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BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

Publication in the Official Journal ~~Yes~~ / No

File Number: T 180/90 - 3.2.3

Application No.: 86 309 845.5

Publication No.: 0230754

Title of invention: Separation of gaseous mixtures

Classification: F25J3/02; C07C7/04; C07C9/04

D E C I S I O N
of 12 September 1991

Proprietor of the patent: The BOC Group plc

Headword: The BOC Group/separation of gaseous mixtures

EPC Article 56

Keyword: "Inventive step (main request: no, auxiliary request: yes)"

Headnote

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number : T 180/90 - 3.2.3

D E C I S I O N
of the Technical Board of Appeal 3.2.3
of 12 September 1991

Appellant : The BOC Group plc
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Representative : Wickham, Michael
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Decision under appeal : Decision of Examining Division of the European Patent Office dated 09.10.89 refusing European patent application No. 86 309 845.5 pursuant to Article 97(1) EPC.

Composition of the Board :

Chairman : C.T. Wilson
Members : H. Andrae
J.-C. Saisset

Summary of Facts and Submissions

I. European patent application No. 86 309 845.5 filed on 17 December 1986 and published on 5 August 1987, was refused by a decision of the Examining Division dated 9 October 1989.

II. The decision was based on Claims 1 to 8 filed on 8 February 1989.

The reason given for the refusal was that the subject-matter of Claim 1 did not involve an inventive step having regard to the prior art disclosed in the document US-A-2 888 807.

III. On 18 December 1989 the Appellant lodged an appeal against the decision. The appeal fee was duly paid and the statement of grounds was received on 16 February 1990 by telecopy, confirmed by letter received on 18 February 1990. The Appellant argued that the features of Claim 1 were nowhere suggested in the citation and maintained that Claim 1 defines an inventive step.

IV. In a communication dated 24 January 1991 the Board expressed a provisional opinion stating that the subject-matter of Claim 1 was novel but appeared to lack an inventive step when taking into account the disclosure of US-A-2 888 807. The Board considered, however, that a new independent Claim 1 comprising the subject-matter of Claims 1 and 5 of the published European patent application would appear to meet the requirements of the EPC.

V. In response to the communication of the Board, the Appellant requested with a letter filed on 23 March 1991 that a patent should be granted on the basis of the

claims underlying the contested decision (main request). As an auxiliary submission, he requested that a patent be granted on the basis of Claims 1 to 7 filed with that letter.

VI. After discussions by telephone dated 18 April and 24 June 1991 between the Appellant and the Rapporteur concerning amendments to the description, the Appellant requests grant of a patent based on Claims 1 to 8 as filed on 8 February 1989, auxiliarily based on Claims 1 to 7 filed on 18 May 1991.

VII. In his written statements, the Appellant put forward the following arguments in support of the main request:

- US-A-2 888 807 contains false thermodynamic data, inconsistencies and lacunae in its specific teaching. If the skilled person did build up the distillation columns and then found them icing up, the logical approach would be to add to the feedstock an inhibitor of the deposition of solid carbon dioxide. If the skilled person did build a plant to operate the process disclosed in US-A-2 888 807 and then found that the distillation columns were icing up under operating conditions that should according to the citation exclude formation of solid carbon dioxide, it would not be a simple matter to adjust the operation of the plant to obtain the desired separation while avoiding such icing-up.

- The teaching in the citation, column 3, lines 23 to 26, is a recommendation to keep the operating pressure as near as possible to 673 psia and hence the bottom carbon dioxide concentration to around 30% by volume of carbon dioxide. Nowhere is it suggested that the level of carbon dioxide should be kept below 12% CO_2.

VIII. The texts of the independent Claims 1 according to the main request and the auxiliary requests, respectively, are as follows:

Main request:

"A method for separating a mixture comprising carbon dioxide and methane, including the steps of feeding said mixture into a first liquid-vapour contact column, performing a first rectification of the mixture at a lower pressure in the first liquid-vapour contact column to form a product vapour fraction containing a reduced concentration of carbon dioxide at the top of the column and a fraction at the bottom of the column which is enriched in carbon dioxide and which contains up to 12% by volume of carbon dioxide, subjecting the said carbon dioxide-enriched fraction to a second rectification in a second liquid-vapour contact column operating at a higher pressure than said first column and thereby forming at the bottom of the second column a liquid fraction in which carbon dioxide predominates, and at the top of the second column a vapour fraction containing a lower proportion of carbon dioxide than the said carbon dioxide enriched fraction, and returning fluid from at or near the top of the second column to the first column, the concentrations of carbon dioxide in the said gaseous mixture and in the said carbon dioxide enriched fraction, and the operating pressures in the columns being selected so as to avoid the deposition of solid carbon dioxide."

Auxiliary request:

"A method for separating a mixture comprising carbon dioxide and methane, including the steps of feeding said mixture into a first liquid-vapour contact column, performing a first rectification of the mixture at a

lower pressure in the first liquid-vapour contact column to form a product vapour fraction containing a reduced concentration of carbon dioxide at the top of the column and a fraction at the bottom of the column which is enriched in carbon dioxide and which contains up to 12% by volume of carbon dioxide, subjecting the said carbon dioxide-enriched fraction to a second rectification in a second liquid-vapour contact column operating at a higher pressure than said first column and thereby forming at the bottom of the second column a liquid fraction in which carbon dioxide predominates, and at the top of the second column a vapour fraction containing a lower proportion of carbon dioxide than the said carbon dioxide enriched fraction, and returning fluid from at or near the top of the second column to the first column, the concentrations of carbon dioxide in the said gaseous mixture and in the said carbon dioxide-enriched fraction, and the operating pressures in the columns being selected so as to avoid the deposition of solid carbon dioxide, wherein said fluid from at or near the top of the second column is taken, is adjusted in temperature to avoid deposition of solid carbon dioxide at the operating pressure of the first column, and is then throttled upstream of where it is returned to the first column."

Reasons for the Decision

1. The appeal is admissible.

2. Main request

2.1 Amendments

2.1.1 Claim 1 is based upon originally filed Claim 1. The further feature in Claim 1 that the carbon dioxide

enriched fraction at the bottom of the first lower pressure column contains up to 12% by volume of carbon dioxide, has been disclosed in the originally filed description on page 5, lines 7 to 17, and page 9, paragraph 2.

2.1.2 Dependent Claims 2 to 8 correspond with originally filed Claims 2 to 8, respectively.

All claims of the main request are therefore acceptable under Article 123(2) EPC.

2.2 Novelty

After examination of the citations revealed by the search report, the Board is satisfied that none of them discloses a method for separating a mixture comprising carbon dioxide and methane including all the features stated in Claim 1. Since this has never been disputed and the claim is unallowable on other grounds anyway, there is no need for a detailed substantiation of the question of novelty. Therefore, the subject-matter as set forth in Claim 1, is novel (Article 54 EPC).

2.3 Inventive step

2.3.1 There is agreement between the Appellant and the Board that US-A-2 888 807 reflects the most relevant prior art.

This document discloses a method for separating a mixture comprising carbon dioxide and methane, including the steps of feeding said mixture into a first liquid-vapour contact column (A) performing a first rectification of

the mixture at a lower pressure (673 psia) in the first liquid-vapour contact column to form a product vapour fraction containing a reduced concentration of carbon dioxide (0% CO_2) at the top of the column and a fraction at the bottom of the column which is enriched in carbon dioxide (30% CO_2), subjecting the said carbon dioxide-enriched fraction to a second rectification in a second liquid-vapour contact column (B) operating at a higher pressure (715 psia) than said first column and thereby forming at the bottom of the second column a liquid fraction in which carbon dioxide predominates (100% CO_2), and at the top of the second column a vapour fraction containing a lower proportion of carbon dioxide (5% CO_2) than the said carbon dioxide enriched fraction, and returning fluid from at or near the top of the second column to the first column, the concentrations of carbon dioxide in the said gaseous mixture and in the said carbon dioxide-enriched fraction, and the operating pressures in the columns being selected so as to avoid the deposition of solid carbon dioxide (cf. figure 1 and the associated description).

Claim 1 differs from this method in that the fraction at the bottom of the first column, enriched in carbon dioxide, contains up to 12% by volume of carbon dioxide.

2.3.2 According to the Appellant all the distillation processes proposed to date are for very low carbon dioxide concentrations, e.g. less than 1%, or for higher carbon dioxide concentrations, e.g. greater than 20%, or necessitate the use of an additive. There is thus a need for an improved process and apparatus for the separation of a methane-carbon dioxide mixture containing up to about 12% by volume of carbon dioxide, and the invention aims at meeting this need (see original description page 3, paragraphs 2 and 3).

The inherent problem may therefore be seen in proposing a distillation process for a mixture of methane and carbon dioxide which is for carbon dioxide concentrations in an intermediate range. An additional aspect is, as the Appellant has set out in his letter dated 21 March 1991, to avoid the formation of solid carbon dioxide or "icing-up" of the distillation columns.

Starting out from a process such as is known from the relevant prior art disclosed in US-A-2 888 807 in which the product fraction at the bottom of the first column contains 30% carbon dioxide, the inherent problem of the invention is solved according to Claim 1 by modifying the known process such that the product fraction at the bottom of the first column contains up to 12% by volume of carbon dioxide.

2.3.3 Since the prior art does not refer to distillation processes for the separation of a methane-carbon dioxide mixture for carbon-dioxide concentrations in the above-cited intermediate range, the need to cover also this range of carbon dioxide concentrations arises from the particular requirements of a distillation process such as the composition of the feed gas and the distillation product gas. The skilled person being aware of the known distillation processes will immediately recognise that the prior art does not refer to processes in the range of carbon dioxides up to about 12% and will be confronted under the requirements of a particular distillation process with the problem of arriving at a process which is perfectly appropriate in the cited intermediate range of carbon dioxide concentrations. The perception of the problem must therefore be regarded as an obvious consequence of the information gaps identifiable in the prior art.

2.3.4 The skilled person would be expected to investigate the relevant prior art, i.e. US-A-2 888 807 with regard to the question whether it was possible to modify the known process such that it yields carbon dioxide enriched fractions in the cited intermediate range.

In this known process, the first column (A, Figure 1) operates at a pressure of 673 psia. The phase diagram for methane-carbon dioxide mixtures at this pressure, as illustrated in Figure 3 of the citation, shows the vapour-liquid triangle defined by points "b", "c" and "d". Column 3, lines 27 to 49, of the citation indicates that the carbon dioxide concentration in the first column must stay within the vapour-liquid triangle in order to avoid the formation of solid carbon dioxide. According to the diagram illustrated in Figure 3, the value of maximum concentration of carbon dioxide without formation of solid carbon dioxide is approximately 30%. In column 3, lines 14 to 26, of the citation it is pointed out that, if the pressure of the first tower (A) is reduced substantially below 673 psia, solid carbon dioxide will form in the tower unless the concentration of carbon dioxide in the feed stream is similarly decreased.

This suggests that the first distillation column may also be operated at a pressure substantially lower than 673 psia. The skilled person is therefore provided with the incentive to choose a lower column operating pressure in the first column. He may decide to choose an operating pressure in the first column of 41.4 atmospheres absolute which in a particular case depending on the design conditions is most appropriate. It arises from the phase diagram for methane-carbon dioxide mixtures under this pressure (cf. the vapour-liquid triangle "A", "B", "C" in Figure 2 of the drawings of the present application),

that the maximum concentration of carbon dioxide without formation of solid carbon dioxide is about 12% (point "C").

In analogy to the above-cited maximum concentration of 30% shown in Figure 3 of US-A-2 888 807, the skilled person having decided to choose an operating pressure of 41,4 atmospheres absolute in the first column, will have to operate the distillation apparatus, in order to avoid the formation of solid carbon dioxide, such that the carbon dioxide enriched fraction in the first column does not contain carbon dioxide in excess of 12%.

Hence, US-A-2 888 807 provides the skilled person with the decisive information that operation of a distillation apparatus under a comparatively lower pressure in the first column is feasible and it remains only as a routine work to consult the corresponding phase diagram in order to obtain the maximum permissible concentration of carbon dioxide without deposition of solid carbon dioxide.

2.3.5 Having regard to the Appellant's argument that US-A-2 888 807 contains false thermodynamic data, inconsistencies and lacunae in its specific teaching whereby the skilled person would be led to reject the teaching of the citation, it is to be noted that the skilled person intending to try the known process experimentally would not rely solely on the thermodynamic data disclosed in the citation but would for the reason of avoiding expensive failures check the data given in the citation by means of the phase diagrams for methane-carbon dioxide mixtures which form part of his basic design material. He would then immediately become aware of any discrepancies in the basic thermodynamic data and would rely on the data consolidated in the art.

There would be no reason for rejecting the teaching of the citations altogether. In particular, the skilled person would not ignore the hint at operating the distillation process with a reduced pressure in the first column and a correspondingly decreased concentration of carbon dioxide in the feed stream because it could be seen immediately from the available phase diagrams that also under such modified conditions operation of a distillation apparatus without the risk of formation of solid carbon dioxide is possible.

2.3.6 For these reasons the Board comes to the conclusion that the subject-matter of Claim 1 according to the main request is arrived at from the cited prior art without an inventive step being involved and Claim 1 consequently cannot be regarded as patentable (Articles 52(1) and 56 EPC).

The main request of the Appellant cannot, therefore, be allowed.

3. Auxiliary request

3.1 Amendments

3.1.1 As compared to Claim 1 according to the main request, Claim 1 according to the auxiliary request has been limited to the further steps that the fluid from at or near the top of the second column is taken, is adjusted in temperature to avoid deposition of solid carbon dioxide at the operating pressure of the first column, and is then throttled upstream of where it is returned to the first column.

These further steps have been disclosed in originally filed Claim 5.

3.1.2 Dependent Claims 2 to 7 are based on originally filed Claims 2 to 4 and 6 to 8, respectively.

Having regard to Claim 2, the amended value of about 41.4 atmospheres absolute (4.2 MPa) of the operating pressure in the lower pressure column has been disclosed in the originally filed description on page 5, second paragraph, and on page 7, lines 10 and 11.

3.1.3 The description has been adapted to the present set of claims in accordance with Rule 27(1)(c) and (d) EPC. Furthermore, an obvious error on page 9, paragraph 2, of the present description has been corrected by eliminating an unclear phrase, cf. the sentence spanning lines 15 and 16 on page 10 of the originally filed description.

3.1.4 The claims and the description are not, therefore, objectionable under Article 123(2) EPC.

3.1.5 The one-part form of the independent claim is considered to be appropriate in the present case. The closest prior art document discloses several of the process steps claimed in the invention such that drafting of the claim in the two-part form according to Rule 29(1) EPC would render the claim lengthy and complex.

3.2 Novelty

The reasons with regard to novelty of Claim 1 according to the main request apply basically also to Claim 1 according to the auxiliary request which has been limited to further process steps (cf. point 3.1.1 above). Therefore, the subject-matter as set forth in Claim 1 is novel (Article 54 EPC).

3.3 Inventive step

3.3.1 In the process for separating a mixture comprising carbon dioxide and methane known from US-A-2 888 807 the product fraction withdrawn from the top of the second distillation column is throttled in a throttling valve. There is a risk that throttling of the product fraction leaving the top of the second distillation column may cause the fluid to enter a state in the phase diagram in which a solid phase of carbon dioxide occurs.

3.3.2 In the light of the relevant prior art disclosed in US-A-2 888 807 the problem underlying the invention may therefore be seen in amending the known process such that in the fluid return duct leading from the top of the second liquid-vapour contact column to the first column deposition of solid carbon dioxide is avoided.

This problem is credibly solved by the features according to Claim 1. In particular, the step of adjusting the temperature of the fluid taken from at or near the top of the second column before throttling the fluid leads to a phase change of the fluid which can be effected by means of heat supply or heat removal such that the fluid after being subjected to the pressure decrease due to throttling arrives at a temperature which safeguards a sufficient distance from the region of solid deposition in the liquid-vapour phase diagram.

3.3.3 US-A-2 888 807 deals with the problem of avoiding the formation of solid carbon dioxide by means of a chilling process, cf. column 5, lines 53 to 75 of the citation. In this passage, the citation discloses essentially that if the feed gas can be chilled by throttling from a higher

pressure down to 673 psia, solid carbon dioxide will not form on the feed plates of tower A (corresponding to the first liquid-vapour contact column according to the invention), even though the carbon dioxide concentration exceeds 8%, providing the concentration remains less than the concentration of the bottom product.

The skilled person is provided hereby with the information generalised with regard to the specific values disclosed that deposition of solid carbon dioxide at the inlet of the distillation tower can be prevented by means of chilling the feed gas by throttling from a higher pressure.

In contrast to this information, Claim 1 of the invention teaches to effect the step of adjusting the fluid taken from at or near the top of the second column, in temperature such as to avoid deposition of solid carbon dioxide at the operating pressure of the first column, and the subsequent step of throttling the fluid upstream of where it is returned to the first column. As compared to the teaching of the citation Claim 1 proposes thus the additional measure of adjusting the fluid temperature before the throttling operating and provides also information as to the manner and extent of effecting this measure, namely such that deposition of carbon dioxide is avoided at the operating pressure of the first column. Since the process disclosed in US-A-2 888 807 does not provide such an additional measure, it cannot suggest the subject-matter of Claim 1.

3.3.4 The further document DE-A-3 510 097 cited in the search report does not constitute part of the prior art in the sense of Article 54(2) and (3) EPC since the claim of the

priority date of the European patent application is justified, all features of the valid claims being disclosed in the priority documents of the application.

3.3.5 Summarising, the Board comes to the conclusion that the subject-matter of Claim 1 according to the auxiliary request is not suggested by the prior art and must therefore be regarded as involving an inventive step (Article 56 EPC).

The same applies to dependent Claims 2 to 7 which concern particular embodiments of the invention according to Claim 1.

Order

For these reasons, it is decided:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to grant a patent on the basis of the following documents:

Claims: 1 to 7 filed on 18 May 1991

Description: Pages 1 to 4, 7, 8 and 11 filed on 18 May 1991;
pages 5, 6, 9 and 10 filed on 23 August 1991.

Drawings: Sheets 1/5 to 5/5 filed on 10 March 1987.

The Registrar:

N. Maslin

The Chairman:

C.T. Wilson

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