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**Datasheet for the decision
of 13 June 2008**

Case Number: T 0815/04 - 3.3.05

Application Number: 99309673.4

Publication Number: 1013333

IPC: B01D 53/86

Language of the proceedings: EN

Title of invention:

Process for the removal of carbon monoxide, hydrogen, carbon dioxide and water from a gas

Applicant:

AIR PRODUCTS AND CHEMICALS, INC.

Headword:

Air purification

Relevant legal provisions:

EPC Art. 52(1), 56

Relevant legal provisions (EPC 1973):

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Keyword:

"Inventive step (no): Formulation of the technical problem based on an alleged disadvantage of the closest prior art: not accepted (Reasons 3.1 to 3.4); obvious further process"

Decisions cited:

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Catchword:

-



Case Number: T 0815/04 - 3.3.05

D E C I S I O N
of the Technical Board of Appeal 3.3.05
of 13 June 2008

Appellant: AIR PRODUCTS AND CHEMICALS, INC.
7201 Hamilton Boulevard
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Representative: Smart, Peter John
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 18 February 2004
refusing European application No. 99309673.4
pursuant to Article 97(1) EPC 1973.

Composition of the Board:

Chairman: G. Raths
Members: B. Czech
C. Vallet

Summary of Facts and Submissions

- I. The appeal is from the decision of the examining division to refuse European patent application No. 99309673.4.
- II. The examining division inter alia cited the following prior art documents:
- D1: JP 2-307 506 A
D2: FR 2 739 304 A
D3: EP 0 438 282 A
- The application was refused on the ground that, taking into account common general knowledge, the process of claim 1 (not amended during examination), lacked an inventive step "over the combined teaching in D2 and D3".
- III. In its statement of grounds of appeal, the appellant requested that a patent be granted on the basis of the claims underlying the contested decision. Alternatively, it requested oral proceedings. The appellant argued that the conclusions of the examining division were based on unsubstantiated allegations concerning common general knowledge and on a hindsight combination of D2 and D3.
- IV. In the annex to the summons to oral proceedings, the board inter alia expressed a negative preliminary opinion concerning inventive step of the claimed subject-matter over the combination of D2 with D3. The board also pointed out that dependent claim 9 and the description suggested that all of the carbon dioxide

may already be removed in the first adsorption step of the claimed process.

- V. With its letter of 24 May 2008, the appellant filed a new main request consisting of a set of amended claims 1 to 9 together with a description adapted to these claims. The appellant took the view that the objections raised by the board in the annex to the summons were overcome by the amended claims and description.

Independent claim 1 according to the said new main request reads as follows:

"1. A process for the removal of carbon monoxide, hydrogen, carbon dioxide and water from air, comprising compressing air and thereby heating the air by heat of compression, substantially no further heat being added to said air after compression, contacting the heated air with a catalyst to oxidise the carbon monoxide to carbon dioxide and part of the hydrogen to water using said heat of compression and so forming a carbon monoxide depleted air stream, contacting the carbon monoxide depleted air stream with a solid adsorbent to remove at least water therefrom to produce a carbon monoxide and water depleted air stream, and contacting the carbon monoxide and water depleted air stream with a catalyst of platinum and palladium supported on alumina to oxidise remaining hydrogen to water and a zeolite adsorbent to adsorb water and carbon dioxide."

- VI. On 9 June 2008, the board issued a further communication by fax. The appellant's attention was inter alia drawn to the Derwent and PAJP abstracts of

D1, and to passages of D2 and D3 referring to the use of zeolite for adsorbing water and carbon dioxide.

VII. Oral proceedings took place on 13 June 2008. At the end of the oral proceedings the board announced its decision.

VIII. The essential arguments of the appellant with respect to the issue of inventive step can be summarised as follows:

The appellant argued that it had realised that with the method according to D2 there was a danger that in the final step some of the hydrogen would not be adsorbed but oxidised to water. This water would eventually be entrained in the purified air and disturb a subsequent cryogenic distillation. With the claimed process, the four impurities carbon monoxide, hydrogen, carbon dioxide and water were effectively removed and in particular all of the water generated in the hydrogen removing step was reliably removed from the air stream. The claimed process provided additional safety and also permitted to additionally remove carbon dioxide at the end of the process. Excluding hindsight considerations, the claimed solution was not suggested by the prior art. In particular, the skilled person would not consider combining the teachings of documents D2 and D3 since they were mutually incompatible and even "hostile". D2 acknowledged the teaching of older document D3 and considered it to be disadvantageous. Moving from D2 to the invention was thus clearly going against the teaching of D2, and there was no positive motivation for making the corresponding changes. First, this would amount to a reversion to a method specifically

condemned in D2. Secondly, the reader of the later document D2 would not expect that hydrogen oxidation would in reality occur as described in D3, since D2 taught specifically that under the conditions hydrogen was not oxidised to water at the relevant temperatures when using the very same materials which in D3 were said to produce oxidation. Furthermore, none of the cited prior art documents disclosed a catalyst containing palladium and platinum supported on alumina, let alone for hydrogen oxidation at ambient temperatures.

- IX. The appellant requested that the decision under appeal be set aside and that a patent be granted in the following version:

Claims 1 to 9 and description pages 1 to 12 filed as main request with letter of 27 May 2008 and figures 1 to 4 as originally filed.

Reasons for the Decision

Inventive step

1. The present application relates to processes for the removal of carbon monoxide, carbon dioxide, hydrogen and water from air for the production of high purity nitrogen gas, see page 1, first paragraph, of the application as filed.
2. D2 relates to a process for the purification of air wherein, like in the process according to present claim 1, the impurities carbon monoxide, hydrogen,

carbon dioxide and water are removed from an air stream by a combination of adsorption and catalytic oxidation steps. In view of the similarities between the process of D2 and the one according to present claim 1, the board shares the view of the examining division that D2 can be considered to represent the closest prior art. This was not contested by the appellant.

- 2.1 More particularly, the process disclosed in D2 comprises compressing air, contacting the compressed air at the elevated compressor outlet temperature, i.e. without additional heating, with a catalyst for the oxidation of carbon monoxide to carbon dioxide, cooling the air stream obtained to ambient temperature, contacting it with an adsorbent material for eliminating carbon dioxide and water by adsorption and finally contacting the stream with an adsorbent material trapping ("piégeant") under conditions not resulting in hydrogen oxidation. This adsorbent material, preferably supported palladium, is periodically regenerated, preferably using a sweep stream of oxygen enriched air at a temperature above ambient. The product stream obtained by the process of D2 is a dry and essentially pure air stream at ambient temperature, wherein the four impurities are reduced to a very low level. Suitable catalysts for oxidising carbon monoxide to carbon dioxide include platinum and palladium, which may optionally be supported. Preferred catalyst support materials include alumina. Preferred materials for adsorbing carbon dioxide and water include zeolites. Reference is in particular made to the following parts of D2: page 1, 1st paragraph, page 5, line 29 to page 6, line 26; page 6, line 27 to page 7, line 17; page 8, lines 14 to 25; claims 1, 5 to 7.

According to the examples (see page 14, line 17 to page 15, line 5) compressed air at 120°C is contacted with a Pd/Al₂O₃ catalyst for the oxidation of carbon monoxide. The air is then cooled and contacted with a Pd/Al₂O₃ adsorbent, which is periodically regenerated with a hot gas.

- 2.2 Some of the hydrogen present in the air is also oxidised upon contact with the carbon monoxide oxidation catalyst, although this is not expressly mentioned in D2. Considering that the oxidation takes place at the elevated compressor outlet temperature, the board is convinced that at least a small part of the hydrogen present will inevitably be oxidised under the conditions described when using supported palladium or platinum as the catalyst. This is confirmed by both the present application as filed (see claims 2 and 4 as originally filed) and D1 (see the PAJP abstract), according to which both supported platinum and palladium are suitable for oxidising carbon monoxide and at least a part of the hydrogen to water at the compressor outlet temperature. Moreover, the appellant acknowledged at the oral proceedings that there was no reason to believe that no hydrogen at all would be oxidised in this step of the process of D2.
3. The board notes that the application as filed contains no express indication of a particular technical problem underlying the invention.
- 3.1 The appellant argued that the method of D2 carried the risk that some of the hydrogen trapped by the adsorbing material, which was preferably selected from noble metals such as platinum and palladium, would be

catalytically oxidised to water rather than simply be adsorbed and trapped upon contact with such a material. The so-formed water would then be adsorbed by the adsorbent material, and would remain adsorbed even upon regeneration of the adsorbent (see D2, page 7, 2nd paragraph). Hence, at some stage, some of this water would be carried along by the stream of purified air produced, thereby leading to problems in a subsequent cryogenic distillation. Regeneration of the palladium adsorbent at higher temperatures would lead to a deterioration of the hydrogen adsorption capacity and to an increased oxidation of hydrogen to water. When using the method of D2, water was thus not reliably and completely removed from the air stream to be purified.

3.2 The appellant thus considered that starting from a process as disclosed in D2, the technical problem consisted in providing an effective and safer process, and that this problem was effectively solved by a process according to present claim 1, which entailed removing any residual hydrogen not by adsorption, but by contacting the stream with a catalyst for oxidising hydrogen to water, followed by adsorption of the water and of residual carbon dioxide. With the process of claim 1, as opposed to the one of D2, all of the residual hydrogen and all of the water produced by hydrogen oxidation was safely removed from the air stream, and carbon dioxide could additionally be removed at the end of the process.

3.3 The board however notes that it is expressly stated in D2 that using the regeneration methods described therein (page 7, second paragraph) the removal of hydrogen to the degree required e.g. for cryogenic

distillation (page 8, second and third paragraphs) can be safely achieved ("on peut garantir une teneur en hydrogène de l'ordre de quelques ppb") over repeated hydrogen adsorption and adsorbent regeneration cycles, and that water and carbon dioxide are also removed to the required extent (page 16, lines 21 to 25). The appellant did not provide any evidence to the contrary in order to corroborate its assertion concerning the alleged disadvantage of the process according to D2. Moreover, although D2 is mentioned as prior art in the present application as originally filed, a technical problem to be solved with respect to D2 is not expressly formulated therein. The risk allegedly associated with the hydrogen adsorption taught by D2 (see point 2.1 above) is not at all addressed in the application as filed.

3.4 The technical problem as formulated by the appellant is thus based on a disadvantage of the closest prior art which was only alleged after the filing date of the present application. Further, said technical problem cannot be deduced either from the application as filed, not even when considering the content of document D2 cited therein. In accordance with established case law (see e.g. Case Law of the Boards of Appeal of the EPO, 5th edition 2006, Section I.D.4.1, second paragraph and Section I.D.4.4, first paragraph) the appellant's formulation of the technical problem is thus not accepted.

3.5 In the light of document D2, the technical problem underlying the application as amended can however be seen in providing a further process for removing the four mentioned impurities from an air stream.

4. According to present claim 1, the solution to this technical problem is a process which inter alia comprises as final steps "*...contacting the carbon monoxide and water depleted air stream with a catalyst of platinum and palladium supported on alumina to oxidise remaining hydrogen to water and a zeolite adsorbent to adsorb water and carbon dioxide*".
5. The board is satisfied that the technical problem is indeed solved by this claimed solution.
6. It remains to be decided whether the claimed solution to the technical problem is obvious in view of the cited prior art.
7. Confronted with the stated technical problem, the skilled person would consider the prior art pertaining to the same technical field as D2, namely the purification of air comprising the said impurities. This prior art includes the processes disclosed in document D3, a document acknowledged in D2.
 - 7.1 Document D3 relates to the purification of gases, in particular of air to be subjected to cryogenic distillation, by removing therefrom the four impurities carbon monoxide, hydrogen, carbon dioxide and water. More particularly, D3 discloses a process comprising compressing atmospheric air, cooling the compressed air and subjecting it to an adsorption step to thereby remove therefrom water vapour and some of the carbon dioxide, contacting the feed stream obtained in the presence of oxygen with one or more oxidation catalysts thereby to convert carbon monoxide to carbon dioxide, and additionally with an oxidation catalyst to convert

hydrogen to water vapour, removing carbon dioxide and water vapour from the gaseous stream obtained. The purified gas stream obtained is substantially free of water vapour, water, carbon monoxide and carbon dioxide. The oxidation catalyst for converting hydrogen to water vapour is preferably "supported palladium or another noble metal catalyst known in the art". Zeolites are mentioned as adsorbents that may preferably be used for removing carbon dioxide and water include. Reference is made in particular to D3, claims 1 to 3, 5 and 6; page 4, line 41 to page 5, line 17; and Figure 1. Example II of D3 describes a process wherein water saturated feed air comprising carbon dioxide and some added carbon monoxide and hydrogen (2 ppm) at a temperature of 25°C and at an elevated pressure is fed to a vessel comprising a series of superposed layers, namely an initial layer of activated alumina, a second layer of "Hopcalite" oxidation catalyst, a third layer of a Pd/Alumina catalyst and a final layer of activated alumina. The treated air contains no hydrogen, no carbon monoxide, less than 0.1 ppm water and less than 1 ppm carbon dioxide.

- 7.2 For the skilled person, it is readily apparent from D3 that hydrogen present in a air stream to be purified to the degree required by a subsequent cryogenic distillation, which air stream is initially compressed and cooled to ambient temperatures, may be removed effectively from the said air stream subsequently to a water and carbon dioxide adsorption step and the removal of carbon monoxide by catalytic oxidation, by oxidising the hydrogen to water vapour using palladium or another noble metal supported on e.g. alumina as the oxidation catalyst, followed by the adsorption of the

water vapour and carbon dioxide present in the stream on a suitable adsorbent, such as a zeolite.

8. Therefore, the skilled person would envisage the replacement of the final hydrogen adsorption step of the process of D2 by the hydrogen oxidation followed by water and carbon dioxide adsorption as disclosed in D3 as a possible solution to the stated technical problem.

8.1 Zeolite adsorbents are a preferred material amongst others for adsorbing water vapour and carbon dioxide according to both D2 (see e.g. page 5, lines 15 to 19, and claim 7) and D3 (see e.g. page 4, lines 52 to 54; page 5, lines 11 to 13). The skilled person would thus consider them to be suitable for the final adsorption step. No unexpected effect associated with the use of a zeolite adsorbent has been invoked by the appellant. The use of this adsorbent material is thus merely one obvious possibility amongst others.

8.2 D3 does not expressly address the possibility of using a hydrogen oxidation catalyst as defined in present claim 1. However, a catalyst comprising, on a carrier material, two or more different catalytic noble metals which are individually suitable for the intended hydrogen oxidation step must be expected to also be suitable for the latter purpose. As apparent from D2 (page 1, lines 22 to 28), and as confirmed by the present application as filed (page 3, lines 23 to 30), palladium and platinum were both known to be interchangeably suitable for catalysing the oxidation of hydrogen present in air to be purified at elevated temperatures. Therefore, the skilled person would also consider platinum to be a "noble metal catalyst"

suitable for being used in a hydrogen oxidation step according to D3, despite the lower temperatures prevailing in the hydrogen oxidation according to D3. At the oral proceedings, the appellant has expressly not invoked any improvement that could be attributed to the claimed use of a catalyst comprising palladium and platinum supported on alumina instead of palladium on alumina as exemplified in D3. The use of the former material as the catalyst is thus merely one obvious possibility amongst others.

- 8.3 Concerning the adsorption of carbon dioxide in a process combining the teachings of D2 and D3, the board notes that according to present dependent claim 6 (claim 9 of the application as filed), "*at least some*" carbon dioxide may be adsorbed on the "*solid adsorbent*", i.e. before hydrogen oxidation. In conjunction with the passage of the description, where reference is (still) made to the adsorption of "*any residual carbon dioxide*" (amended page 12, lines 17 to 19), this means that present claim 1 has to be understood as also encompassing processes wherein, like in the process of D2, substantially all of the carbon dioxide present after the carbon monoxide oxidation is removed by adsorption on the "*solid adsorbent*", and that a further significant adsorption of residual carbon dioxide on the zeolite used for adsorbing the water formed by hydrogen oxidation need not necessarily occur. Correspondingly, the appellant has acknowledged (reply of 27 May 2008; page 6, seventh paragraph) that according to the application "carbon dioxide may or may not be present at the stage of the final water adsorption" and merely stated in this connection that "in the normal practice of the invention it is

envisaged that carbon dioxide will remain to be adsorbed at the end of the process".

- 8.3.1 When combining the teachings of D2 and D3 in the manner addressed under point 7. above, the skilled person will immediately recognise that the carbon dioxide may be removed either by i) full carbon dioxide adsorption in the first adsorption step according to D2 or by ii) partial carbon dioxide adsorption in the first water adsorption step and final carbon dioxide adsorption in the final water adsorption step according to D3. More particularly, the skilled person will recognise that by keeping the concept of full carbon hydrogen adsorption before hydrogen oxidation (as disclosed in D2), the final water adsorption may be designed without having to consider the presence of any residual carbon dioxide. On the other hand, he will recognise that by foreseeing a carbon dioxide removal in the final water adsorption step (as disclosed in D3), the carbon dioxide need not be fully removed prior to the hydrogen oxidation step.
- 8.3.2 The process alternatives according to claim 1 wherein the carbon dioxide content (originally present and formed by oxidation of carbon monoxide) of the air stream is removed by either alternative i) or ii) are thus two possibilities for combining the teachings of D2 and D3 involving only routine engineering considerations.
- 8.4 The replacement of the final hydrogen adsorption according to D2 by the hydrogen oxidation step using a catalyst of palladium and platinum supported on alumina, followed by the adsorption of the water formed and of any residual carbon dioxide present on a zeolite

adsorbent thus leads to a process falling within the ambit of claim 1 in an obvious manner, involving no inventive skills.

9. The appellant's arguments aiming at establishing the presence of an inventive step are not convincing for the following reasons:

9.1 As pointed out by the appellant, the authors of D2 considered the process of D3 to be disadvantageous (D2, page 2, lines 4 to 21) since it requires an additional adsorbent bed for removing the water vapour and carbon dioxide generated by the catalytic oxidation of carbon monoxide and hydrogen. Water vapour and carbon dioxide thus needed to be removed twice, namely in a first adsorption step, and then again after being generated in the oxidation steps. However, this particular aspect of the process of D3, considered as to be disadvantageous by the authors of D3, is not related to the purity of the treated air that may be achieved. Moreover, the authors of D2 do not challenge the efficiency of the hydrogen and water vapour removal achieved according to D3. There is also no clear indication in D2 that the cyclic hydrogen adsorption/adsorbent regeneration proposed would be more efficient in terms of capital or operating costs than a hydrogen oxidation/water adsorption according to D3.

The board is convinced that under these circumstances the skilled person would expect the combination of hydrogen oxidation and subsequent adsorption of the water produced to give satisfactory results in terms of the air purity achieved. Therefore, the skilled person

starting from D2 and considering the acknowledgement of D3 therein would not be discouraged by the particular aspect of the method of D3 which the authors of D2 merely declare to be disadvantageous, but would definitely envisage the replacement of the final hydrogen adsorption by a catalytic hydrogen oxidation with subsequent water adsorption as a possible solution to the stated technical problem, despite the need for an additional adsorption step.

- 9.2 According to a further argument of the appellant, the disclosure of D2 casts doubt on what is said in D3 regarding the ability of a material containing a supported noble metal to act as a catalyst for the oxidation of hydrogen under the conditions described in D3. However, the board notes that the authors of D2, when acknowledging the teaching of D3, have not questioned the teaching of this document. The board therefore takes the view that the fact that D2 is the later document and refers to conditions under which hydrogen is not oxidised to water (D2; page 5, lines 29 to 35) is, as such, not sufficient to harbour such doubts, let alone to stop the skilled person from considering the replacement of the hydrogen removal technique of D2 by the one of D3.
10. The board thus concludes that the subject-matter of claim 1 is not based on an inventive step as required by Articles 52(1) and 56 EPC.
11. The appellant's request can thus not be allowed.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar

The Chairman

C. Vodz

G. Raths