

**Internal distribution code:**

- (A)  Publication in OJ  
(B)  To Chairmen and Members  
(C)  To Chairmen  
(D)  No distribution

**D E C I S I O N**  
**of 22 June 2005**

**Case Number:** T 0825/02 - 3.3.5

**Application Number:** 98110012.6

**Publication Number:** 0908219

**IPC:** B01D 53/22

**Language of the proceedings:** EN

**Title of invention:**

Multi-storage process for the separation/recovery of gases

**Patentee:**

GKSS-Forschungszentrum Geesthacht GmbH, et al

**Opponent:**

Membrane Technology and Research, Inc.

**Headword:**

Membranes/GKSS-PETROBRAS

**Relevant legal provisions:**

EPC Art. 54(1), (2), 111(1)

**Keyword:**

"Novelty - main request (no), implicit features"  
"Auxiliary request (yes)"

**Decisions cited:**

-

**Catchword:**

-



Case Number: T 0825/02 - 3.3.5

**D E C I S I O N**  
**of the Technical Board of Appeal 3.3.5**  
**of 22 June 2005**

**Appellants:** GKSS-Forschungszentrum Geesthacht GmbH  
(Proprietors of the patent) Max-Planck-Strasse  
D-21502 Geesthacht (DE)

PETROLEO BRASILEIRO S.A. - PETROBRAS  
Cidade Universitária - Quadra 7  
Ilha do Fundão  
BR-21949-900 Rio de Janeiro (BR)

**Representative:** Grebner, Christian, Dr.  
Patentanwälte  
Seemann & Partner  
Ballindamm 3  
D-20095 Hamburg (DE)

**Respondent:** Membrane Technology and Research, Inc.  
(Opponent) 1360 Willow road  
Menlo Park, CA 94025 (US)

**Representative:** Setna, Rohan P.  
Boult Wade Tennant  
Verulam Gardens  
70 Gray's Inn Road  
London WC1X 8BT (GB)

**Decision under appeal:** Decision of the Opposition Division of the  
European Patent Office posted 11 June 2002  
revoking European patent No. 0908219 pursuant  
to Article 102(1) EPC.

**Composition of the Board:**

**Chairman:** M. M. Eberhard  
**Members:** J. D. Schwaller  
H. Preglau

## Summary of Facts and Submissions

- I. This appeal lies from the decision of the opposition division revoking European patent No. 0908219 on the grounds that it does not comply with the requirements of novelty set out in Article 100(a) EPC.
- II. The opposition division held that the subject-matter of claim 1 of the main request (claim 1 as granted) lacked novelty over D2: JP-A-1043329 whereas claim 2 of the main request (claim 2 as granted) lacked novelty over D1: "The behaviour of a Membrane-Membrane Hybrid", D.L. Roberts, Proceedings of the North American Membrane Society, 6<sup>th</sup> Annual Meeting 1994. The auxiliary request was rejected because the subject-matter of claim 1 was considered to be anticipated by D2.
- III. During the appeal proceedings, the appellants (proprietors) filed several sets of amended claims as auxiliary requests. In particular, with a letter dated 23 May 2005, they submitted three new sets of claims as auxiliary requests I to III and on 20 June 2005, two additional sets of claims as auxiliary requests IV and V, each of these requests I to V comprising only two independent claims.
- IV. During the oral proceedings which took place on 22 June 2005, the appellants withdrew their main request and filed three complete sets of amended claims as a main request and two auxiliary requests I and II.

Claims 1 and 2 of the main request read as follows (the *bold characters have been added by the board to identify the features not present in claims 1 and 2 as*

*granted; the features deleted from claims 1 and 2 as granted have been crossed out)*

1. Process for the ~~separation and/or~~ recovery of gases **as process products** from gas and/or gas vapour mixtures by means of a membrane separating device to which the gas and/or gas vapour mixture is supplied, characterised by a multi-stage, simultaneous separation of such gas and/or gas vapour mixtures in the membrane separating device, by two different membrane types, one type being at least one organophilic membrane, the at least one such membrane unit separating the gas and/or gas vapour mixture into a pressure-reduced permeate, **recovered as and being a first process product**, enriched with desired condensable gas(es)/gas vapours and a retentate enriched with desired gas(es) of small kinetic diameter at essentially the same pressure set at the inlet end of the organophilic membrane unit(s), combined with at least one glassy membrane, the at least one such glassy membrane unit separating the retentate from the organophilic membrane unit(s) in the membrane separating device into a pressure-reduced permeate, **recovered as and being a second process product**, enriched with the desired gas(es) of small kinetic diameter and a retentate enriched with such desired condensable gas(es)/ vapours at essentially the same pressure set at the inlet end of the glassy membrane unit(s).
  
2. Process for the ~~separation and/or~~ recovery of gases **as process products** from gas and/or gas vapour mixtures by means of a membrane separating device to which the gas and/or gas vapour mixture is supplied, characterised by a multi-stage, simultaneous separation of such gas

and/or gas vapour mixtures in the membrane separating device, by two different membrane types, one type being at least one glassy membrane, the at least one such membrane unit separating the gas and/or gas vapour mixture into a pressure-reduced permeate, **recovered as and being a first process product**, enriched with desired gas(es) of small kinetic diameter and a retentate enriched with desired condensable gas(es)/vapours at essentially the same pressure set at the inlet end of the membrane unit combined with at least one organophilic membrane, the at least one such organophilic membrane unit separating the retentate from the glassy membrane unit(s) in the membrane separating device into a pressure-reduced permeate, **recovered as and being a second process product**, enriched with the desired condensable gas(es)/vapours and a retentate enriched with such desired gas(es) of small kinetic diameter at essentially the same pressure set at the inlet end of the organophilic membrane unit(s).

Claims 1 and 2 of the 1<sup>st</sup> auxiliary request correspond to the claims of the main request with the following additional features: "said condensable gas(es)/gas vapours being C<sub>2</sub> and higher hydrocarbons" and "said gases of small kinetic diameter being H<sub>2</sub> or He".

- V. The appellants requested that the decision under appeal be set aside and that the patent be maintained on the basis of the main request or alternatively on the basis of one of auxiliary requests I or II filed during the oral proceedings or of the auxiliary requests III or IV, originally filed by fax on 20 June 2005 as auxiliary requests IV and V.

The respondent requested that the appeal be dismissed. In case the board would consider the subject-matter of one set of claims as novel, he requested that the case be remitted to the opposition division for further prosecution.

VI. The appellants presented mainly the following arguments:

VI.1 The features "recovered as and being a first process product" and "recovered as and being a second process product" appearing in claims 1 and 2 of the main request were disclosed respectively in paragraphs [0001], [0007] with respect to the word "recovery" and in paragraphs [0015], [0020], [0026], [0031] and [0036] of the patent in suit as regards the first and second process products. They furthermore stated that the said first and second process products corresponded to the permeates of the first and second membranes, respectively.

VI.2 With respect to novelty of claim 1, they stressed that the permeates obtained in both the first and second membranes of the separation process disclosed in D2 could not be recovered as process products because they were supposed to be discarded or burnt. The appellants further were of the opinion that in the absence of any indication as to the pressure at the feed inlet of the second membrane in D2, it was speculative to suppose that it was essentially the same as that of the retentate of the second membrane.

VI.3 As to the novelty of the subject-matter of claim 2 according to the main request, the appellants submitted

that in the air/toluene separation process of D1, the air recovered as the permeate of the first membrane was rejected to the atmosphere and therefore could not be considered as being recovered as a process product. They also argued that the retentate exiting the second membrane and recycled to the inlet of the first membrane was recompressed by means of a compressor. Thus the pressure of said retentate could not be substantially the same as the pressure set at the inlet of the second membrane. They finally stated that the expression "at essentially the same pressure" stated in claims 1 and 2 would be interpreted by the skilled person as meaning  $\pm 10 \%$ .

VI.4 With respect to claims 1 and 2 of the 1<sup>st</sup> auxiliary request, they argued that neither D1 nor D2 disclosed that the feed to the first membrane may be C<sub>2</sub> and higher hydrocarbons. Furthermore, D1 was silent about the presence of H<sub>2</sub> or He in the feed stream to the separation process.

VII. The respondent (opponent) essentially argued as follows:

The permeates from the first and second separation membranes used in D1 and D2, whether they were desired or not, were in any case process products and recovered as such. The silicone rubber membranes used e.g. in D2 have intrinsic properties belonging to the general knowledge, one of these properties being to reject hydrogen which thus remains in the retentate. This was confirmed by D17 = US-A-5755855 (Example 1 and Table 2) and the patent in suit, both documents using this kind of membrane for the same purpose. Since air inherently comprises helium and hydrogen, the permeate of the

first membrane of D1 necessarily comprised these gases. The C<sub>2</sub> and higher hydrocarbons were explicitly disclosed in the Table of D2 and hydrogen was considered as technically equivalent to CO<sub>2</sub> in this document.

Therefore the subject-matter of claim 1 of each of the main and 1<sup>st</sup> auxiliary requests was anticipated by D2 and claim 2 of each of the main and 1<sup>st</sup> auxiliary requests was not novel over D1.

### **Reasons for the Decision**

1. The appeal is admissible
2. *Main request*
  - 2.1 Allowability of the amendments under Article 123 EPC

The appellants having acknowledged at the oral proceedings that the first and second process products defined in amended claims 1 and 2 were nothing else than the permeates of the first and second membranes, respectively, the respondent had no objection against the allowability of this request.

The amended features in claims 1 and 2, in particular the expressions "recovered as and being a first product process" and "recovered as and being a second product process" have a basis in the following passages of the application as originally filed: page 1, first paragraph; page 3, third paragraph; page 5, third paragraph; page 6, fifth paragraph; page 9, second paragraph and page 10, last paragraph. Dependent



claims 3 to 24 being unamended and corresponding to claims 3 to 24 as originally filed, the requirements of Article 123(2) EPC are therefore fulfilled with respect to claims 1 to 24.

The restriction of the expression "separation and/or recovery" to "recovery" and the above-mentioned additional amendments to claims 1 and 2 do not give rise to an extension of the scope of protection, thus no objection under Article 123(3) EPC arises.

## 2.2 Novelty

2.2.1 As a preliminary remark, it is noted that the claims contain the expressions "gas(es) of small kinetic diameter", "condensable gas(es)/vapours" and "at essentially the same pressure". In the absence of well-recognized technical meaning of these expressions and of definitions thereof in the patent in suit, they must be interpreted in the broadest way. In this connection, the appellants have never disputed that "air" is encompassed by the expression "gas(es) of small kinetic diameter" and similarly "toluene" by "condensable gas(es)/ vapours". They nevertheless contended that the expression "at essentially the same pressure" would mean  $\pm 10 \%$ ; the board notes in this respect that this assertion is not supported by any value in the examples of the patent in suit or any other information therein. In these circumstances, this ambiguous expression cannot be interpreted as being limited to variations of 10 % or less and has therefore to be construed broadly, the limits for the pressure difference defined in the claims being in fact unclear.

2.2.2 It was argued that D1 destroyed the novelty of the process as defined in claim 2. In fact, D1 (page 2, 2<sup>nd</sup> full paragraph; Figure 1) discloses a process for the recovery of toluene and air from a toluene/air gaseous mixture by means of two different membranes,

- (i) the first one being an air-selective membrane based on perfluorodimethyl dioxole-tetrafluoroethylene (PDD-TFE) polymers, said membrane being of the glassy type (see paragraph bridging pages 1 and 2) and separating the binary mixture into a pressure-reduced (15 psia) permeate A enriched in air and a retentate enriched with toluene, the pressure at the inlet end of the first membrane unit being 22.5 psia, combined with
  
- (ii) a second membrane, namely an organic-selective membrane based on a silicone rubber polymer, separating the retentate from the glassy membrane unit into a pressure-reduced (7.5 psia) permeate B enriched with toluene and a retentate enriched with air, the retentate from the second membrane being recycled to the feed inlet of the first membrane.

2.2.3 It is observed that neither Figure 1, nor the text of D1 discloses an intermediate compression between the two membranes. As shown on Figure 1, a "recycle blower" is used to return the retentate from the second membrane to the feed inlet of the first membrane at the high pressure, exhaust side of the feed compressor. The board notes that the author used on the one hand the terms "feed compressor" in connection with the

compression of the feed mixture and on the other hand the terms "recycle blower" in the retentate recycle loop. In the board's judgment, the different wording used and the fact that the recycle fluid is returned after the feed compressor imply that no substantial recompression of the recycle fluid was required and, as pointed out by the respondent, the said blower mainly ensures gas flow in the loop. The pressure of the retentate from the second membrane is therefore essentially the same as the pressure of the feed to the first membrane. Since, in addition, no further compression between the two membranes is disclosed in D1, it follows that there is no substantial pressure drop from the first membrane feed to the first retentate and no significant pressure drop from the second membrane feed to the second retentate. This means, in other words, that the pressure of the retentate from the first membrane is "essentially the same" as the feed pressure to the first membrane and the pressure of the retentate from the second membrane is "essentially the same" as the feed pressure thereto.

The board is aware that a small pressure drop may occur between the inlet of a membrane unit and the retentate thereof, in particular due to piping; claim 2 however is not limited to identical pressures (see the expression "essentially the same pressure") but, as confirmed by the appellants at the oral proceedings, indeed encompasses a small pressure drop.

- 2.2.4 The appellants stressed that one essential difference between the subject-matter of claim 2 and D1 would be, on the one hand, that in the process claimed a first process product is recovered as the permeate of the

first membrane and, on the other hand, that a second process product is simultaneously recovered as the permeate of the second membrane.

The board notes that according to D1, a toluene-rich permeate and an air-rich permeate are recovered, said permeates being optionally further treated to recover pure toluene and essentially toluene-free air (see page 2, paragraph 2). Thus, both permeates are recovered as products of a process.

Contrary to the appellants' contention, the subject-matter of claim 2 is not limited to the recovery of products which later on should not be discarded or burnt. The board can thus only interpret the said features relating to the recovery of a first and a second process products in the way given by the appellants themselves, namely that they are the permeates of respectively the first and second membranes, i.e. exactly what they also are in D1. The process of D1 being moreover run continuously, the recovery of the process products as permeates of the first and second membranes is implicitly made simultaneously.

2.2.5 For the above reasons, the subject-matter of claim 2 lacks novelty over D1 (Article 54(1) and (2) EPC). The main request is therefore rejected.

3. *First auxiliary request*

3.1 Allowability of the amendments under Article 123 EPC

The independent claims 1 and 2 of this request differ from those of the main request in that:

(i) the gas(es) of small kinetic diameter are defined as being **H<sub>2</sub> or He**, and

(ii) the condensable gas(es)/gas vapours are defined as being **C<sub>2</sub> and higher hydrocarbons**.

These features having a basis e.g. at page 1, first paragraph and page 3, second and third paragraphs of the application as filed, they do not contravene the requirements of Article 123(2) EPC. Concerning the remaining amendments to claims 1 and 2 and the dependent claims 2-24, the observations made in item 2.1 *supra* apply likewise to this request, which therefore meets the requirements of Article 123 EPC.

3.2 Novelty

3.2.1 The board understands the wording "C<sub>2</sub> and higher hydrocarbons" as necessarily implying the presence of a C<sub>2</sub> and at least two other higher hydrocarbons; this was confirmed by the appellants during the oral proceedings.

3.2.2 As explained in item 2.2.2 *supra*, D1 discloses a process using two different membranes with reverse selectivities for recovering air and toluene from a binary mixture of these two compounds. D1 further teaches to use the same process for the recovery of

volatile organic compounds (VOCs) from process gases (page 1, last two lines of the first paragraph), or alternatively for separating nitrogen from methane (page 1, 2<sup>nd</sup> and 3<sup>rd</sup> lines of the paragraph bridging pages 1 and 2).

The board accepts that air implicitly contains traces of hydrogen and/or helium as gas(es) of small kinetic diameter, however neither toluene (a hydrocarbon with seven carbon atoms), nor the generic term "volatile organic compounds" can be acknowledged as disclosing the specific term "C<sub>2</sub>".

Bearing in mind the interpretation of the wording "C<sub>2</sub> and higher hydrocarbons" (see item 3.2.1 *supra*), and D1 being considered as not disclosing the term "C<sub>2</sub>", the subject-matter of claim 2 is therefore novel over D1.

3.2.3 It was not disputed that the process of claim 1 is also new with respect to the disclosure of D1. In fact, in D1 the feed first passes through a glassy membrane whereas it first passes through an organophilic one in the process according to claim 1.

3.2.4 The respondent argued that D2 destroyed the novelty of claim 1. In fact, D2 discloses a separation process in which a gas mixture consisting of light hydrocarbons and at least H<sub>2</sub>, CO<sub>2</sub> or CO is passed through a silicone membrane wherein the high-boiling hydrocarbons pass through the membrane more quickly than methane, the retentate depleted with the high-boiling hydrocarbons being then passed through a cellulose acetate membrane which allows H<sub>2</sub>, CO<sub>2</sub> and CO to pass through more quickly than the light hydrocarbons; H<sub>2</sub>, CO<sub>2</sub> or CO being thus

separated from the gas mixture (see D2, abstract and Figure; English translation of D2, paragraph bridging pages 1 and 2; page 3, fourth paragraph; page 4, third paragraph).

D2 (English translation, page 2, paragraph headed "industrial field of application") discloses to use this process for treating a mixture of light hydrocarbon gases with carbon dioxide, like natural gas or digestive gases. As submitted by the respondent, it can be seen from the Table on page 154 of D2 that the specific natural gas used in the example contains  $C_2$  and higher hydrocarbons. However, the board notes that it **contains neither  $H_2$ , nor  $CO$** . Furthermore, according to the data reported in said Table, the retentate 5 of the silicone membrane (first membrane) is not enriched with  $CO_2$  whereas the permeate 4 is enriched with  $CO_2$  and  $C_2$ - to  $C_{6+}$ -hydrocarbons.

D2 (last paragraph of page 5 of the English translation) further teaches "that the process according to the invention can furthermore be used in the same manner on gas mixtures consisting of light hydrocarbon and hydrogen and/or carbon monoxide, to effectively prevent condensation in the membrane stage by reduction of the dew point in the feed gas". This sentence, although referring to the process defined in a general way in D2, does not directly and unambiguously teach that in the case of a gas mixture containing  $H_2$  and "light hydrocarbon", the latter would inevitably contain  $C_2$ . It is also not directly and unambiguously derivable from the said sentence that the retentate 5 of the first separation step would be

enriched with H<sub>2</sub>, although in the example involving CO<sub>2</sub> it is not enriched therewith.

As regards the respondent's argument that CO<sub>2</sub> and H<sub>2</sub> are technical equivalents in the process of D2, it should be emphasized that according to the table on page 154, the retentate 5 of the silicone membrane (first membrane) is, as already indicated above, not enriched with CO<sub>2</sub>. Therefore, if (as alleged by the respondent) H<sub>2</sub> and CO<sub>2</sub> were technically equivalent, the silicone membrane would be expected to behave in a similar way with respect to hydrogen as with respect to carbon dioxide and thus, contrary to what is required in claim 1, the retentate 5 would consequently also not be enriched with H<sub>2</sub>.

In a second line of argument, the respondent further argued that a silicone membrane, such as the one used in D2, would inherently reject hydrogen. In the board's judgment, in view of the data in the table of D2, this would mean that the first membrane would behave differently with respect to CO<sub>2</sub> and H<sub>2</sub> and thus that they cannot be considered as technically equivalent in the process of D2. Under these circumstances the information disclosed in the table of D2 cannot be transferred to mixtures of H<sub>2</sub> and light hydrocarbon.

As a conclusion even if C<sub>2</sub> is disclosed in combination with CO<sub>2</sub> in the example, CO<sub>2</sub> cannot simply be interchanged with H<sub>2</sub> therein and even if it could, then the feature that the retentate of the first membrane is enriched with H<sub>2</sub> would not be directly and unambiguously derivable from D2. Thus, the board is not convinced



that D2 destroys the novelty of the process according to claim 1.

The subject-matter of claim 1 is therefore novel over D2.

3.2.5 The subject-matter of claim 2 is also novel over the disclosure of D2 at least due to the reverse sequence of the membranes, the first membrane being the glassy one in the process as defined in claim 2. This was not disputed.

3.2.6 Neither the other documents cited as novelty-destroying during the opposition procedure, namely D3, D4 and D17, nor the remaining documents on file anticipate the subject-matter of present claims 1 and 2. In particular D3 (US-A-4994094) and D4 (US-A-5407467) do not deal with the simultaneous recovery of H<sub>2</sub> or He on the one hand, and of C<sub>2</sub> and higher hydrocarbons, on the other hand. D17 (US-A-5755855) discloses exclusively in example 1 a two-membranes system. However it is silent as to the feed/retentate pressure ratio of the second membrane.

3.2.7 The subject-matter of claims 1 and 2 of auxiliary request I being novel over the cited prior art and claims 3-24 being dependent on these claims, they also meet the requirements of Article 54(1) and (2) EPC.

3.3 Inventive step of the subject-matter as claimed in auxiliary request I was neither discussed during the oral proceedings before the opposition division nor considered at all in the decision under appeal. The board, in the exercise of its discretionary power

pursuant to Article 111(1) EPC, finds it appropriate to remit the case to the opposition division for further prosecution.

## **Order**

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

1. The decision under appeal is set aside
2. The case is remitted to the opposition division for further prosecution on the basis of the first auxiliary request as filed during the oral proceedings

The Registrar:

The Chairman:

A. Wallrodt

M. Eberhard