only reference to a "product" was to be found in the examples, where this term was used to designate the permeate side gas after reaction. The retentate was never designated as "product" in D1 itself, and the reference to other documents such as e.g. D7 was not permissible when assessing the disclosure of D1. D1 thus clearly and unambiguously disclosed recycling gas containing reaction products withdrawn from the permeate zone back to the permeate zone inlet as diluent, a measure also described in the patent in suit (see Figure 1, "exhaust recirculation stream" 48). The respondent argued that starting from D1 as the closest prior art, modifying the process disclosed therein by using a diverted portion of the retentate, rather than some external diluent, would result in an improved

control of the combustion reaction taking place on the permeate side, since in this manner the temperatures, pressures and compositions of the gases on both sides of the membrane were interrelated. This modification was neither suggested by D1 itself nor by D5 or the other prior art cited. D5 related to a process wherein a portion of the gas withdrawn from the retentate side was used to purge the permeate side of the membrane separator. This gas was inert and no combustion reaction took place near the membrane. Therefore, polymeric membranes could also be used according to D5, which membranes would not be suitable for carrying out the process of the invention. The "reflux" of retentate disclosed in D5 merely served the purpose of purging the permeate side, and not the purpose of controlling a combustion reaction. The skilled person would thus not, without relying on ex post facto considerations, combine D1 with D5, a document from a more remote field, to arrive at the claimed process in an obvious manner.

IX. The appellant requested that the decision under appeal be set aside and that the patent be revoked.

The respondent requested that the decision under appeal be set aside and that the patent be maintained according to the set of claims filed during the oral proceedings.

Reasons for the Decision

1. *Allowability of amended claim 1*

1.1 The allowability of the amendments to the claims as granted have been not been challenged by the appellant. The board also has no reason to question the positive finding of the opposition division having regard to the replacement of the term "*combines*" by the term "***reacts***" in claims 1, 4 and 5. The further amendment to claim 1 consisting in the insertion of the term "***diverted***" into claim 1 to specify that the "*reactive purge stream includes a **diverted** portion of output from said first feed zone*" finds a basis in the application as originally filed inter alia on page 8, lines 25 to 27 referring to Figure 1 and on page 16, lines 2 to 4 referring to Figure 3. Both figures show embodiments with one separator and a conduit comprising a valve which permits diverting some of the retentate gas leaving the separator and joining the diverted portion to the purge gas stream entering the permeate side of the separator.

1.2 The board is thus satisfied that this amendment meets the requirements of Article 123(2) EPC and, in view of its restricting nature, of Article 123(3) EPC.

2. *Novelty*

2.1 Document D1 discloses processes for the separation of oxygen from an oxygen-containing gas such as air by an oxide ion conducting mixed metal oxide membrane. Two distinct types of processes are generally described in D1: The oxygen extracted by means of the membrane is

removed from the second surface of the membrane either i) by reaction with an oxygen-consuming substance such as a hydrocarbon gas or ii) as molecular oxygen, see e.g. column 2, lines 6 to 52; column 5, lines 54 to 60, figures 1 and 3.

- 2.1.1 Concerning the first type of process, it is indicated in column 3, lines 33 to 39 that the feed gas should contain at least about 0.1 mole percent of molecular oxygen, more preferably at least about 5 mole percent, and that air is particularly preferred. The reduction of the oxygen concentration occurring in the feed gas, i.e. on the retentate side, is neither addressed in this passage nor in the examples of D1, which also relate to the first type of process. On the other hand, in the only passage of D1 explicitly referring to complete elimination of the oxygen from the feed gas (column 3, lines 53 to 59), no mention is made of the use of a reacting gas. The skilled person thus has no reason to read the latter passage in conjunction with the passage in column 8, lines 5 to 13 of D1, where more details are given concerning the case wherein an oxygen consuming substrate, typically a reactant gas, is used.
- 2.1.2 Exemplary reaction gases are stated to include hydrogen, carbon monoxide and hydrocarbons. Moreover, it is said that "the reaction gas can also contain inerts or diluents, such as nitrogen or steam, or **a recycled product stream**" (emphasis added by the board). However, it is not explicitly indicated in D1 what is meant by "recycled product". In particular, the output on the retentate side is nowhere in D1 designated as "product". The only other occurrences in D1 of the term "product"

are in examples 1 (column 9, lines 22, 67 and 68) and 2 (column 10, lines 22 to 28). In these examples, it is undisputedly the gas stream leaving the permeate side of the membrane separator which is called "**product** gas flow" (emphasis added by the board). Moreover, recycling of some of this gas flow together with the reactant gas entering the permeate zone makes sense technically (see e.g. the contested patent, page 4, lines 21 to 23 and lines 32 to 33). The fact that in D7, a document also relating to the membrane process for separating oxygen from air (page 15, line 27 to page 16 line 31), both the permeate and the retentate are designated as "product" cannot alter the disclosure of D1 and does not permit considering that the term "product" as used in column 8, line 13 of D1 could designate the retentate.

2.1.3 As acknowledged by the appellant during the oral proceedings, the term "*diverted*" inserted into claim 1 implies that there must be valve and conduit means or similar means leading a volume portion of the retentate stream leaving the retentate side of the separator to the permeate zone inlet. However, such means are not disclosed in D1.

2.2 The board concludes that a process with the combined features of present independent claims 1 or 4, i.e. wherein the oxygen content of a feed gas is reduced by membrane separation to a concentration of less than 5 % by volume, and which also comprises the use of a reactive purge gas containing a diverted portion of output from the feed zone of the separator, is not directly and unambiguously derivable from D1.

2.3 The processes according to independent claims 1 or 4 are also not disclosed in any of the other documents cited by the appellant. As this was not disputed, further consideration in this respect is not necessary.

2.4 The subject-matter of claims 1 to 8 is thus novel.

3. *Inventive step*

3.1 Closest prior art

Considering the similarities between the processes disclosed in D1 and the process of present claim 1, the board can accept that D1 represents the closest prior art.

3.2 Technical Problem

3.2.1 In the board's view, the technical problem formulated by the appellant during the oral proceedings, i.e. "to look for an alternative purge stream" contains a pointer to the solution and cannot, therefore be accepted. On the other hand, there is no evidence on file corroborating any improvement over the process of D1. Hence the board doubts whether the problem stated on page 8 (first paragraph) of the contested decision ("improved control of the combustion on the permeate side of an oxygen transferring solid electrolyte membrane"), upon which the respondent relied during the oral proceedings, is actually solved by the claimed process. The technical problems relied upon by the parties can thus not be accepted in accordance with constant jurisprudence of the boards of appeal (see e.g.

"Case Law of the Boards of Appeal of the EPO", 5th edition 2006, sections I.D.4.2 and I.D.4.3.1).

3.2.2 The technical problem can however be seen in providing a further way of controlling a process wherein oxygen is removed from a gas mixture in a separator comprising an oxygen ion conducting solid electrolyte membrane.

3.2.3 It is immediately apparent from the patent in suit and it has not been disputed that this technical problem is solved by the process of present claims 1 and 4.

3.3 Non-obviousness

3.3.1 Document D1 does not suggest the use of a diverted part of the retentate output as diluting component of the reactant gas in a process leading to a high purity retentate.

3.3.2 The combination of D1 with D5

D5 discloses a process for drying wet gas streams in a two stage membrane system. The "wet component" of the feed gas permeates through a membrane in a first permeator and is thereby partially separated from the feed gas. The partially dried retentate gas from the first permeator is fed to a second permeator. In this second permeator, further "wet component" permeates through a further membrane and the permeated wet component is purged from the lower pressure permeate side of this permeator by a "reflux purge". The "reflux purge" consists in diverting a portion of the non-permeated, dried product gas stream leaving the retentate side of the second permeator by means of a

valve and re-introducing it into said permeator on its permeate side. Reference is made in particular to claim 1, Figure 1, column 4, lines 10 to 17 and column 6, lines 3 to 41.

D5 is specifically concerned with a membrane process for drying a wet feed gas by separating the "wet component", i.e. easily condensable components such as water, from the gaseous mix. In such a process, there is a risk of water condensing on the permeate side of the membrane, see D5, column 1, lines 42 to 57. On the other hand, D5 neither relates to processes for separating other types of gas components, such as oxygen, let alone using ion-transporting solid electrolyte membranes, nor to processes operated at particularly high temperatures. Moreover, the reflux purge stream of D5 consists only of dried retentate. It is thus not used to dilute a reactive purge gas component, and hence does not have an influence on a reaction of a permeated component with a reactant on the permeate side of the membrane. In view of these differences the board is not convinced that a skilled person starting from D1 would be induced by D5 to adopt a particular feature of the process of D5, such as the recycling of a part of the retentate, whilst keeping the reactive purge of D1, in order to solve the stated technical problem. D5 being a patent publication, its disclosure, and in particular its teaching concerning the "reflux purge", cannot, in the absence of further corroborating evidence, be considered to belong to common general knowledge in the field of membrane separation. The fact that the inventor indicated in D5 is also one of the inventors stated in the patent in suit does not imply that the notional person skilled in

the art would obviously combine the teachings of D1 and D5 in a manner leading him to the process according to present claims 1 or 4.

3.3.3 At the oral proceedings, the appellant has not invoked any other combination of the documents cited in the appeal and opposition proceedings to substantiate its objection concerning the alleged lack of inventive step of the amended claims according to the sole remaining request. The board is also convinced that the remaining documents contain no additional relevant information which would point towards the claimed subject-matter.

D3 relates to a process for purifying crude argon gas recovered from a cryogenic, adsorptive or membrane separation of air. Oxygen is removed from the argon gas by selective permeation of oxygen through a high temperature solid electrolyte membrane in one or two stages. D3 discloses the use of a suitable sweep gas such as nitrogen to lower the oxygen partial pressure on the permeate side. In one embodiment, some of the nitrogen contained in the sweep gas stems from the downstream separation by distillation of the substantially oxygen-free argon stream obtained as retentate in the membrane separation. This nitrogen containing argon could thus be considered as "a portion by ingredient" of the membrane separator's output. In D3 it is neither envisaged to use of a reactive sweep gas, nor is it suggested to divert a portion of the output, i.e. to use a portion by volume as a component of the sweep gas. Reference is made in particular to D3, column 2, lines 25 to 51; column 3, lines 42 to 55; column 3, line 62 to column 4, line 9; , claims 1 and 3; figures 1 and 2.

D7 also discloses the separation of oxygen from a gas mixture such as air by means of an oxygen ion-conducting ceramic membrane. According to a preferred embodiment, the oxygen transported across the membrane is reacted on the permeate side with a gas stream containing at least one organic compound such as light hydrocarbon gases, optionally diluted with inert gases, preferably nitrogen or steam. Diluting the hydrocarbon stream with a diverted portion of oxygen depleted retentate is, however, not suggested by D7. Reference is made in particular to page 9, lines 6 to 10; page 15, line 27 to page 17, line 17; page 29, line 23 to page 31, line 6.

3.3.4 Since the subject-matter of claims 1 and 4 and, consequently, of dependent claims 2, 3, and 5 to 8, is not obvious having regard to the prior art cited by the appellant, it is also based on an inventive step.

4. The claims as amended during the oral proceedings are consistent with the amended description on file. This was not disputed by the appellant.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the first instance with the order to maintain the patent with the following documents:

Claims 1 to 8 filed during the oral proceedings

Description:

Pages 2,5,7,8	of the patent specification
Page 3	filed with letter of 27 May 2003
Pages 4, 6	filed during the oral proceedings of 23 November 2004

Drawings: Figures 1 to 5 of the patent specification

The registrar

The chairman

C. Vodz

M. Eberhard