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D E C I S I O N
of 21 February 2002

Case Number: T 0947/98 - 3.3.5

Application Number: 91300307.5

Publication Number: 0438282

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Language of the proceedings: EN

Title of invention:
Production of pure nitrogen from air

Patentee:
THE BOC GROUP, INC.

Opponent:
L'AIR LIQUIDE, S.A. pour l'étude et l'exploitation des
procédés Georges Claude

Headword:
-

Relevant legal provisions:
EPC Art. 56, 100(a)

Keyword:
"Inventive step (no); obvious combination of features"

Decisions cited:
-

Catchword:
-



Case Number: T 0947/98 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 21 February 2002

Appellant: THE BOC GROUP, INC.
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 5 August 1998
revoking European patent No. 0 438 282 pursuant
to Article 102(1) EPC.

Composition of the Board:

Chairman: R. K. Spangenberg
Members: A. T. Liu
J. H. Van Moer

Summary of Facts and Submissions

I. European patent No. 0 438 282 was granted with a set of claims consisting of an independent claim 1 for a process and claims 2 to 14 depending thereon. Claim 1 read as follows:

"A process for separating air comprising

(i) purifying a gaseous feed air stream by substantially freeing it of water vapour, carbon monoxide and carbon dioxide impurities by a method comprising the steps of:

(a) removing water vapour from the gaseous feed air stream;

(b) contacting the feed stream from step (a) with one or more oxidation catalysts thereby to convert carbon monoxide to carbon dioxide; and

(c) removing carbon dioxide and, if present, water vapour from the gaseous stream obtained from step (b) to obtain the purified air; and

(ii) distilling the purified air to produce nitrogen".

II. A notice of opposition was filed against the patent on the grounds of Article 100(a) EPC. During the opposition proceedings, six prior art documents were cited, of which reference shall be made to the following four in the present decision:

D1: JP-A-61-228 286 (English translation)

D2: US-A-3 672 824

D3: US-A-4 579 723

D5: US-A-4 054 428

- III. In the course of the opposition proceedings, four sets of amended claims were filed as auxiliary requests by the patent proprietor.
- IV. At the end of the oral proceedings which were held on 24 July 1998, the opposition division came to the conclusion that, essentially, the subject-matter of claim 1 as granted and that of claim 1 according to the auxiliary requests did not involve an inventive step with respect to the closest prior art D1.
- V. The appeal was from the decision of the Opposition Division revoking the European patent.
- VI. By letter of 18 January 2002, the appellant submitted six amended sets of claims termed first to sixth auxiliary requests. The correspondingly amended pages of description were submitted by letter of 21 January 2002.
- VII. The first, fourth and fifth auxiliary requests were withdrawn at the oral proceedings held on 21 February 2002. Claim 1 of the remaining auxiliary requests were as follows.

Claim 1 of the second auxiliary request differed from claim 1 as granted in that it further incorporated the stipulation:

"... in which the said steps (a) to (c) are performed by passing the feed gas through a single treatment zone contained in a single vessel comprising a first section of adsorbent for performing step (a), a second section of oxidation catalyst for performing step (b), and a third section of adsorbent for performing step (c)".

Claim 1 of the third auxiliary request differed from claim 1 of the second auxiliary request in that it further incorporated the stipulation:

"... and in which the treatment zone containing the two adsorption sections and the catalyst section is periodically regenerated by purging the accumulated adsorbed impurities".

Claim 1 of the sixth auxiliary request read as follows:

"A process for separating air comprising:

- (i) compressing a gaseous feed air stream, cooling the compressed feed air stream in a heat exchanger and removing liquid water from the cooled air stream, so as to form a resulting feed air stream;
- (ii) purifying the resulting feed air stream by substantially freeing it of water vapour, carbon monoxide and carbon dioxide impurities by a method comprising the steps of:
 - (a) removing water vapour from the gaseous feed air stream;

(b) contacting the feed stream from step (a) with one or more oxidation catalysts thereby to convert carbon monoxide to carbon dioxide; and

(c) removing carbon dioxide and, if present, water vapour from the gaseous stream obtained from step (b) to obtain the purified air; and

(iii) distilling the purified air to produce nitrogen;

in which the said steps (a) to (c) are performed by passing the feed gas through a single treatment zone contained in a vessel comprising a first section of adsorbent for performing step (a), a second section of oxidation catalyst for performing step (b), and a third section of adsorbent for performing step (c), in which the treatment zone containing the two adsorption sections and the catalyst section is periodically regenerated by purging the accumulated adsorbed impurities, and in which the treatment zone is operated in a pressure swing mode or a temperature swing mode, the said resulting feed air stream being formed at a temperature of from 5 to 50 °C when the treatment zone is operated in the pressure swing mode, or at a temperature of from 5 to 20 °C when the treatment zone is operated in the temperature swing mode."

VIII. The appellant's arguments, submitted orally and in writing, may be summarised as follows:

- With regard to the closest prior art represented by D1, the problem to be solved was to reduce the cost in the production of highly pure nitrogen.
- In D1, the catalyst had the dual function of

oxidizing both CO and H₂. The catalyst used was based on palladium which was only efficient at elevated temperatures and not known to be sensitive to water.

- The catalyst used in the patent in suit was different from the catalyst of D1 since it was only needed for the oxidation of CO.
- The temperature of air fed to the purification zone was not obvious in view of D1.
- The teachings of D1 and D2 (or D5) were mutually contradictory.
- D2 and D5 concerned a technical field different from that of D1 and the patent in suit, namely the purification of room air. In these processes, the mixed oxides catalysts were applied to the removal of excess and not traces of CO from room air. A combination of D1 with either D2 or D5 would be based on hindsight.
- The use of the single vessel for the air purification, which further reduced the operational costs, was not obvious.

IX. The respondent's arguments were briefly as follows:

- It was obvious for the skilled person to try and apply the catalyst of D2 or D5 as an alternative to the palladium catalyst in the process of D1.
- The use of a desiccant at ambient temperatures upstream of the catalyst was also known from D2

and D5.

- D2 was relevant since the level of CO in purified air as given in D2 corresponded to the level of detection achievable at that time.
- The use of single vessels as claimed was common in the art.
- The stipulated temperature ranges of feed air were known from D2.

X. The appellant (patentee) requested that the decision under appeal be set aside and that the patent be maintained as granted or in the alternative on the basis of the claims submitted with letter of 18 January 2002 as second, third and sixth auxiliary requests.

The respondent (opponent) requested that the appeal be dismissed.

Reasons for the Decision

1. *Main request*

1.1 The patent in suit is directed to a process for separating air to produce nitrogen which is free of water vapour, carbon dioxide and carbon monoxide (page 2, lines 3 and 4). It concerns in particular an "upfront separation process of air" in which these impurities are first removed from a gaseous feed air stream and the purified air subsequently separated to produce high purity nitrogen for use in the semiconductor industry (page 2, lines 5 to 8, lines 34

to 48 and claim 1).

- 1.2 It is common ground that D1 is considered to comprise the closest prior art.

D1 discloses a conventional "upfront separation process of air" in which feed air is compressed and its moisture removed, the compressed and dried air cooled, any water vapour and CO₂ contained in the cooled compressed air removed by adsorption and the purified air sent to a distillation column for producing nitrogen (page 4, last paragraph to page 6, first paragraph and Figure 8).

It is further known from D1 that the presence of carbon monoxide and hydrogen even at very low concentrations in the feed air is detrimental to the application of the final product in the semiconductor manufacturing industry. It is therefore proposed to pass the compressed air directly over a palladium catalyst to oxidise these impurities into carbon dioxide and water, respectively, before further processing (page 6, last paragraph to page 7, last full paragraph; page 11, second paragraph and Figure 1).

- 1.3 The Board accepts the appellant's submission that the problem to be solved with regard to D1 can be seen in the reduction of operational costs. As is already indicated in the patent in suit, these costs may arise due to the extensive use of noble metal catalysts and the requirements in terms of equipment and energy (page 2, lines 18 to 24).

- 1.4 The solution proposed in claim 1 differs from the processes disclosed in D1, be it the conventional

process illustrated in Figure 8 or the proprietary process illustrated in Figure 1, in the steps of:

- (a) removing water vapour from the gaseous feed air stream and
- (b) contacting the feed stream from step (a) with one or more oxidation catalysts thereby to convert carbon monoxide to carbon dioxide.

According to the appellant, the stipulation in step (b) that the oxidation catalyst is "to convert carbon monoxide to carbon dioxide" has to be interpreted as a functional feature. The oxidation catalyst implied by this feature is thus necessarily different from the palladium catalyst of D1 which has to fulfil the dual function of oxidising both carbon monoxide and hydrogen. The Board can accept this explanation which is corroborated by the description. Indeed, in contrast to the process illustrated in Figure 1 of D1, the present process primarily seeks to remove carbon monoxide and the catalytic system to be applied is not expected to oxidize hydrogen (point VIII above and description, page 2, line 57 to page 3, line 6).

- 1.5 As is indicated above, there is in the present case no need for a catalyst with a dual function. Furthermore, there is no doubt that the use of noble metal catalysts entails high costs of operation. Thus, when seeking to reduce the costs, the first and most obvious modification the skilled person would consider is to replace the palladium catalyst with one which is selective for the oxidation of carbon monoxide to carbon dioxide but less expensive. Such a catalyst based on manganese and copper oxides (generally known

as hopcalite) is well known in the art (see for example D2, column 1, lines 35 to 44 and D5, column 1, lines 14 to 16). The Board therefore holds that it is straightforward for the skilled person to apply these mixed oxides to the present process as an alternative to the palladium catalyst of D1. The selection of these mixed oxides as the catalyst for the present process is thus not an indication of an inventive step (see Examples I-III).

The skilled person also knows that this oxidation catalyst is sensitive to water or moisture in any form and therefore needs protection from poisoning by moisture (see D2, column 1, lines 48 to 56 and D5, column 1, lines 16 to 20). In the Board's judgment, therefore, the insertion of a step for removing any water vapour from feed air prior to its contacting with the mixed oxides catalyst is an obvious consequence of the choice of the catalyst.

- 1.6 The appellant has asserted that D2 is directed to the purification of room air and the excess carbon monoxide is only removed to the extent that its content in the discharged air is reduced to 20 ppm. The skilled person would therefore not expect the same oxidation catalyst to be suitable for the present purpose of removing traces of carbon monoxide.

The appellant has, however, not provided convincing arguments, let alone proof, to refute the respondent's submission that the level of 20 ppm indicated in D2 merely reflects the threshold of detection which could be reached before 1972, the year in which D2 was published. The Board therefore holds that, knowing the analytical constraints in earlier years, the skilled

person would not be deterred by the relatively high level of residual carbon monoxide indicated in D2 from taking this piece of prior art into consideration with a view to solving the present technical problem. In contrast, he would rather interpret the data in D2 as demonstrating a high efficiency - in absolute as well as in relative terms - of the mixed oxides catalyst for oxidizing carbon monoxide. In the Board's judgment, D2 therefore provides a strong incentive for the skilled person to try and apply the same catalytic system to the purpose envisaged by the patent in suit. Under these circumstances, the observed fact that the mixed oxides oxidation catalyst when applied under the same operational conditions as in D2, is indeed efficient in removing traces of carbon monoxide, is no more than the result of an obvious routine experiment.

- 1.7 The appellant has also advanced the argument that the teachings of D1 on the one hand, and D2 and D5 on the other hand, are mutually contradictory. The skilled person does not have any incentive in substituting the palladium catalyst of D1 which is efficient at elevated temperatures with a catalyst according to D2 (or D5) working at ambient temperatures. This would go against the teaching of D1 whose salient feature is to make the best use of the heat of compression by feeding air at a high temperature to the palladium catalyst. It would be nonsensical to operate that process with a desiccant directly in front of the catalyst since the desiccant would not be effective at that temperature. Thus, according to the appellant, the only way in which D1 could be modified in view of D2 or D5 would be to compress the air, cool it, remove the water by adsorption with the desiccant, reheat the air to the operating temperature of the palladium catalyst and

cool the air again to ambient temperature for the removal of CO (see also letter of 14 December 1998, page 5, points 26 to 29).

- 1.7.1 The Board remarks that the temperature at which the oxidation catalyst is applied in step (b) is not explicitly stipulated in claim 1. However, if the stipulation "to convert carbon monoxide to carbon dioxide" is to be interpreted as a functional feature, then the Board can also accept said feature as implying that the catalyst is employed at such temperatures where it is efficient for the conversion in question.
- 1.7.2 As is indicated in point 1.2 above, in a conventional "upfront separation process of air", for example as illustrated in Figure 8 of D1, feed air is compressed, cooled and condensed water removed therefrom for further treatment. It is true that when the palladium catalyst is required for the oxidation of hydrogen and carbon monoxide, the hot compressed air is first contacted with the catalyst section before it is cooled and the condensed water removed therefrom (see D1, Figure 1 and point 1.2 above). As is not disputed by the appellant, the palladium catalyst is, on the one hand, known to be efficient at higher temperatures, and on the other hand, not known to be sensitive to moisture. In the Board's judgment, the skilled person would however not apply a particular teaching which is coupled with the use of the palladium catalyst to a process in which mixed oxides are used as catalysts, which are known to be efficient at ambient temperatures but sensitive to water vapour or moisture (see D2, Example 1; column 4, lines 3 to 7 and point 1.5 above). When modifying a conventional process such as one according to Figure 8 of D1, he would first, in the

knowledge of D2 (or D5), aim at inserting the mixed oxides catalyst appropriately so as to protect it from water poisoning.

1.7.3 Thus, the Board concurs with the appellant in that a combination of the D1 with D2 or D5 would lead the skilled person to compress the air, cool it to the temperature of the desiccant and remove the water before contacting air with the catalyst. However, since no palladium catalyst is involved here, there is no reason to reheat the air to the operating temperature of such catalyst. Therefore, the skilled person would in the present case, after passing air through the desiccant, directly contact the dried air which is already at the appropriate temperature with the mixed oxides catalyst for removing CO. The application of the mixed oxides catalyst at ambient temperatures directly after the water vapour removal thus arises straight from the choice of the catalyst.

1.8 The appellant has further observed that the inventors of the process of D1 (published in 1986) have chosen to use a palladium catalyst for the oxidation step. In his opinion, it is thus pure hindsight to revert to the older teachings according to D2 or D5, published in 1972 and 1975, respectively, when seeking to improve the process of D1.

The Board cannot accept this line of argument since the problem which D1 particularly sets out to solve is to simultaneously remove from the air any minute trace of hydrogen and carbon monoxide (see paragraph bridging pages 6 and 7 and point 1.2 above). Compared to D1, the problem to be solved by the present process is less severe in the sense that only carbon monoxide has to be

removed from the feed whilst the removal of hydrogen is optional and necessitates an additional catalyst (see patent in suit, page 2, line 59 to page 3, line 6). Moreover, the mixed oxides catalysts utilised in the examples of the patent in suit were not only known up until 1975 to be efficient for the desired purpose but were still readily available as commercial products at the priority date of the patent in suit (see patent in suit page 6, line 5 and line 32 to 33). In the Board's judgment, the application of these commercial products to the oxidation of carbon monoxide is therefore straightforward and not based on hindsight.

1.9 As corollary to the above, the Board holds that the subject-matter of claim 1 is an alternative process to that according to D1, Figure 1 which is obvious in the light of the disclosure of either D2 or D5.

2. *Auxiliary requests*

2.1 The additional key aspect common to the subject-matter of claim 1 of the auxiliary requests is the stipulation that the three steps of air purification (adsorption / oxidation / adsorption) are performed "by passing the feed gas through a single treatment zone contained in a single vessel" (see point VII above).

2.1.1 The Board recognises that, by carrying out the air purification in a single vessel, the process is simplified as compared to the process of D1. It is therefore accepted that the operational costs can be further reduced following a decreased requirement in

terms of equipment and energy (see also point 1.3 above). The Board, however, holds that the use of a single vessel for performing the three steps as stipulated is common in 21 March 2002 the art.

2.1.2 For example, in the process of D3, a gas stream comprising carbon monoxide is supplied to a bed of catalytic material wherein carbon monoxide is oxidised to carbon dioxide. The resulting gas stream is supplied to a second bed of getter material effective to trap carbon dioxide. Furthermore, it is explicitly suggested to add alumina at the inlet of the catalyst material in the event that relatively moist feed is to be purified. Alumina can also be added to the getter material in the event that it is desirable to balance a carbon dioxide breakthrough (column 6, lines 39 to 42 and 45 to 47). D3 does not use the term "adsorbent" for the materials used in the beds before and after the catalyst bed. However, it is clear that the alumina will perform the same function as in the patent in suit, namely first to remove water vapour from the feed gas and last, together with the getter, to remove carbon dioxide from the gas after the oxidation (compare patent in suit, page 3, lines 43 to 46 and page 4, lines 1 to 2). It is further explicitly stated in D3 that the beds may be disposed in a single container (column 2, lines 37 to 57, column 3, line 8, column 6, lines 3 to 6). Since D3 not only discloses the same essential gas purification steps in the same sequence as in claim 1 but also suggests performing these steps in a single vessel, the use of a single vessel in the present process cannot be regarded as involving an inventive step.

The Board does not concur with the appellant that D3 is

of no relevance to the patent in suit. In fact, D3 concerns the same technical field as D1 and the patent in suit since it is also directed to a method for producing pure nitrogen for use in the semiconductor industry (column 1, lines 5 to 20). The essential difference between D3 on the one hand and D1 and the patent in suit on the other hand is that the first discloses a "back end separation process of air" wherein the air is first separated into its components and the impurities are subsequently removed from the nitrogen gas whilst the latter two processes are "upfront separation processes of air" in which the purification takes place before the air separation (see also points 1.1 and 1.2 above). However, the appellant has not argued and the Board cannot see in which way this basic difference should have any bearing on the consideration as to whether or not to combine the purification steps into one vessel.

2.2 A further additional feature common to claim 1 of the third and sixth auxiliary requests is the stipulation that "the treatment zone containing the two adsorption sections and the catalyst section is periodically regenerated by purging the accumulated adsorbed impurities". This is however already known from D3 which teaches that, in order to assure continued effectiveness of the beds, these are regenerated by purging to remove trapped impurities (column 2, lines 63 to 58).

2.3 Claim 1 according to the sixth auxiliary request further stipulates prior to the step of purification:

- (i) "compressing a gaseous feed air stream, cooling the compressed feed air stream in a heat exchanger

and removing liquid water from the cooled air stream, so as to form a resulting feed air stream", and

(ii) operating the treatment zone in a pressure swing mode or a temperature swing mode, with "the said resulting feed air stream being formed at a temperature of from 5 to 50 °C when the treatment zone is operated in the pressure swing mode, or at a temperature of from 5 to 20 °C when the treatment zone is operated in the temperature swing mode."

2.3.1 Re. feature (i)

It is undisputed that in conventional processes, feed air is also compressed, cooled and condensed water removed therefrom for further treatment (see point 1.2 above). Thus the presence as such of feature (i) in the present process is common in the art.

Additionally, the mixed oxides catalyst used in the present process for oxidising carbon monoxide particularly requires protection from poisoning by moisture. As is already set out above, the positioning of feature (i) into the present process before the air purification treatment is merely an obvious consequence of the catalyst requirement (see point 1.7, including point 1.7.3).

2.3.2 Re. feature (ii)

It is irrefutable that the operation of the treatment zone in a pressure swing mode or a temperature swing mode as such does not relate to the technical problem

to be solved, namely the reduction of operational costs. Furthermore, as is already acknowledged in the patent in suit (page 2, lines 14 to 16), such mode of operation is common in the art (see also D1, page 2 and Figure 8). In addition, it is already known, for example from D2, that, where the mixed oxides catalyst is used, the air to be treated is introduced at 20 °C (column 4, lines 3 to 7 and preceding paragraph). The choice of the temperature ranges for both modes is therefore also obvious in view of the choice of the catalyst type (see also point 1.7, including point 1.7.3).

2.4 Combination of features

As summary of the above, the Board holds that the additional features of the auxiliary requests are either common in the art or a direct consequence of the choice of the mixed oxides as catalyst, which choice results from the kind of impurity to be removed, here carbon monoxide in feed air. The appellant has not submitted any arguments showing an unexpected interaction of these additional features either among themselves or between any of those and that of claim 1 of the main request. Consequently, the combination of features as claimed in any of the auxiliary requests does not imply an inventive step.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

U. Bultmann

R. Spangenberg