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D E C I S I O N
of 15 September 1994

Case Number: T 0765/93 - 3.4.2

Application Number: 88107943.8

Publication Number: 0336999

IPC: B01D 13/04, B01D 53/22

Language of the proceedings: EN

Title of invention:
Reactive posttreatment for gas separation membranes

Applicant:
E.I. DU PONT DE NEMOURS AND COMPANY, ET AL

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54(3), 56, 123(2)

Keyword:
"Novelty - yes, after amendment"
"Inventive step - yes"
"Added subject-matter - no"

Decisions cited:
-

Catchword:
-

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Case Number: T 0765/93 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 15 September 1994

Appellant: E.I. DU PONT DE NEMOURS AND COMPANY
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Decision under appeal: **Decision of the Examining Division of the European Patent Office dated 23 March 1993 refusing European patent application No. 88 107 943.8 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: E. Turrini
Members: C. Black
L. C. Mancini

Summary of Facts and Submissions

I. European patent application No. 88 107 943.8, filed 18 May 1988 and published on 18 October 1989 with the publication No. 0 336 999, claiming the priority date 13 April 1988 of a US application, was refused by a decision of the Examining Division on the ground that its subject-matter lacked novelty having regard to the disclosure in WO88/10140 (D1), filed 25 June 1987 and published 29 December 1988 and therefore state of the art according to Article 54(3) EPC. The Examining Division further observed, under "Additional Information" stated to be not forming part of the decision, that the subject-matter of the application lacked an inventive step having regard to the disclosure in JP-A-5959222 (D2) or JP-A-5959220 (D3), in both cases in the form of abstracts, taking into account common general knowledge as evidenced by Ullmanns Encyklopädie der Technishen Chemie, 1978, Volume 16, pages 515 to 517 and 532, 533.

II. The gist of the Examining Division's argumentation is that Claim 1 then under consideration required applying to the surface of a gas separation membrane at least two monomers and causing these to react together to form a polymer. D1 discloses applying two monomers to respective different sides of a membrane and causing these to react to seal defects in the membrane. In the Examining Division's view a membrane has only one surface comprising the two opposite sides and, where present, the walls of pores extending between the two. For this reason the subject-matter of Claim 1 lacked novelty.

III. The present appeal lies against this decision.

IV. Oral proceedings were held, at which the Appellant requested that the decision under appeal be set aside and a patent granted on the basis of amended Claim 1 submitted at the oral proceedings, reading as follows:

"Process for improving the selectivity of a gas separation membrane by sealing defects or imperfections in the surface of the gas separation membrane having a dense separating layer, comprising applying to the side of the gas separation membrane having the dense separating layer at least two monomers, causing the monomers to react with each other to plug the holes in the dense separating layer without forming a continuous layer of polymer."

V. The Appellant's written and oral argumentation may be summarised as follows. Claim 1 as amended makes clear that the two (or more) monomers are applied to the same side of the membrane, thus distinguishing its subject-matter over D1. D2 and D3 disclose methods of preparing composite membranes, not sealing defects in already formed membranes. The difference is emphasised by the feature in Claim 1 that the monomers react with each other to plug the holes without forming a continuous layer of polymer.

Reasons for the Decision

1. Claim 1 now under consideration differs from that originally filed in several respects and the question of compliance with Article 123(2) EPC has to be investigated. The claim now relates to a process for improving the selectivity of a gas separation membrane

and this has a counterpart in the original Claim 1 which contained the wording "which causes the selectivity of the membrane to be improved".

The claim then requires that the gas separation membrane is one "having a dense separating layer." The whole of the description relates to asymmetric membranes, which may be integral or composite (page 8, lines 20, 21), which have a separating skin (for example page 3, lines 13 to 16) otherwise called a dense separating layer (for example page 5, lines 23 to 27).

The claimed process is to seal defects or imperfections in the surface of the gas separation membrane. Page 3, lines 13 to 16 refers to sealing defects in the separating skin, and page 5, lines 23 to 31 refers to the imperfections and defects which arise in the membrane formation process and states that the disclosed process achieves the goal of sealing the defects and, it can be assumed, imperfections.

The claim then requires that at least two monomers are applied to the side of the gas separation membrane having the dense separating layer. On page 3 under "Detailed Description" methods of making gas separation membranes are referred to, including "casting a film or extruding a hollow fibre from a solution of a polymer in a solvent mixture, evaporating a portion of the solvent from one side of the film or the outside of the hollow fibre and quenching in a non-solvent". The said "one side" or "outside" becomes the skin side or dense separation layer of the membrane and this is the side wherein defects are sought to be sealed (page 3, lines 14, 15). Further all of the Examples of the invention describe the application of two monomers to the outside (skin side) of a hollow fibre membrane. It

is clear therefore from the description as a whole that the two (or more) monomers are to be applied to the same side of the membrane.

The foregoing conclusion requires some qualification. Examples 1 to 3, 5 to 22, 24 to 26, 29 to 39 and 41 to 49 all refer to the outer surfaces of the membranes, which is consistent with the above interpretation, because this can be read as referring to the outer surface of each membrane. Example 40, referring to the outer surface of the membrane, presents no problems. However, Control Examples 1 to 3, Examples 4, 23, 27, 28, 40, Comparative Example 1 and Example 50 all refer to the outer surfaces of the membrane, suggesting that each membrane can have more than one outer surface. This point was discussed at the oral proceedings, and it was concluded that any ambiguity resulted from somewhat loose wording in the last-mentioned set of Examples and that for the reader familiar with the art there was no other interpretation than that the monomers were to be applied to the side of an asymmetric membrane having the skin or dense separating layer and that in the case of the Examples this side is the outside.

The Board can agree with the Examining Division that strictly speaking a membrane has only one surface. However, in the present case, in the context of the description the terms surface and side are synonymous. In any case the claim has been clarified in this respect.

Finally the two or more monomers are caused to react with each other (a) to plug the holes in the dense separating layer and (b) without forming a continuous layer of polymer.

As regards (a) page 6, lines 29 to 31, states that the reactive chemical substances are then anchored in place through the reaction. Then according to page 9, lines 32 to 34 a larger range of pore size distribution can be effectively plugged by the disclosed procedure than in the prior art. Page 10, lines 29, 30 states that the procedure allows the substantial plugging of pore sizes too small for other procedures and lines 32, 33 that the material reacts in place to form a plug to fit the pore size. There is therefore a clear basis for plugging holes, which holes are in the dense separating layer because it is this layer which is required to be pin-hole free - see for example the description, page 5, lines 11 to 22.

The Appellant acknowledges that there is no explicit disclosure that no continuous layer of polymer is formed over the dense separating layer. An implicit basis can be found on page 10, lines 2 to 7, where it is indicated that prior art processes are only effective by applying a thin coating, which can be assumed to be substantially continuous, on top of the membrane material and that this causes a substantial loss in membrane productivity. It can be accepted that it follows that the claimed process seeks to avoid this, and this borne out by the Examples, which disclose an acceptable reduction in permeability (productivity) as compared with the large increase in selectivity which is achieved.

Moreover the expressions "anchored in place" (page 6, line 30), "effectively plugged" (page 9, line 33), "substantial plugging" (page 10, line 29) and "reacts in place to form a plug" are more suggestive of individual plugs filling separate pin-hole defects that of a continuous coating. This is confirmed by SEMs which accompanied the Appellant's communication dated 18 August 1994. These show, on the surface of membranes

treated by the claimed process, isolated rounded "bumps" which can be accepted as being the surface portions of individual plugs which have possibly encroached a little on to the surface of the membrane but remain isolated from each other.

It is true that on page 6, lines 34, 35 it is stated that the polymer is **believed** to form and grow within the membrane and on page 9, lines 22 to 27 that the effect that the membrane defects and imperfections have on the gas-pair selectivity is **believed** to be due to healing of these through plugging or partial plugging, so that these passages qualify the interpretation of the description for the purpose of ascertaining the compliance of the amended Claim 1 with Article 123(2) EPC. Nevertheless the Board accepts that the general picture obtained from reading the description as a whole, and taking into account the above-mentioned SEM's, is that of plugging of individual pin-holes without forming a continuous layer of polymer. The requirement of Article 123(2) EPC is therefore satisfied.

2. From the foregoing it will be apparent the subject-matter of Claim 1 differs from the disclosure in D1 at least in that the two or more monomers are applied to one side of the membrane, so that the said subject-matter is novel.

3. Although the Examining Division's observations concerning inventive step did not form part of the decision, the Appellant gave counter-arguments in the grounds for the appeal and in the circumstances of the present case the Board considers it appropriate to make use of its powers under Article 111(1) EPC, second

sentence and to decide also on the question of inventive step rather than remit the case to the Examining Division to this end.

The Appellant in effect argues that D2 and D3 are describing methods for forming composite membranes by depositing comonomers on a porous support followed by polymerisation. The support may be a membrane, but it is not a gas separation membrane. Deposition of the polymer converts it into a gas separation membrane, it does not seal defects in an already formed gas separation membrane. The Board agrees that this is so.

The Board, in the communication accompanying the summons to oral proceedings, referred to a standard reference work, Gas Purification, fourth edition, Gulf Publishing Company, Houston, Texas (1985) which can be considered as reflecting common general knowledge. On page 872, second paragraph, it is stated that these base materials (sheet or hollow fibre gas separation membranes) may be coated on the active face with a thin layer of another polymer to seal surface pores or modify separation properties. The Board expressed the provisional view that this might suggest to the average skilled person that the same process could be used for completing a composite gas separation membrane or for sealing an already formed one. That is, the processes disclosed in D2 or D3 for forming composite membranes might also be used for sealing defects. However, it is accepted that in Gas Purification, it seems that a preformed polymer is coated on the active face and that any conclusion based on coating with a preformed polymer does not necessarily apply also to deposition of comonomers followed by polymerisation as in D2 and D3. In any case in Gas Purification, D2 and D3 a substantially continuous layer of polymer is formed. The subject-

matter of Claim 1 cannot therefore be said to be obvious over these documents and thus involves on inventive step.

4. Finally the Board can accept that the concentration of the monomers used (about 0.1 to 1.0 weight per cent solutions) and the contact times (about 10 minutes) in the numerous examples give the necessary instruction as to how the claimed process is to be carried out in order to achieve the desired plugging of defects (see the grounds for the appeal, paragraph bridging pages 2 and 3.

5. The description requires to be adapted to take into account the amendments made to Claim 1, and in the Board's opinion this adaptation is best done at leisure by the Examining Division. The Board has moreover noted the following clerical errors in the description:

page 4, line 31, "selectively" should be "selectivity"
page 4, line 34, "mateiral" should be "material"
page 8, line 15, "therein" should be "thereof"
page 9, line 24, "selectively" should be "selectivity"
page 21, line 8, "rises" should be "rinses"
page 23, line 15, "loosing" should be "losing"
page 33, line 5, "SLYGARD" should be "SYLGARD"

This list is not guaranteed to be exhaustive. The opportunity may be taken, in adapting the description, also to make these corrections.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Examining Division with the order to grant a patent on the basis of Claim 1 as filed at the oral proceedings and claims 2 to 24 as originally filed, with description to be adapted.

The Registrar: The Chairman:

E. Görgmaier E. Turrini