DECISION
of 31 May 2001

Case Number: T 0486/97 - 3.3.5
Application Number: 91108835.9
Publication Number: 0461478
IPC: B01D 53/04

Language of the proceedings: EN

Title of invention: Improved air separation pressure swing adsorption process

Patentee: PRAXAIR TECHNOLOGY, INC.

Opponent: Bayer AG Konzernbereich RP Patente und Lizenz
The BOC Group plc
L’AIR LIQUIDE, S.A. pour l’étude et l’exploitation des procédés Georges Claude
LINDE AKTIENGESELLSCHAFT

Headword: -

Relevant legal provisions:
EPC Art. 84, 123(2), 123(3), 52(1), 56

Keyword: "Inventive step (no): obvious combination of prior art teachings with expected results"

Decisions cited:
-

Catchword:
Case Number: T 0486/97 - 3.3.5

DEcISION
of the Technical Board of Appeal 3.3.5
of 31 May 2001

Appellant: PRAXAIR TECHNOLOGY, INC.
(Proprietor of the patent) 39 Old Ridgebury Road
Danbury, CT 06810-5113 (US)

Representative: Schwan, Gerhard, Dipl.-Ing.
Schwan Schwan Schorer
Patentanwälte
Elfenstrasse 32
D-81739 München (DE)

Respondent: Bayer AG
(Konzernbereich RP
Patente und Lizenzen
D-51368 Leverkusen (DE)

Representative: -

Respondent: The BOC Group plc
(Opponent) Chertsey Road
Windlesham
Surrey GU20 6HJ (GB)

Representative: Wickham, Michael
(c/o Patent and Trademark Department
The BOC Group plc
Chertsey Road
Windlesham
Surrey GU20 6HJ (GB)

Respondent: L'AIR LIQUIDE, S.A.
(Opponent) pour l'étude et l'exploitation
des procédés Georges Claude
75 quai d'Orsay
FR-75321 Paris (FR)

Representative: Le Moenner, Gabriel
L'AIR LIQUIDE, Société Anonyme
pour l'étude et l'exploitation des procédés
Georges Claude
75, Quai d'Orsay
FR-75007 Paris (FR)
Respondent: LINDE AKTIENGESELLSCHAFT
(Opponent) Zentrale Patentabteilung
D-82049 Höllriegelskreuth (DE)

Representative: -

Decision under appeal: Decision of the Opposition Division of the European Patent Office posted 13 March 1997 revoking European patent No. 0 461 478 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. The appeal is from a decision of the Opposition Division revoking the patent in response to four oppositions filed on the grounds of Articles 100(a) and (b) EPC.

II. The decision under appeal was based on the independent claims 1, 2 and 4 as amended with the letter of 28 July 1996, with dependent claims 3 and 5 to 10 as granted. Independent claim 1 reads as follows:

"1. A pressure swing adsorption process for the production of high purity oxygen by separation of air, employing a pressure swing adsorption system containing at least two adsorbent beds of adsorbent material capable of selectively adsorbing nitrogen from feed air, with oxygen and argon passing through and being discharged from the beds, said process comprising, on a cyclic basis, adsorption at an upper adsorption pressure and desorption at a lower desorption pressure, wherein said adsorption/desorption sequence is carried out under subatmospheric cycle conditions in which said upper adsorption pressure is at or slightly below atmospheric pressure and said lower desorption pressure is a (sic) subatmospheric pressure, the adsorption/desorption pressure ratio range being from about 1.4:1 to about 4:1, and wherein said adsorbent material comprises NaX zeolite adsorbent highly exchanged to replace Na⁺ ions therein with lithium cations resulting in a zeolite adsorbent in which the molar ratio of the SiO₂/Al₂O₃ framework is less than 3.0 and at least about 88% of its Al₂O₃ tetrahedral units are
associated with lithium cations, or modified by caustic digestion at elevated temperature to increase the zeolite content thereof, or modified by a combination thereof, whereby the separation of oxygen and argon from nitrogen is desirably accomplished at advantageously low power consumption levels for the adsorption/desorption sequence and with favourable adsorbent utilization requirements."

Independent claims 2 and 4 substantially relate to the same process, with the only difference that the pressure swing conditions are transatmospheric or superatmospheric, respectively.

III. Of the documents cited in the opposition proceedings, reference shall be made to the following in the present decision:

D1 EP-A-0 297 542


D9 FR-A-1 303 222

D13 US-A-4 810 265

IV. In the impugned decision, the opposition division essentially held that the introduction of the expression "production of high purity oxygen" into the amended claims did not contravene Articles 84, 123(2) and (3) EPC.
D1 was considered to represent the closest prior art. The technical problem to be solved with regard to D1 was seen in the cost reduction in the production of oxygen. The solution proposed in claim 1 was essentially the selection of an adsorption / desorption pressure ratio within the range from 1.4 : 1 to 4 : 1. The proposed solution was however found to be obvious in view of D6. The presence of an inventive step was therefore denied.

V. With the statement of the grounds of appeal, the appellant (proprietor) filed amended pages of description.

VI. Oral proceedings took place on 31 May 2001, at which the opponent Bayer AG, as announced in the letter of 9 February 2001, was not represented.

VII. The appellant concurred with the opposition division on the finding concerning the compliance of the amended claims with Articles 84 and 123 EPC. His arguments, given in writing and at the oral proceedings, may be summarised as follows:

- D9 should be considered to represent the closest prior art.

- The problem was seen in reducing the cost associated with a pressure swing adsorption process for producing a specified quantity of high purity oxygen.

- The solution proposed in claim 1 was to carry out the process at an unusually low ratio of adsorption to desorption pressure.
The art, in particular D13, did not give the skilled person an incentive for lowering the pressure ratio.

VIII. The arguments of the respondents were essentially the following:

- The amended claims contravened Articles 84, 123(3) and 100(c) EPC.

- The reasoning and the assessment of inventive step by the opposition division was correct.

- Alternatively, one could start from D6 as closest prior art. In that case, the claimed processes were also obvious in combination with D1.

IX. The appellant (patentee) requested that the decision under appeal be set aside and that the patent be maintained on the basis of amended claims 1, 2 and 4 as submitted with the letter of 28 July 1996 with claims 3 and 5 to 10 as granted.

The respondents - opponents BOC group PLC, Air liquide and Linde AG requested that the appeal be dismissed.

Reasons for the Decision

1. Amendments

Claim 1 as amended differs from claim 1 as granted in that it is now directed to "a pressure swing adsorption process for the production of high purity oxygen"
(emphasis added) by separation of air" instead of being directed to "a pressure swing adsorption process for the separation of air" (emphasis added)."

1.1 The Board concurs with respondent BOC group plc that the purity level of the oxygen stream is not indicated in the examples. This objection, however, would only imply that the amended claims may not be supported by the description as required by Article 84 EPC but it is not a reason for querying the basis for the amendment under Article 123(2) EPC. In fact, the application of the pressure swing adsorption process to the "production of high purity oxygen" is disclosed in the description as filed, page 7, lines 31 to 33.

1.2 It is well known in the art and also originally disclosed in the patent application that a pressure swing adsorption process for air separation provides oxygen and nitrogen component streams (see page 7, lines 13 to 30 and page 8, lines 12 to 13). The Board therefore holds that the amendment stipulating the production of oxygen by air separation corresponds to the deletion of the alternative of producing nitrogen by air separation, which alternative was previously encompassed by the wording of claim 1 as granted. As a consequence, claim 1 also satisfies the requirements of Article 123(3) EPC.

2. Clarity

The expression "high-purity oxygen", newly introduced into claim 1, is indeed not precise in the sense that it is not defined by a concrete numerical range. It is however irrefutable that the oxygen product obtained in D6, with a purity of 90 to 95% by volume, is also
considered as being of "high-purity" (see page 6, "Product purity"). The exact interpretation for this term is thus inessential for the assessment of inventive step. Since the appeal has to be dismissed as a result of the assessment of inventive step (see point 4 below), the issue of clarity need not be considered further.

3. **Novelty**

None of the cited prior art documents disclose the combination of features of claim 1; the claimed process is thus novel. This is undisputed.

4. **Inventive step**

4.1 In the Board's judgment, the closest prior art is represented by D6 which is directed to the same subject-matter as claim 1, namely the use of pressure swing adsorption processes for the recovery of oxygen from air (see page 1, Sections "1. Introduction" and "3. Description of the process"). Although D1 also relates to processes involving the selective adsorption of nitrogen from air, it more specifically concerns the recovery of nitrogen (see page 2, lines 4 to 6).

D6 discloses a pressure swing adsorption system comprising three adsorber vessels filled with zeolitic molecular sieves and working under subatmospheric cycle conditions (page 1, last two paragraphs; page 2, Figure 1).

4.2 With respect to D6, the technical problem can be seen in the provision of an improved method for the recovery of oxygen by adsorption.
4.3 The solution proposed in claim 1 is the stipulation that:

(i) the adsorption/desorption pressure ratio range is from about 1.4:1 to about 4:1, and

(ii) the adsorbent material comprises NaX zeolite adsorbent highly exchanged to replace Na\(^+\) ions therein with lithium cations resulting in a zeolite adsorbent in which the molar ratio of the \(\text{SiO}_2/\text{Al}_2\text{O}_3\) framework is less than 3.0 and at least about 88% of its \(\text{Al}_2\text{O}_3\) tetrahedral units are associated with lithium cations, or modified by caustic digestion at elevated temperature to increase the zeolite content thereof, or modified by a combination thereof.

4.4 Although a direct comparison has not been made with the process of D6, test results given in the patent in suit allow a comparison to be made between the processes which are carried out according to the conditions as stipulated in claim 1 and those outside of these boundaries.

The data in the patent in suit show that when the adsorbent NaX is used at a pressure ratio of 6:1, the power consumption is 100 kW and the bed size factor 914 (Table I). In contrast, with the adsorbent LiX (2.5) used at a pressure ratio of 2:1, the power consumption is reduced to 73 kW. At the same time, however, the bed size factor rises to 1 127 (Table II). The conclusion which can be drawn from the available experimental data is that the present process results in the separation of oxygen from nitrogen at relatively low power consumption levels for the adsorption / desorption...
sequence but at the expense of adsorbent utilization requirements. Since the appellant argues that the power consumption reduction is more important from the economic point of view, the Board can in the present case accept that the technical problem stated in point 4.2 is solved by the process according to claim 1.

4.5 The remaining question is whether the solution to the technical problem, as proposed in claim 1, is obvious in view of the available prior art.

4.5.1 Re characterising feature (i)
Adsorption/desorption pressure ratio

The basic process for the oxygen recovery from air is carried out in D6 with the adsorption and desorption pressure ranges preferably between 1 and 1.1 bars and between 200 and 300 mbars, respectively (see page 2, paragraph above Figure 1). The corresponding pressure ratio range of 3.3:1 to 5.5:1 thus overlaps the ratio range stipulated in claim 1.

It is further recognised in D6 that the pressure conditions affect the economy of the adsorptive oxygen recovery process (see page 3, Section "4. Influence on economy" and page 7, paragraph 2). In particular, it is found that, for a given molecular sieve, the required quantity of molecular sieve - which corresponds to the bed size factor "BSF" in the vocabulary of the patent in suit - is reduced as the adsorption pressure rises. At the same time, the energy consumption increases linearly in first approximation. On the other hand, the BSF decreases with desorption pressure, at the cost of increasing energy consumption. The choice of the most
economic pressure conditions is thus a compromise between these two factors (see Sections "4.2 Adsorption pressure" and "4.3 Desorption pressure").

D6 thus not only discloses a pressure ratio range overlapping the range stipulated in claim 1. It also teaches the skilled person how to optimise that ratio according to need.

4.5.2 Re characterising feature (ii)
Adsorption material

Experiments are also conducted in D6 to determine the influence of various types of commercial molecular sieves on the size of the adsorber and energy consumption (see page 3, Section "4.1 Adsorption material" including Figure 2). It is undisputed that D6 is silent about the exact type of the zeolitic materials investigated therein. However, the conclusion of these studies is clear in that it teaches the skilled person to select those materials with high N₂/O₂ loading capacities when looking for alternatives for the molecular sieves already in use in the recovery of oxygen by adsorption (page 3, last two paragraphs).

A molecular sieve described as having extraordinary capacity and selectivity toward the adsorption of nitrogen from gas streams containing oxygen, is disclosed in D1, published in 1989, the same year as D6 (see abstract and page 3, lines 3 to 7). It is thus obvious for the skilled person to apply the zeolite of D1 to the oxygen recovery processes according to D6. The adsorbent concerned is a lithium exchanged zeolite in which the framework SiO₂/Al₂O₃ molar ratio is from about 2.0 to 3.0 and in which at least about 88% of its
Al₂O₃ tetrahedral units are associated with lithium cations (claims 1 to 5). It is undisputed that this type of lithium exchanged zeolite is exactly the same adsorbent as stipulated in claim 1.

4.5.3 Re combination of features of claim 1

The explicit teaching of D6 is that of the parameters influencing the economy of the oxygen recovery process, considerable importance is attached to the adsorption material and the adsorption/desorption pressure. The skilled person further learns from D6 that the most economic pressure conditions are the result of a compromise between the size of the adsorber bed (BSF) and the energy consumption.

In the patent in suit, tests are carried out at an approximately constant adsorption pressure of 150 kPa or 140 kPa, and at two distinct desorption pressures of 25 and 70 kPa, corresponding to a pressure ratio of 6:1 and 2:1, respectively (Examples 2 to 3). As expected, the power consumption is found to be reduced with increased desorption pressure (i.e. with reduced pressure ratio). However, the gain in power efficiency is also obtained at the expense of a considerably larger adsorbent requirement (compare Table I and II and see page 6, lines 38 to 42). Thus, the finding is essentially the same as in D6. The appellant has not argued and it is not plausible that the combination of features of claim 1, in particular the use of the stipulated adsorbent material under the prescribed pressure conditions would result in any unexpected technical effect.

As is expressly acknowledged in the patent in suit, the
selection of the pressure ratio range as stipulated in claim 1 is based on the ground that power savings are considered a more significant economic factor than adsorbent requirements (see page 6, lines 44 to 45 and point 4.4 above). This particular view, however, entirely depends on subjective considerations which may well vary with time and extraneous circumstances. In the Board's judgment, the optimisation of the process in function of these circumstances belongs to the routine tasks of the notional person skilled in the art. The resulting selection of the pressure ratio range, which is within the framework of D6, therefore does not require inventive skill. In consequence, the process of claim 1 is obvious in view of D6 in combination with D1.

4.6 The Board would not have arrived at a different conclusion, had it followed the appellant and accepted D9 as representing the closest prior art.

4.6.1 D9 concerns a pressure swing adsorption process for recovering oxygen with a purity of approximately 93% by separation from a mixture with nitrogen (page 1, right hand column, first full paragraph; page 3, second paragraph following Tab. I; page 6, left hand column, last full paragraph). One of the adsorbents used in the process is X zeolite exchanged with lithium cations, in which 86% of the \( \text{Al}_2\text{O}_3 \) tetrahedral units are associated with lithium cations (page 3, Table I).

4.6.2 With regard to D9, the Board can also see the technical problem in the provision of an improved method for the recovery of oxygen by adsorption.

4.6.3 The solution proposed in claim 1 is also characterised
here by the choice of the adsorbent material and the pressure conditions (compare point 4.3).

4.6.4 It is already apparent in D9 that the efficiency of the zeolite is in proportion with its degree of cation exchange (page 4, right hand column, first paragraph). In the Board's judgment, the skilled person thus must recognise that the adsorbent used in D9, published in 1962, is practically obsolete, considering the developments that have been made in the field of zeolitic adsorbent materials in the meantime. It is thus straightforward for the skilled person to replace the adsorbent of D9 with the higher exchanged adsorbent of D1 which is precisely known as a better adsorbent for the same application.

The Board further holds that, for the same reasons as indicated in points 4.5.1 and 4.5.3, the skilled person would follow the general teaching of D6 and optimise the process parameters such as the pressure conditions in relation to the type of adsorbent applied.

4.7 The appellant has also argued that, according to D13, the lowest pressure ratio applicable is 4.5 when LiX is used as adsorbent (Table V and column 27, lines 30 to 35). The skilled person therefore would not look into the claimed pressure ratio range in order to reduce the specific power consumption.

The Board, however, notes that the cited passage concerns an example given for illustrative purposes. It is expressly indicated that such examples should not be construed as limiting the teaching of D13 (see column 26, lines 11 to 21). The Board therefore has no reason to presume that the pressure ratio of that
particular embodiment should represent the lowest ratio envisaged by D13.

The Board further observes that D13 is specifically directed to a pressure swing adsorption process for recovering nitrogen from air. It may be accepted that the efficiency of such a process is based on the same parameters as for a similar process where the product to be recovered is oxygen. However, it is also undisputed that a skilled person would expect that some variation in the specific reaction conditions is necessary depending on whether oxygen or nitrogen is the desired product (see for example patent in suit, page 9, lines 9 to 12). Thus, irrespective of the interpretation attributed to the teaching of D13, the Board holds that this cannot be construed as a prejudice against the application of a lower pressure ratio in the oxygen recovery by gas separation.

**Order**

For these reasons it is decided that:  
The appeal is dismissed.

The Registrar: The Chairman:

G. Rauh R. Spangenberg