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**D E C I S I O N**  
**of 16 February 2001**

**Case Number:** T 0661/97 - 3.3.5

**Application Number:** 93307703.4

**Publication Number:** 0590946

**IPC:** B01D 53/04

**Language of the proceedings:** EN

**Title of invention:**  
Production of nitrogen

**Applicant:**  
THE BOC GROUP, INC.

**Opponent:**

-

**Headword:**  
Nitrogen/BOC

**Relevant legal provisions:**  
EPC Art. 54(1)

**Keyword:**  
"Novelty - no - a prior art document must be interpreted in a technically realistic context"

**Decisions cited:**  
T 0450/89

**Catchword:**

-



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Boards of Appeal

Chambres de recours

**Case Number:** T 0661/97 - 3.3.5

**D E C I S I O N**  
**of the Technical Board of Appeal 3.3.5**  
**of 16 February 2001**

**Appellant:** THE BOC GROUP, INC.  
575 Mountain Avenue  
Murray Hill  
New Jersey 07974 (US)

**Representative:** Wickham, Michael  
c/o Patent and Trademark Department  
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**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office posted 28 January 1997  
refusing European patent application  
No. 93 307 703.4 pursuant to Article 97(1) EPC.

**Composition of the Board:**

**Chairman:** G. J. Wassenaar  
**Members:** A.-T. Liu  
M. B. Günzel

### Summary of Facts and Submissions

I. European patent application No. 93 307 703.4, publication No. 0 590 946, was refused by a decision of the Examining Division.

II. The Examining Division held that the subject-matter of the claims then on file lacked an inventive step in view of

D1: US-A-5 085 674

and the common general knowledge of a person skilled in the art of gas phase adsorption.

III. With the statement of grounds of the appeal the appellant filed a new set of claims. In a communication of the Board the preliminary opinion was expressed that the subject matter of at least the independent claims 1 and 8 seemed to lack novelty over

D2: US-A-4 746 332.

In reply the appellant filed a new set of claims 1 to 7. In a communication annexed to the summons to attend oral proceedings the Board expressed as its preliminary opinion that the novelty of the process according to claim 1 was still questionable in view of D2. In response the appellant submitted two new sets of claims 1 to 7 as main and auxiliary request, together with an amended description for the main request.

IV. Claim 1 of the main request read as follows:

"A process for producing high purity nitrogen product comprising removing carbon monoxide from a feed gaseous nitrogen or air stream which contains carbon monoxide as an impurity by a temperature swing adsorption process comprising the step of passing said stream through a bed of carbon monoxide-selective adsorbent at a temperature below 150 K, thereby producing a substantially carbon monoxide-free stream, wherein the said carbon monoxide-selective adsorbent is selected from calcium exchanged X zeolite, copper exchanged Y zeolite, 5A zeolite, 13X zeolite, and mixtures thereof, and wherein when the feed stream is gaseous nitrogen, the said substantially carbon monoxide-free stream is the high purity nitrogen product, but when the feed stream is air the said substantially carbon monoxide-free stream is fractionally distilled to form the high purity nitrogen product."

Claim 1 of the auxiliary request differed therefrom in that the carbon monoxide-selective adsorbent was limited to 5A zeolite.

V. The appellant's arguments with respect to novelty may be summarized as follows:

D2 concerned the removal of oxygen impurities and did not treat the problem of removing carbon monoxide impurities. In view of the disclosure in D2 that 5A zeolite is a Ca-A type zeolite and that Ca-A- type zeolites have remarkably large adsorption capacity for nitrogen, so that it is practically used for selectively removing nitrogen to separate oxygen, the actual technical teaching of D2 was to use an A-type

zeolite but not to use 5A zeolite for the adsorption of oxygen at cryogenic temperatures. No instructions were provided how to use 5A zeolite for doing just the opposite, ie the selective adsorption of oxygen from nitrogen comprising oxygen as an impurity. Example III of the present application showed that oxygen was not adsorbed to a substantial amount by a 5A zeolite. In the process according to D2 the feed gaseous nitrogen to be treated did not necessarily contain carbon monoxide. The air from which the feed gaseous nitrogen was obtained could be free from carbon monoxide, or the carbon monoxide in the air could have been removed from the nitrogen feed before the oxygen removal. In this respect reference was made to the text book "Separation of Gases" by W. H. Isalski, Clarendon Press Oxford, 1989, Table 3.1. Whether carbon monoxide was removed depended on the bed size and adsorption time, important parameters about which D2 was silent. With reference to T 450/89 it was stressed that for lack of novelty it was necessary that the prior art document comprised clear and unmistakable instructions to perform the claimed subject-matter of the later invention, a requirement that D2 clearly did not fulfil.

- VI. The appellant requested that the decision under appeal be set aside and a patent be granted on the basis of claims 1 to 5 of the main request filed with the letter dated 12 January 2001. As auxiliary request, the appellant requested that a patent be granted on the basis of claims 1 to 5 of the auxiliary request filed with the same letter, claims 6 and 7 of both requests having been deleted during the oral proceedings which were held on 16 February 2001.

## Reasons for the Decision

1. Claim 1 of the main request concerns a process for producing high purity nitrogen comprising alternative solutions to solve that problem. One of these solutions is to pass a stream of nitrogen comprising carbon monoxide as an impurity through a bed of 5A zeolite at a temperature below 150K, whereby substantially carbon monoxide-free nitrogen is obtained and to desorb the bed at a higher temperature for reuse of the bed; ie by a temperature swing adsorption process (TSA).
  
2. The prior art document D2 also discloses a process for producing high purity nitrogen whereby a stream of nitrogen comprising oxygen as an impurity is passed through a bed with an A-type zeolite in a TSA process at an adsorption temperature of  $-100^{\circ}$  to  $-196^{\circ}\text{C}$  (77 to 173K). Specifically disclosed are 4A and 5A zeolites and the adsorption temperature is preferably between  $-150$  and  $-196^{\circ}\text{C}$  (77 to 123K); see column 2, lines 39 to 44 and 60-63. It is further disclosed that Ca-A type zeolite (=5A) has a remarkably large adsorption capacity as to nitrogen, so that it is practically used for selectively removing nitrogen to separate oxygen and that the use of Na-A type zeolite is thus preferable (column 2 line 63 to column 3, line 2). On the basis of the last cited passage the appellant argued that D2 did not disclose the use of 5A zeolite for the process disclosed therein. The Board cannot accept this for the following reasons.
  
3. In the claims, the "summary of the invention" and the examples of D2, the adsorbent is constantly referred to as A-type zeolite without any restriction. Together with the statement that 4A zeolite is preferable it

follows that the other specifically mentioned 5A zeolite, although not preferable, is also suitable. This is not contradictory to the statement that 5A zeolite has a remarkable large adsorption capacity as to nitrogen and is practically used for selectively removing nitrogen to separate oxygen. The latter statement is understood by the Board to relate to earlier adsorption processes not performed at the required cryogenic conditions. As shown in Figure 1 of D2 the adsorption capacity of zeolites for nitrogen and oxygen is very much dependent upon the adsorption temperature. According to said Figure 1 the adsorption capacity of 4A zeolite for nitrogen is also higher than for oxygen at temperatures between -10 and -100°C, so that at these higher temperatures 4A zeolite could also be used for selectively adsorbing nitrogen. Although, according to column 2, lines 66 to column 3 line 1, the graph in Figure 1 is based on experiments with a 4A zeolite, in the description of the preferred embodiments the graph of Figure 1 is presented as representative for A-type zeolite in general (column 2, lines 10 to 24).

4. The appellant's argument that example III of the present application confirmed that 5A zeolite did not adsorb oxygen at cryogenic temperatures so that a skilled person would understand that in D2, 5A zeolite was not intended for removing oxygen from nitrogen under the conditions mentioned there, is not convincing. From present example III it might be derived that oxygen was less strongly adsorbed on 5A zeolite at cryogenic temperatures than carbon monoxide but not that oxygen was less strongly adsorbed than nitrogen and that 5A zeolite was thus not suitable for separating oxygen from nitrogen under the conditions

mentioned in D2. Moreover the skilled person was not aware of present example III before the publication of the present application and could not have used its information for considering whether D2 disclosed the use of 5A zeolite or not for the removal of oxygen. There is no prior art evidence that at cryogenic temperatures 5A zeolite does not adsorb oxygen to such an extent that it cannot be used for its removal from nitrogen.

The Board holds, therefore, that 5A zeolite is disclosed in D2 as being only gradually different in adsorption behaviour from 4A zeolite and less preferable but still suitable for the adsorption of oxygen from nitrogen at the required cryogenic conditions.

5. The starting gas to be treated according to D2 is one obtained from a rectifying column in a low temperature separating apparatus for air that comprises 99,9% by volume or more of nitrogen and 0.1% by volume or less of oxygen (claim 1 and example 1). D2 is silent about the carbon monoxide content of the gas to be treated but that does not mean that carbon monoxide is absent. As acknowledged in the present application, if carbon monoxide is present in the air feed as an impurity and it is not removed from the feed stream prior to entry of the feed stream into the distillation system, it will end up in the nitrogen-enriched stream, because its boiling point is very close to that of nitrogen (page 2, lines 10-13 of the published application). Although the amount of carbon monoxide in air may be practically zero as testified by Table 3.1 of the cited textbook of W. H. Isalski, normal air and certainly air in industrial areas where air separation plants are



normally operated, always contain carbon monoxide. If that were not the case, there would have been no need to remove carbon monoxide. According to said Table 3.1 the amount of carbon monoxide in the air may be up to 35 v.p.m. The determination of the disclosure of a prior art document should be based on realistic operation conditions and not on theoretically possible extreme conditions. Thus when performing the process of D2 under realistic conditions the air used in the air separation plant always contains detectable amounts of carbon monoxide.

6. In the process according to Figure 2 of the present application the nitrogen gas containing carbon monoxide passed through the adsorbent bed also comes from the rectifying column in a cryogenic air separation plant (page 4, lines 41 to page 5, line 19 of the published application). The feed treated in the present application is thus identical to the feed treated according to D2 so that in the process according to D2 the nitrogen feed also comprises carbon monoxide and the latter is removed if passed through a bed containing 5A zeolite.
  
7. The appellant's argument that in the process according to D2 carbon monoxide could have been removed before the feed nitrogen entered the adsorption bed is not convincing. In the plants according to Figures 3 and 4 of D2 no such removal units are present. There is also no basis for the appellant's assumption that after the normal rectifying column an additional rectifying step could have been used to separate carbon monoxide by fractional distillation from the nitrogen stream from the rectifying column. In view of the acknowledged close boiling points of nitrogen and carbon monoxide

this alleged option would be unrealistic. The appellant's further argument that D2 does not disclose bed volume and adsorption times so that time and volume could be insufficient to remove carbon monoxide is also not convincing. Present claim 1 does not require the complete removal of carbon monoxide or the removal to a minimum amount. In a bed volume and adsorption time sufficient to remove substantial amounts of oxygen it is unavoidable that at least some carbon monoxide is removed if 5A zeolite is used as adsorbent.

8. The Board concurs with the decision T 450/89 of 15 October 1991, 3.11 of the reasons, cited by the appellant, in that novelty should be affirmed if the prior art document does not comprise clear and unmistakable disclosure for the subject-matter of the later invention (point 3.11). Whether subject-matter is clearly and unmistakably disclosed is, however, a matter of facts to be decided by the Board depending on the specific circumstances of the case. For the reasons given above the Board holds that in the present case D2 clearly and unmistakably discloses the use of 5A zeolite as adsorbent for the purification of nitrogen at cryogenic temperatures and that by using that adsorbent under the conditions mentioned in D2, carbon monoxide, inevitably present in crude nitrogen from the air separation plant, is removed from the feed nitrogen.
  
9. For these reasons the Board concludes that the subject-matter of claim 1 of the main request lacks novelty over D2. Since claim 1 of the auxiliary request also requires the use of 5A zeolite as adsorbent the same applies to the subject-matter of claim 1 of the auxiliary request.

**Order**

**For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

The Chairman:

S. Hue

G. Wassenaar