BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

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File Number: T 379/88 - 3.3.2

Application No.: 83 401 022.5

Publication No.: 0 096 610

Title of invention: Air separation process for the production of krypton and xenon

Classification: C01B 23/00

D E C I S I O N
of 15 January 1991

Applicant:

Proprietor of the patent: Union Carbide Corporation

Opponent: Linde Aktiengesellschaft

Headword: Separation of air/UNION CARBIDE

EPC Article 56

Keyword: "Inventive step - (no) - combination of known processes - no evidence for alleged improved efficiency"

Headnote

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number : T 379/88 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 15 January 1991

Appellant : (Opponent)
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Representative :

Respondent : (Proprietor of the patent)
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Decision under appeal : **Decision of the Opposition Division of the European Patent Office dated 3 December 1987, posted on 7 June 1988, rejecting the opposition filed against European patent No. 0 096 610 pursuant to Article 102(2) EPC**

Composition of the Board :

Chairman : P. Lançon
Members : I.A. Holliday
R. Schulte

Summary of Facts and Submissions

I. European patent No. 0 096 610 was granted on the basis of ten claims contained in European patent application No. 83 401 022.5. Claim 1 reads as follows:

1. In a process for the separation of air wherein air at greater than atmospheric pressure is subjected to rectification in a high pressure column (10) and a low pressure column (20) which are in heat exchange relation at a heat exchange stage, the improvement, whereby a fraction containing a relatively high concentration of krypton and xenon is produced, comprising:

(a) introducing a gaseous oxygen-rich stream (72), containing krypton and xenon, taken from the low pressure column above said heat exchange stage, into a rare gas stripping column (40) provided with a first bottom reboiler (86);

(b) introducing a liquid oxygen-rich stream (73), taken from the low pressure column at a point above that from which said gaseous oxygen-rich stream is taken, into the rare gas stripping column as descending liquid reflux in an amount such that the reflux ratio of the rare gas stripping column is from 0.1 to 0.3;

(c) stripping krypton and xenon from the gaseous oxygen-rich stream into the descending liquid reflux;

(d) partially vaporizing the liquid reflux in the first reboiler by indirect heat exchange with a first condensing gaseous nitrogen-rich stream (83) taken from the high pressure column;

(e) returning the resulting condensed nitrogen-rich stream (84) from step (d) into either the high pressure column or the low pressure column;

(f) recovering from the rare gas stripping column a liquid first rare gas stream (90) comprising krypton, xenon and oxygen wherein krypton and xenon are in a concentration greater than their concentration in the descending liquid reflux;

(g) introducing said liquid first rare gas stream into an oxygen exchange column (50) provided with a second bottom reboiler (95);

(h) introducing a gaseous nitrogen stream (85, 97), taken from the high pressure column, into the oxygen exchange column in an amount such that the reflux ratio is from 0.15 to 0.35;

(i) passing in said oxygen exchange column said liquid first rare gas stream against said gaseous nitrogen stream such that oxygen in the liquid first rare gas stream is replaced by nitrogen;

(j) withdrawing the resulting oxygen-containing gaseous nitrogen-rich stream (104) of step (i) from the oxygen exchange column and introducing it into the low pressure column;

(k) partially vaporizing the resulting nitrogen-containing liquid first rare gas stream of step (i) in the second reboiler by indirect heat exchange with a second condensing gaseous nitrogen-rich stream (85, 98) taken from the high pressure column;

(l) returning the resulting condensed nitrogen-rich stream (101) of step (k) into either the low pressure column or the high pressure column; and

(m) recovering a liquid second rare gas stream (100) comprising krypton, xenon and nitrogen wherein krypton and xenon are in a concentration greater than their concentration in the liquid first rare gas stream.

II. The Appellant filed a notice of opposition against the European patent requesting revocation of the patent on the grounds that its subject-matter lacked an inventive step.

Two prior art documents were cited in support of the opposition:

(1) US-A-3 751 934
(2) DE-A-1 122 088.

III. The Opposition Division rejected the opposition taking the view that the subject-matter claimed was novel vis-à-vis the prior art cited. The Division were also of the opinion that a simple hypothetical combination of documents (1) and (2) would not have led to the process of the patent in suit. Document (1) has a clear teaching to perform the working-up process for krypton and xenon only after oxygen is recycled from the stripping column to the main column thus loading the main column with hydrocarbons. By avoiding this recycling in the process of the patent, a safety hazard was reduced. It was also the Division's view that the proprietor had produced convincing arguments in favour of the efficiency of the process, i.e. the energy demand in relation to the yield of xenon and krypton.

IV. The Appellant lodged an appeal against this decision in which it was sought to introduce a further document into the procedure, namely:

(3) Ullmann's Encyclopädie der technischen Chemie, Band 10, 4 Auflage, 1975, Seite 286.

V. Oral proceedings took place on 15 January 1991.

VI. The arguments of the Appellant, in the Statement of Grounds of Appeal, a further communication and at the oral proceedings may be summarised as follows.

Document (1) disclosed steps (a) to (c) of Claim 1 of the disputed patent and document (2) steps (f) to (m). It was argued that column 12 according to (1) fulfilled the same function as column 40 of the process according to the patent in suit. However, in the process of (1), the liquid in the sump of column 12 consisting of xenon, krypton and oxygen was recirculated to the sump of the low pressure column where concentration of Kr and Xe took place, the stream 19 according to (1) being equivalent to the stream 90 leaving column 40 in the process of the disputed patent. In other words, the provision of heat exchange 86 at the foot of the rare gas stripping column in the disputed patent, whereby a gaseous nitrogen stream taken from the high pressure main column was recirculated either to the high pressure or low pressure main column, was an obvious alternative to the provision in (1) of heat exchange at the foot of the low pressure main column, i.e. steps (d) and (e) of the claimed process. The Appellant referred to (3) to demonstrate that it was already known to introduce an indirect heat exchange into a Kr-Xe concentration column (Anreicherungskolonne).

The Appellant also argued that stream 17 according to document (1) was equivalent to stream 89 leaving the rare gas stripping column 40 of the patent in suit. Thus, most of the hydrocarbons contained in this feed air would leave with the oxygen in streams 17 and not be returned to the main column. In support of this argumentation, the Applicant pointed to column 3, lines 26-40 and column 4, lines 40-51 of (1).

VII. The Respondent, who is proprietor of the patent in suit, argued in response that the Appellant's submissions represented an oversimplification. The provision of heat exchange within the rare gas stripping column could not be regarded as the mere equivalent of heat exchange, with recycling of the oxygen streams, at the foot of the low pressure main column. The Respondent pointed to the increased concentration of Kr and Xe in the stripping column whilst, in contrast to the prior art, the hydrocarbon concentration was not increased. In addition, the provision of the heat exchange or bottom boiler in the stripping column was an additional burden and on economic grounds was not obvious.

The Respondent also argued that the process according to document (2) was not strictly equivalent to steps (f) to (m) of the claimed process. In particular, it was argued that there would be losses of Kr and Xe at stages 9 and 10 (supplementary condenser and separator respectively) of the process of (2).

The Respondent also requested that the Board should exclude document (3) from the proceedings since it appeared less relevant than the prior art already cited.

VIII. The Appellant requests that the decision of the Opposition Division be set aside and the patent revoked in its entirety.

The Respondent requests that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

2. The Board has considered whether it should disregard document (3) in accordance with Article 114(2) EPC. However, since (3) is a page from a well known chemical encyclopedia, it can be regarded as common general knowledge and, accordingly, the Board has decided to admit the document into the proceedings. This follows decision T 271/84 (O.J. EPO, 9/1987, 405, see Reasons, Point 3) which was referred to by the Respondent at the oral proceedings. A document indicating the scope of the common general knowledge of a skilled person in a special technical field cannot be disregarded as not submitted in due time because it only serves to confirm what is already known to the parties working in that field.

3. The patent in suit is concerned with an air separation process for the production of krypton and xenon.

3.1 The closest state of the art is considered to be document (1), which is also concerned with the concentration of krypton and xenon in an air separation process. According to (1), a gaseous stream of oxygen containing krypton and xenon is passed to a rectifying column where it is washed with a downward flow of liquid oxygen drawn from above the sump of the low pressure column of an air separation plant. The reflux in the rectifying

column causes gaseous oxygen together with some methane to leave the top of the column whilst a liquid mixture of oxygen together with krypton and xenon collects in the sump of the rectifying column. The said mixture is recycled to the sump of the low pressure main column where enrichment in Kr and Xe takes place. The concentrate from the said sump then passes to further concentration stages which are not specified (cf. column 5, lines 51-54).

3.2 In relation to the above prior art, the problem to be solved by the disputed patent is to provide an improved process for the concentration of Kr and Xe which avoids the safety hazard of hydrocarbon build up in contact with oxygen.

The problem is solved by providing heat exchange (or a bottom boiler) at the foot of the rectification or rare gas stripping column instead of the prior art recirculation to the main column and by using an additional countercurrent exchange with nitrogen.

Having regard to the data which appear in the Tables in the description of the patent in suit, the Board is satisfied that the problem is solved in a plausible manner.

4. None of the prior art cited discloses a process having the individual steps (a) to (m) set out in Claim 1 of the disputed patent; the process can accordingly be recognised as new. In any event, novelty has not been contested.

5. It remains to consider whether or not Claim 1 satisfies the requirements of Article 56 in respect of inventive step.

5.1 A comparison of Figure 1 of document (1) with the drawing of the patent in suit shows that both relate to closely similar processes. Both processes have a main air

separation column in which a lower high pressure column is linked to a low pressure column situated above, with heat exchange at the foot of the low pressure column. In both processes a gaseous stream of oxygen, krypton and xenon together with residual hydrocarbons is passed from the low pressure column to a rare gas stripping column which is also referred to in (1) as a separator. Thus, stream 11 according to (1) corresponds to stream 72 according to the disputed patent, both streams entering the respective columns below the bottom tray. Both processes also have a liquid oxygen stream entering above the top tray of the column shown as stream 15 in document (1) and stream 73 according to the disputed patent. In the process of the patent in suit, a stream of gaseous oxygen 89 leaves the top of the stripping column 40 and carries with it hydrocarbon impurities (column 5, lines 49-55). A similar gaseous oxygen stream 17 leaves the top of column 12 according to (1). As stated in column 4, lines 45-40 of (1), the reflux ratio in column 12 is adjusted so that the amount of methane leaving with the gaseous oxygen is equal to the amount entering the plant with the feed air, thus ensuring that the methane level in the liquid oxygen will stabilise. Having regard to the figures which appear in Table I of the patent in suit, it appears that similar conditions are established in stripping column 40; the figures for the hydrocarbon content of the liquid oxygen from the bottom tray 87 being essentially the same as the hydrocarbon content of the liquid oxygen stream 90 which leaves the bottom of the column. It is also to be noted that the reflux ratio of the column according to (1), i.e. 0.05 to 0.2 (column 4, lines 38-40) overlaps to a substantial extent the ratio 0.1-0.3 mentioned in column 6, lines 46-48 of the disputed patent. It is, accordingly, the opinion of the Board that the conditions established in column 12 of (1) are substantially the same as those established in column 40 according to the process of the

patent in suit. This was not disputed by the parties at the oral proceedings. Thus, the only difference between the said processes is that in accordance with the disputed patent a bottom reboiler is used to establish the reflux whilst, in accordance with document (1), the liquid oxygen stream is recycled to the sump of the main low pressure column. As indicated at column 4, lines 63-65 of (1), the amount of Kr and Xe increases as the plant continues to operate, the increase in rare gases corresponding to that which is recorded in Table I of the disputed patent relating to column 40. The Respondent has argued that the provision of heat exchange at the foot of column 40 provides an advantage in comparison with the process of document (1) insofar as it avoids a build-up of hydrocarbons in the main column. There is, however, no evidence of the extent of such a build-up available to the Board. Further, it does not appear, anyhow, once the steady state has been reached. Moreover, it is apparent from the disclosure of (1) that the inventor was aware of such a possibility but withdrew the liquid oxygen stream 19 containing Kr and Xe before any dangerous concentration of hydrocarbons could occur (cf. column 5, lines 46-51).

5.1.1 The Respondent has argued that the process according to the disputed patent gives a more efficient concentration of Kr and Xe at this stage than that known in the prior art. If the Respondent relies on such an advantage as evidence for inventive step, the burden is upon him to demonstrate such an effect. In the absence of such evidence, the Board can only conclude that the provision of a reboiler in column 40 of the process of the disputed patent is an obvious alternative to the recycling known from (1), without any apparent advantage. It has also been admitted that the provision of the said reboiler is an additional burden in terms of energy consumption. This could obviously not be

considered as an advantage in favour of the patent in suit.

5.1.2 It must also be added that the provision of heat exchange in a rare gas enrichment column by circulation of nitrogen from the high pressure column is also known from document (3). As was admitted by the Respondent at the oral proceedings, the "Anreicherungskolonne" according to (3) falls within the definition of "stripping column" which appears at column 3, lines 57-59 of the disputed patent.

5.1.3 In summary, it is the Board's view that the liquid stream (19) leaving the low pressure column of the process according to document (1) and the stream 90 leaving column 40 according to the process of the patent in suit can be regarded as obvious equivalents.

5.2 As indicated above (point 3.1), document (1) makes no precise reference to the further processing of stream 19, merely indicating further processing by known methods. Document (2), however, is concerned with exactly the same problem as that which is encountered in processing the stream 90, leaving column 40 in the process of the disputed patent which, according to Table I, contains, along with the Kr and Xe, 99.7% oxygen and 216 ppm hydrocarbons. Although document (2) is silent concerning the exact constitution of the streams, it is apparent that the hydrocarbon containing liquid oxygen stream 7 leaving the separator (Abscheider) (10) (cf. Figure 1 and Figure 2) is analogous to the said stream 90. It is recognised in (2) that such a mixture is potentially explosive (cf. column 1, lines 16-20). Thus, the problem to be solved according to (2) is to avoid the further processing of liquid oxygen which contains hydrocarbons (column 1, lines 35-38). The problem there is solved by passing the stream through a column against a counterflow of gaseous nitrogen. An

exchange takes place so that a nitrogen stream containing hydrocarbons leaves the bottom of the column, gaseous oxygen (containing some methane) leaving at the top (cf. column 1, lines 39-43). Document (2) makes no reference to the presence of Ar and Xe in the stream 7. It was, however, agreed by the parties at the oral proceedings that the said rare gases would be present in stream 7. The Respondent argued that the process according to document (2) was not analogous to that of the patent in suit since there would be losses of rare gases at the top of separation 11. The Board, however, accepts the counterargument of the Appellant that, owing to the low vapour pressure of Kr and Xe, the losses would be minimal. A comparison with the very small amounts of Kr and Xe leaving with the gaseous streams 89 and 104 from columns 40 and 15 respectively, according to the patent in suit, renders this counterargument plausible.

5.2.1 Accordingly, a man skilled in the art seeking a solution to the dangers of processing a liquid oxygen stream which contains hydrocarbons, such as stream 90 according the patent in suit, would, being aware of the disclosure of document (2), be led to seek a solution in replacing the oxygen with nitrogen using a countercurrent exchange.

5.3 From the preceding it follows that Claim 1 is not allowable under Article 56 since the subject-matter thereof represents an obvious combination of known processes.

6. Since Claim 1 falls, Claims 2-10, which represent preferred embodiments of the claimed process, must share the same fate.

Order

For these reasons, it is decided that:

1. **The decision under appeal is set aside.**

2. **European patent No. 0 096 610 is revoked.**

The Registrar: **The Chairman:**

M. Beer P. Lançon