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Bezeichnung der Erfindung: Separation of multicomponent gas mixtures

Title of invention:

Titre de l'invention :

Klassifikation / Classification / Classement : B01D 53/04

ENTSCHEIDUNG / DECISION

vom / of / du 9 April 1987

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /

Titulaire du brevet :

Air Products & Chemicals, Inc.
(Respondent)

Einsprechender / Opponent / Opposant : Linde AG (Appellant)

Stichwort / Headword / Référence :

EPO / EPC / CBE Article 56

Kennwort / Keyword / Mot clé : Inventive step (yes)

Leitsatz / Headnote / Sommaire

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Boards of Appeal

Office européen
des brevets

Chambres de recours



Case Number : T 155 /84

D E C I S I O N
of the Technical Board of Appeal 3.4.1
of 9 April 1987

Appellant :
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Decision under appeal : Decision of Opposition Division of the European
Patent Office dated 3 May 1984 rejecting
the opposition filed against European patent
No. 8512 pursuant to Article 102(2) EPC.

Composition of the Board :

Chairman : K. Lederer
Members : J. Roscoe
R. Schulte

Summary of Facts and Submissions

I. European patent No. 8512 was granted on 14 April 1982 with 22 claims in response to the European patent application No. 79 301 598.3 filed on 7 August 1979. The two independent Claims 1 and 21 read as follows:

1. In the separation of a multicomponent feed gas mixture with the individual recovery of a primary key component and a secondary key component present in such mixture, by selective sorption, wherein said secondary key component is more strongly sorbed than the primary key component and there is present in said mixture at least one tertiary component which is present in a concentration less than either said primary key component or said secondary key component and is less strongly sorbed than the secondary key component but more strongly sorbed than said primary key component; the method which is characterized in that it comprises, in an adiabatic adsorption pressure swing cycle the steps of:

(a) passing said multicomponent gas mixture at initial superatmospheric pressure and in selected flow direction through a first sorbent bed (A) selective for preferential retention of said secondary key component and then passing the effluent from said first sorbent bed through a second sorbent bed (B) selective for retention of said tertiary component as opposed to said primary key component, and discharging from said second sorbent bed unadsorbed primary key component, said passing of the multicomponent feed gas mixture being continued for a controlled time period until or short of breakthrough of said secondary key component from said first sorbent bed, while retaining all of the said tertiary component in said second sorbent bed;

(b) thereafter discontinuing gas flow communication between said first and second sorbent beds,

and

- (i) rinsing said first sorbent bed by flowing a stream of relatively pure secondary key component therethrough at substantially said initial superatmospheric pressure for a controlled time period effective to purge most of the void and displaced gases from the said first sorbent bed, and during this time period (b),
 - (ii) lowering the pressure in said second sorbent bed to an intermediate level by withdrawing a gas stream therefrom including void and desorbed gases, and thereafter;
 - (iii) further depressurizing said second sorbent bed to near ambient pressure
- (c) after said rinsing step in (b) above reducing the pressure in said first bed to an intermediate level by desorption of gas therefrom including previously sorbed secondary key component and during this step (c),
- (i) purging the second sorbent bed at near ambient pressure with a stream of primary key component;
 - (ii) repressurizing the second sorbent bed to an intermediate pressure level by flow thereinto of gas essentially free of secondary key component; and
 - (iii) repressurizing said second sorbent bed to about said superatmospheric pressure;
- (d) following step (c) above further desorbing gas from said first bed to lower the pressure therein to substantially ambient level, and thereafter;
- (e) evacuating said first bed to subatmospheric level;

- (f) after attaining the subatmospheric level in the said first bed, introducing therein a gas stream substantially free of the secondary key component to bring said first bed to an intermediate pressure level, and thereafter;
- (g) further repressurizing said first bed to initial superatmospheric feed pressure level by flowing therein primary key component via a second sorbent bed already pressurized to the intermediate pressure level (step c), thereby bringing both beds to the feed pressure level and making them ready to repeat the defined sequence of steps beginning with the reintroduction of the multicomponent feed gas mixture into the said first sorbent bed.

21. In the separation of a multicomponent feed gas mixture with the individual recovery of a primary key component and a secondary key component present in such mixture, by selective sorption, wherein said secondary key component is more strongly sorbed than the primary key component and there is present in said mixture at least one minor tertiary gas component less strongly sorbed than the secondary key component; the method which comprises effecting such selective sorption in a system comprising a plurality of first sorbent beds (A) operated in parallel in timed sequence and a plurality of second sorbent beds (B) operated in parallel in timed sequence, said first beds containing solid sorbent selective for preferential retention of said secondary key component and said second beds containing solid sorbent selective for retention of said tertiary component(s) as opposed to said primary key component; and wherein during a completed cycle each of said first beds (A) undergoes a successive sequence of steps comprising

- (a) passage of the multicomponent feed gas mixture there-
through at superatmospheric pressure in a selected
positive flow direction; with discharge therefrom of an
effluent gas stream composed of the unadsorbed portion
of the feed gas mixture;
- (b) discontinuing the passage of feed gas mixture into the
bed and discharge of effluent therefrom, followed by
rinsing of the bed at substantially initial feed gas
pressure level by flow therethrough of relatively pure
secondary key component;
- (c) after step (b) depressurizing the bed to an
intermediate pressure level by gas withdrawal
therefrom;
- (d) further withdrawing contained gas from said bed to
bring the bed to substantially ambient pressure level;
- (e) lowering the bed pressure to subatmospheric level by
evacuation;
- (f) then again bringing the bed to an intermediate pressure
level by introduction therein of a product gas
substantially free of secondary key component;
- (g) and finally bringing the bed to a superatmospheric feed
pressure level by flow thereinto of substantially pure
primary key component, and during the time period of
such complete cycle undergone by each first (A) bed,
each of the second (B) beds undergoes twice in
succession the steps of:

- (h) receiving the previously unadsorbed effluent portion of the initial feed gas mixture discharging from a first bed undergoing step (a) above, with discharge therefrom of an effluent gas stream composed of the unadsorbed portion of the primary key component of the feed gas mixture;
- (i) after discontinuing passage therein of said first bed effluent, lowering the pressure of such second (B) bed by transfer of gas therefrom into an evacuated first bed that has undergone step (e) above;
- (j) after step (i), further depressurizing such second bed by transfer of gas therefrom into another second bed which has undergone the purge step (l) below;
- (k) after step (j) further depressurizing such second bed to or near ambient pressure;
- (l) purging such second bed at or near ambient pressure with primary key component product;
- (m) bringing such second bed to an intermediate pressure level by introduction of desorbed gas into another second bed undergoing step (j);
- (n) and finally introducing substantially pure primary key component into such second bed to bring the bed to substantially feed gas pressure level.

In the above, certain obvious clerical errors appearing in the text of the claims of the granted patent have been corrected. Thus, in that text "intermediate" in feature c(ii) of Claim 1 reads "immediate", the article "a" appeared at the end of line 10 of Claim 21, and "solid" at line 13 of Claim 21 reads "said".

II. On 7 January 1983 the Opponents (Appellants) lodged an opposition against the European patent and requested that it be wholly revoked on the grounds that its subject-matter did not involve an inventive step having regard to:

DE-A-2 604 305 (1)
DE-A-2 258 299 (2)
US-A-3 703 068 (3)
and
DE-A-2 315 113 (4).

III. By a decision of 3 May 1984 the Opposition Division rejected the opposition.

IV. A notice of appeal was filed by the Appellants on 3 July 1984 against the decision of the Opposition Division, and the appeal fee duly paid. The Statement of Grounds submitted on 25 August 1984, relied on the documents (1) and (2) mentioned above and on the following additional documents:

DE-A-2 745 088 (5)
DE-B-1 259 844 (6)
FR-A-1 455 900 (7)
and
DE-B-1 919 557 (8).

Of these additional documents (7) had been cited in the European search report, but the remaining documents were completely new.

- V. In a letter filed on 25 September 1984 the patentee (respondent) drew attention to these additional documents, questioned the admissibility of their introduction into the appeal proceedings and sought a preliminary decision of the Board on this point.
- VI. Acting on behalf of the Board as then constituted the rapporteur, in a communication dated 7 December 1984 expressed the provisional opinion that since the documents in question were referred to in the grounds of appeal which were themselves filed within the time limit laid down in Article 108 EPC they had been submitted in due time within the meaning of Article 114(2) EPC and must therefore be admitted. Even if the Board were to deem them to have been late filed it would have to examine their relevance to the matter to be decided, that is to say whether the decision of the Opposition Division to maintain the patent in unamended form was correct in the light of all the facts available to the Board.
- VII. In the statement of grounds and at the oral proceedings held on 9 April 1987 the appellant argued essentially as follows:

Doc. (5) describes a pressure swing process which enables primary and secondary components to be recovered in pure form from a gas stream consisting of these two components. The purpose of the invention in the patent-in-suit was to recover such components from a gas stream containing in addition a tertiary component which is absorbed more readily than the primary but less readily than the secondary component.

The use of such a feed gas in the method of doc. (5) would result in a product stream consisting, instead of pure primary component, of a mixture of primary and tertiary components. It would be obvious for the skilled man to remove the tertiary component from this stream by an additional pressure swing process since processes for separation of several components from a gas stream by a succession of such processes were common and described for example in doc. (6). There, in each of two parallel processes, the air is dried in a first, and oxygen and nitrogen separated from the dried air in a second, pressure swing process. The separate removal of various components of a gas stream by use of series-connected adsorbers was a widely used process, described in doc. (2).

In the process claimed in Claim 1 of the patent-in-suit the two pressure swing processes operated largely independently of one another, a functional coupling between them existing only during adsorption, which was inevitable, and during the immediately preceding bed repressurizing step. In such a repressurizing step the use of gas which is not adsorbed was a widely used and described measure. The parallelism in time required between process steps in the first and second pressure swing processes was to a large extent uncritical and left to the free choice of the skilled man designing the process and thus not inventive.

As far as the steps followed in the first pressure swing process were concerned the differences between what was claimed and what happened in the apparatus of doc. (5) were trivial and obvious. Thus, it was obvious for the skilled man primarily interested in a good yield of secondary gas to dispense with the second rinsing with foreign gas disclosed in doc. (5). The two-stage pressurisation (features (f) and (g) of Claim 1) of a previously evacuated

first adsorber also involved nothing inventive since on the one hand in doc. (5) the single stage repressurisation was also effected with gas from a first sorbent bed which was not adsorbed in the first sorbent bed and on the other hand repressurisation in two or more stages in a pressure swing process was well known, see doc. (7). Furthermore steps (c) and (d) of Claim 1 were merely parts of a single continuous step such as performed in doc. (5). Thus such differences as there were from doc. (5) were within the compass of the skilled man.

The sequence of steps performed in the sorbent beds of the second pressure swing process was a conventional one described in doc. (7) with reference to Table I and elsewhere in the prior art, for example with reference to Fig. 7 in doc. (8).

The same considerations applied to Claim 21. The further requirement of this claim that the cycle time of the second beds be half that of the first beds was a consequence of the fact, not explicit in the claim, that there were twice as many first as second beds. In any system the skilled man knew he had to choose the adsorbing capacity of each stage in accordance with the amount of gas to be adsorbed, and that this could be done by using different sizes and/or numbers of beds in the two stages, which in the example of the patent had to adsorb secondary (20% of feed gas) and tertiary (4% of feed gas) component respectively.

VIII. The patentee disputed the arguments of the appellant. In his submission to omit the second rinsing, which was stated to be an essential feature in doc. (5) ran contrary to its teaching and could not therefore be obvious. With regard to the process of doc. (2) it was not seen how this equated to recovery of pure secondary key component and the relevance of doc. (6) was not apparent.

The arguments brought against the claims were based on *ex post facto* reasoning rather than on what was disclosed or suggested by the prior art relied on. The appellant had drawn steps at random from other citations to replace steps of the process of doc. (5) to arrive at the processes claimed without giving reasons as to why the skilled man should choose these in preference to others and had dismissed the ingenuity of the relative timing of the steps as "to a large extent non-critical".

- IX. The appellant (opponent) requests that the decision under appeal be set aside and the patent revoked.

- X. The respondent (patentee) requests that the appeal be dismissed and the patent maintained.

Reasons for the Decision

1. The appeal complies with the requirements of Articles 106 to 108 and Rule 64 EPC and is therefore admissible.

2. The claims and description being those of the application as originally filed and also of the granted patent no objection arises under Article 123(2) or (3) EPC.

3. After carefully considering the documents (5) to (8), first mentioned in the statement of grounds of appeal, and the arguments based thereon, pursuant to Article 114(1) EPC, using the powers conferred on it by Article 111(1) EPC, the Board concluded that even if it were to take this prior art into account it would not come to a different decision on the appeal. Having regard to this conclusion and bearing in mind that the appellant has not requested that the case be

remitted to the first instance for consideration of the relevance of these documents, the Board has decided to disregard them in accordance with Article 114(2) EPC as having not been submitted in due time. No further mention will therefore be made of them in this decision (cf. T 156/84, to be published).

4. Novelty

The Board finds, following a thorough examination of the documents (1) to (4) filed within the opposition period, that none of them discloses the subject-matter of either of the two independent Claims 1 and 21, and hence that of any of the claims. Hence the subject-matter claimed is novel. Since the appellant has not raised the issue of novelty either during the opposition itself or in the present appeal proceedings it is not deemed necessary to further discuss this matter as such.

5. Inventive step

It remains therefore to be determined whether the subject-matter of Claims 1 and 21 involves an inventive step over the prior art represented by docs. (1) to (4) and the routine knowledge of the skilled man.

- 5.1 The only one of these documents which discloses a process for individually recovering primary and secondary key components (in the examples hydrogen and carbon monoxide respectively) from a feed gas which additionally contains a tertiary component (in the examples methane) which is present in a lesser concentration and is sorbed more strongly than the primary but less strongly than the secondary key component is doc. (1). In the adsorption

phase of this process the feed gas flows through the first and second sorbent bed in series as in the process of Claims 1 and 21.

This process is therefore in the Board's opinion the prior art which comes closest to the processes claimed in Claims 1 and 21.

However, even the most relevant embodiment, that described with reference to Figs. 4 and 5, differs from the process of Claim 1 of the patent-in-suit in a number of material respects. Thus, whereas Claim 1 requires discontinuance of gas flow communication between the first and second beds (feature (b)) after completion of the adsorption phase, this phase is followed in the known process by purging of the still series-connected first and second beds with gas containing all three components, whereafter the beds remain connected during an expansion step in which the pressure is lowered from 20 to 3.4 bar. It is only then that the first sorbent bed is rinsed, and rinsing is not performed at the initial superatmospheric pressure but at a much lower pressure.

After this rinsing the pressure in the first bed is reduced (as it is in step (c) of Claim 1 of the patent-in-suit) but in a single continuous step to lowest pressure in the cycle, which may be subatmospheric rather than in two steps as in (c) and (e) of Claim 1. There is however, no repressurizing of the second sorbent bed during this lowering of pressure (cf. steps (c) (ii) and (iii) of Claim 1). Furthermore, there is no desorption of the gas from the first bed after the second sorbent bed is repressurised (step (d)). In fact the first and second beds are connected together and simultaneously repressurised in two stages generally equivalent to steps (f) and (g) of the claim.

- 5.2 From the benefits referred to on page 16 of the description of the patent-in-suit which are stated to flow from the invention it can be deduced that the complex of problems to be solved was how to improve the recovery rate and purity of both the primary and secondary key components, to obtain a tertiary product stream free of secondary key component and to improve operational efficiency and process economy.
- 5.3 That the claimed processes afford a solution to these problems has not been disputed by the appellant and appears plausible to the Board.
- 5.4 No contribution to inventive step is to be found in the recognition of these problems, since it is the constant endeavour of skilled practitioners in all technical fields to identify inefficiency and waste in processes and to seek to eliminate or reduce them, and it is clear that for many purposes purer products are desirable from gas separation methods.
- 5.5 Doc. (1) also sets out to solve at least some of the problems referred to above, e.g. recovery of two components from a multicomponent gas mixture with high purity and yield but arrives at a solution which is different in many respects as the above analysis shows.
- 5.6 The question to be answered is whether it would have been obvious at the priority date of the patent-in-suit for the skilled man, starting from the process described with reference to Figs. 4 and 5 of doc. (1) and confronted with these problems to modify it in such a way as to arrive at a process according to Claim 1, or Claim 21, of the patent-in-suit.

The Board is of the opinion that the answer to this question is no.

- 5.7 There is absolutely nothing in the other embodiments of doc. (1) itself to suggest to the skilled man that he should modify the steps of the method in such a way as to arrive at the process of Claim 1.
- 5.8 Doc. (3) is primarily concerned with a pressure swing adsorption process for separating a single component from a gas mixture in a single set of cyclically operated sorbent beds, and makes no provision for individual recovery of components from the residual gas. It therefore affords the skilled man no hint as to how the relative timing of the operations taking place in the first and second beds of doc. (1) might be altered. Moreover, the sequence of steps followed in the described processes do not correspond to that in either the first or the second bed of Claim 1 even if the interactions between the beds are left out of account.
- 5.9 Doc. (4) relates specifically to an adsorption process for the removal of sulphur compounds from gas streams and makes no provision for the recovery of an adsorbable component of a gas as such. The adsorbed component is in fact converted into elemental sulphur in part in the main beds where it is adsorbed and in part in auxiliary beds to which it is transferred. During the adsorption stage of the process the main and auxiliary beds are not connected in series and indeed at no stage does feed gas pass through an auxiliary bed. Therefore, since this process is directed to quite a different purpose than that of doc. (1) and the auxiliary (or second) set of beds plays a different role it cannot be seen why the skilled man looking to improve or modify the process of doc. (1) should seek guidance from it. Even if he were to do so he would not be led to modifications which would transform this process to that claimed in Claim 1.

Apart from the fact that the step (b) has no counterpart in doc. (4) it can be seen from TABLE I that there is no correspondence between the sequence of steps in the main and auxiliary beds with those performed in the first and second beds of Claim 1.

- 5.10 In the adsorption phase of the process described with reference to Fig. 3 of doc. (2) moist air flows through a first sorbent bed wherein its carbon dioxide and water content is adsorbed into a second sorbent bed in which nitrogen is selectively adsorbed. A flow of pure nitrogen is then pumped through the second bed to purge it of any adsorbed oxygen and then through the first bed which it purges of water and carbon dioxide. Finally with the first bed inactive the pressure in the second bed is lowered to desorb the nitrogen. None of the features of Claim 1 which are absent from the process of doc. (1) are to be found in this simple process which cannot therefore lead the skilled man to apply them to that known process.

Accordingly, the subject-matter of Claim 1 is not in the Board's view rendered obvious by any combination of the teachings of docs. (1) to (4). Claim 1 is therefore allowable.

- 5.11 Claims 2 to 20 relate to particular embodiments of the process of Claim 1 and are therefore likewise allowable.

6. Claim 21

- 6.1 The process described with reference to Figs. 4 and 5 of doc. (1) is in the Board's opinion also the closest prior art to the process claimed in Claim 21 as indicated above.

The claimed process differs from it in the following respects:

- (i) the rinsing of step (b) is performed with relatively pure secondary key component gas as opposed to gas containing only 35% of secondary key component (carbon monoxide);
- (ii) three successive steps (c) to (e) of reducing the pressure in the first bed to subatmospheric pressure replace two such steps separated by a second rinsing step at constant pressure with substantially pure secondary key component gas, a step which is stated in doc. (1) to further improve the purity and yield of this product;
- (iii) the pressure reduction in the second bed (step (i)) directly follows discontinuance of flow of first bed effluent into it, instead of being preceded by a rinsing of the bed at superatmospheric pressure with pure secondary key component or with essentially pure secondary key component;
- (iv) the step (j) has no counterpart in the known process where the second phase of depressurizing of the second bed is effected by evacuation;
- (v) the second beds each complete a full cycle of operations twice during the time taken for a first bed to complete a cycle.

6.2 The difference (i) involves no inventive step since in the general part of the description of doc. (1) dealing with a

variant corresponding to that described with reference to Figs. 4 and 5 it is suggested as an alternative to use product A (i.e. secondary key component) for such rinsing.

There is however, nothing in the document which could incite the skilled man to so modify the process as to eliminate the remaining differences.

- 6.3 The purpose of the beds in doc. (4) and the cycles followed in each set of beds are so radically different from those of the respective beds in doc. (1) that the skilled man would not in the Board's opinion consider this document relevant to the process described in that document. Even were he to do so he would find no teaching of a three-phase pressure reduction in either set of beds nor of transfer of depressurizing gas from one second bed to any other (see differences (ii) and (iv) in item 6.1). Furthermore since the first and second beds do not adsorb different gas components as they do in doc. (1) use of half the number of second beds cycling twice as fast as the first beds provides the skilled man with no motivation to adopt the same arrangement, for which no reason is given, in the process of doc. (1).
- 6.4 In doc. (2) the first beds are not subjected to any pressure reduction steps after the rinsing step, and there is no depressurizing of a second bed by transfer of gas to another second bed. Therefore no combination of the teaching of this document with that of doc. (1) could lead the skilled man to the process claimed in Claim 21.
- 6.5 Turning finally to doc. (3) this relates to a process using only a single set of beds and cannot therefore lead the skilled man to the employment of different numbers of first

and second beds in the doc. (1) process. Although the pressure reduction in the beds, which directly follows discontinuance of the flow of feed gas through them is, in the embodiment of Fig. 5, a three-stage process it terminates at atmospheric pressure and does not therefore correspond to that in the first beds of Claim 1 and it lacks step (i), which involves interaction between a second and a first bed and more importantly also step (j), also missing from docs. (1), (2) and (4), since the depressurizing gas is actually used to purge another second bed which has not been purged with primary key component or anything else.

- 6.6 From the above discussion it emerges that even by selectively combining features from docs. (1) to (4) the skilled man could not arrive at the process claimed in Claim 21. The Board is therefore satisfied that the subject-matter of this claim involves an inventive step, and the claim is thus allowable.
- 6.7 Claim 22 relates to a particular embodiment of the process of Claim 21 and is likewise allowable.

Order

For these reasons, it is decided that:

The appeal against the decision of the Opposition Division is dismissed.

The Registrar:

F. Klein

The Chairman

K. Lederer