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D E C I S I O N
of 8 June 1994

Case Number: T 0806/92 - 3.4.2

Application Number: 84105108.9

Publication Number: 0144493

IPC: B01D 13/04

Language of the proceedings: EN

Title of invention:
Method of preparing asymmetric membranes

Patentee:
Memtec America Corporation

Opponent:
01) Hoechst Aktiengesellschaft
02) Sartorius AG

Headword:
-

Relevant legal norms:
EPC Art. 56

Keyword:
"Inventive step - No"

Decisions cited:
-

Catchword:
-

Case Number: T 0806/92 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 8 June 1994

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Decision under appeal:
Decision of the Opposition Division of the European Patent Office dated 12 April 1991, written decision posted on 26 June 1992 revoking European patent No. 0 144 493 pursuant to Article 102(1) EPC.

Composition of the Board:

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

Summary of Facts and Submissions

I. The Appellant is the proprietor of European patent no. 0 144 493 which was granted with nine claims of which Claim 1 reads as follows:

"A method of preparing an asymmetric integral membrane comprising a skin and a porous support having a reticulated structure, which comprises casting a polymer dope while said dope is in a turbid metastable liquid dispersion condition, the concentration of polymer in said polymer dope being high enough to produce a coherent membrane yet low enough to form said reticulated structure."

II. The patent was revoked by decision of the Opposition Division on the ground that the subject-matter of amended claims presented in the course of the opposition proceedings, although novel, did not involve an inventive step. The documents relied on by the opposition were:

US-A-3 615 024, (D1)
K. Koch's Dissertation "Untersuchungen über den Bildungsmechanismus asymmetrischer Membranen für die umgekehrte Osmose", Dissertation zur Erlangung der Würde eines Doktors der Naturwissenschaften dem Fachbereich Chemie der Eberhard-Karls-Universität zu Tübingen (1975) (D2)
R.E. Kesting, Synthetic Polymeric Membranes, McGraw Hill, 1971, Chapter 5, pages 116 to 157, (D3).

The documents Journal of Chemical Physics (John H. Cahn) Vol. 42, No. 1, (1965) pages 93 to 99,

and Kolloid-Z. u. Z. Polymere (C.A. Smolders et al), 243 (1971) pages 14 to 20 which had been introduced by the Patentee during oral proceedings, were also referred to in the decision.

III. The present appeal lies against this decision. In the course of the appeal proceedings, the Respondent Sartorius (O2) introduced the document J.Appl.Polym.Sci. 21 (1977) pages 199 to 215 (D4). At oral proceedings the Appellant requested that the decision under appeal be set aside and the patent maintained in amended form on the basis of a set of Claims 1 to 6 of which Claim 1, amended during the oral proceedings, reads as follows:

"A method of preparing an asymmetric integral membrane comprising a skin and a porous support which comprises preparing a polymer dope comprising a polymer, a solvent for the polymer, and a diluent which is a non-solvent for the polymer, casting the polymer dope as a film on a support surface, and quenching the cast film by contact with a quench liquid in which the polymer is insoluble, **characterised in that**

(a) the polymer dope is cast while it is a turbid metastable liquid-liquid dispersion which has been brought close to its spinodal by adding non-solvent or adjusting the temperature,

(b) the membrane is formed as a result of phase separation by spinodal decomposition,

(c) the concentration of polymer in the polymer dope is high enough to produce a coherent membrane yet low enough to form substantially all reticulated structure within said porous support,

(d) the skin has an average pore diameter of from about 0.05 µm to about 3 µm, and

(e) the pores in the porous support are largest at the surface opposite the skin and have gradually decreasing size towards the skin with the smallest pores being those in the skin."

Both Respondents requested that the appeal be dismissed.

IV. The gist of the Appellant's written and oral argumentation is as follows. The claimed process differs from prior art processes particularly in that the casting dope is in the form of a metastable liquid-liquid dispersion which has been brought close to the spinodal and the membrane is formed by spinodal decomposition. Previously it was thought that such dispersions would be unsuitable because of their instability, and casting dopes which had undergone phase separation were normally discarded. D1 indeed discloses adding diluents (solution modifiers) to the casting dope but these can increase as well as decrease the solvating effect of the solvent system. Those that decrease the said effect lead to a reduction in pore size of the eventual membrane and in this respect their use can be considered disadvantageous, as decreasing the flux rate.

D1 moreover discloses the use of comparatively small amounts of the said solution modifiers (up to 10%, usually 1 to 6% - column 10, lines 44 to 47) and when the term non-solvent is used (column 10, line 48) again a small quantity, functioning e.g. as a thickening agent, is disclosed. Such small quantities are

insufficient to bring the casting dope close to the spinodal.

The passage in D1 starting at line 57 of column 10 leads away from the claimed process, in that it proposes clarification of the casting dope by centrifugation before drawing films therefrom. If the metastable dispersion required by Claim 1 were to be centrifuged, segregation into two layers would result. The fact that the Tyndal effect need not be eliminated (D1, column 10, line 62) does not mean that the casting dope is in a metastable condition.

V. The argumentation of the Respondents can be summarised as follows. The main distinctions sought to be drawn between the subject-matter of Claim 1 and the disclosure in D1 reside in features (a) and (b). Feature (b) is not based on a reproducible technical teaching but rather seeks to provide a theoretical explanation of how the membrane is formed. Such a feature cannot serve to distinguish the claimed process from the prior art.

In feature (a) the expression "close to" is indefinite and therefore does not provide a clear distinction over the prior art. The part-feature "by adding non-solvent or adjusting the temperature" belongs to the competence of the average skilled person. Therefore there only remains that the dope is cast while it is in a metastable liquid-liquid dispersion. This is arguably known from D1 which discloses the addition of non-solvent to the casting dope and that the casting dope should be agitated between preparation and casting, thus indicating that it is not a pure solution. But

even if the subject-matter of Claim 1 were considered to be novel over D1, it would not involve an inventive step over a combination of D1 and D3 since the latter contemplates the use of casting dopes containing non-solvent and which are emulsoid or otherwise comprise a two-phase dispersion.

Reasons for the Decision

1. The appeal is admissible.

2. Since, as will be seen, the appeal fails because the subject-matter of Claim 1 does not involve an inventive step, the Board does not propose to examine in detail the various amendments introduced into the claim as compared with the granted Claim 1 (which is moreover identical to Claim 1 as originally filed) for conformity with other requirements of the EPC. The said amendments will be considered where necessary in the discussion of inventive step.

3. The Opposition Division, in paragraph D of the reasons for its decision found the subject-matter of Claim 1 then under consideration novel. The Respondent 02, in paragraph 3 of the response to the grounds for the appeal, appeared to accept this in respect of present Claim 1 which does not differ substantially, but in the oral proceedings reopened the question of lack of novelty. The Board can agree with the finding of the Opposition Division, for reasons which will become apparent in the discussion of inventive step.

4. Claim 1 is now drafted in the two-part form in accordance with Rule 29(1) EPC and it is not disputed that the first part is based on D1. The characterising portion is conveniently sub-divided into features (a) to (e) which will now be considered in turn.

5. Feature (a) includes, as compared with the original Claim 1, the requirement that the casting dope dispersion has been brought close to its spinodal by adding non-solvent or adjusting the temperature. The Appellant acknowledged that "close to" is a relative term, but the Board can agree that the use of relative expressions does not necessarily lead to lack of clarity in claims containing them. The problem arises when, as here, there is pertinent prior art to be taken into consideration, so that the term "close to" has to be investigated in order to establish whether it contributes something by way of clear definition in feature (a) which might constitute a distinction over the prior art.

To this end, recourse to the description is required, particularly in respect of the binodal and spinodal curves shown in the phase diagrams in Figures 1a and 1b. The process as originally claimed required that the casting dope was in a turbid metastable liquid dispersion condition, that is a condition represented by the shaded area between the binodal and spinodal curves, and column 4, lines 50 and 51 stated that this was an essential part of this invention. The claim is now restricted to the use of a casting dope whose state is defined by a part of this shaded area, namely, close to the spinodal. However the description gives little, if any, guidance as to how close is close. The

expression "close to" first appears in column 3, lines 38 to 52 in a passage talking in general terms about phase diagrams. This passage refers to a region close to the binodal in which the unstable system is believed to undergo phase separation by a nucleation and growth mechanism, and to a region at or close to the spinodal in which the mechanism may change to the so-called spinodal decomposition. It is not clear whether these two regions account for the whole of the region between the binodal and spinodal or whether there is an intermediate region. The expression "close to" appears again in column 5, lines 6 to 8, where it is stated that it is also believed that spinodal decomposition is favoured by bringing the casting dope close to its spinodal. Other passages of interest in connection with the spinodal are in column 4, lines 5 to 7, "toward the spinodal side of this region, microporous membranes (pore sizes of about 0.05 to about 3 micrometres) may be produced; lines 39 to 41 "When the system is further inside the binodal, segregation may occur within hours or even minutes" and lines 55 to 57 "in general, as the system approaches the spinodal from the binodal, the turbidity of the dispersion increases". Nothing in the foregoing gives a clear indication of how "close to" is to be interpreted in establishing the scope of Claim 1. Figure 2 also does not assist. It seeks to show the variation of water permeability and protein retention of the cast membranes with increasing non-solvent in the casting dope, and indicates that when the binodal is crossed there is a sudden increase in water permeability which then levels off. The spinodal is not shown, so that there is no indication of any additional role which operating near the spinodal would play in respect of

permeability. As regards protein retentivity, the sudden change takes place completely before the binodal is reached.

The specific examples do not mention the binodal or spinodal. Of these Examples I and II have been cancelled because they are outside the scope of the present Claim 1 (no non-solvent). Example VI is also outside the scope of the claim, because the membrane obtained has a pore size of 0.01 µm (column 8, lines 6 to 9). Examples VII and VIII could also be considered as not falling within the scope of the claim because the casting dope is cast directly into the quenching liquid and not first on to a support surface as in the claim. However this is not a serious objection because it could no doubt have been readily overcome. In addition to the foregoing, the Respondent 01 questioned whether any of the examples could be seen to fall clearly within the scope of present Claim 1. These were examples of the invention as originally claimed, and from their content there was no way of knowing whether they remained within the scope of the amended claim. In the Board's view the onus would have been on the Respondent 01 to show that the examples did not fall within the scope of the claim. Nevertheless, it does appear that the examples contribute nothing positive which would assist in interpreting "close to", particularly when the subject-matter of Claim 1 is compared with the disclosure in D1.

D1 discloses, as stated above and not disputed by the parties, the features of the first part of Claim 1. D1 also discloses adding to the casting dope solution modifiers (column 9, line 11 et seq.) which, inter

alia, reduce the solvating effect of the overall solvent system and are therefore at least poor solvents for the polymer, or non-solvents for the polymer (column 10, line 48 et seq.). D1 goes on to say that clarification of the casting dope by centrifugal action before drawing films therefrom may be advantageous, but need not be so complete as to cause for example elimination of a Tyndal effect from a casting dope containing inorganic salts. It is further stated that it is also desirable to keep the casting dope agitated between preparation and use in making film draw-downs.

The Board can agree with the Respondents that the foregoing, particularly in respect of agitation of the casting dope, indicates that for the person of average skill in the art, D1 contemplates the use of casting dopes that are not pure solutions.

It is true that D1 does not disclose addition of non-solvent to the casting dope to bring it close to the spinodal, which is not surprising since the role of spinodal demixing in the field of polymer blends, let alone asymmetric membranes was not described until the 1970s, and D4 (published 1977) is the first document of which the Board is aware which mentions spinodal demixing in respect of membrane formation. Nevertheless the general knowledge of the person of average skill in the art at the priority date of the patent in suit may be taken into account, and this is reflected by D3, a standard reference work in the field, possibly the only one at the said priority date.

Chapter 5 (pages 116 to 157) of D3 relates to porous phase-inversion membranes, therefore membranes of the

kind prepared by the process of the patent in suit. Page 116 refers to the structure of the membranes as being due to the presence of voids or vacuoles which originate in the emulsoid nature of the solutions from which they are formed. Page 119 refers to the appearance of two interdispersed liquid phases (during the formation of the membrane) and on page 120 it is stated that the droplets of the dispersed liquid phase need not be present in the initial solution; the corollary is that they may be present and this is then confirmed by the statement that if, inter alia, non-solvent is present (in the casting solution) two phases may be present initially. Page 121 goes on to say that the choice and concentration of non-solvent will determine the precise moment of phase inversion, droplet size and homogeneity. The sentence bridging pages 125 and 126 makes it clear that the preceding description is relevant to the technique which employs partial evaporation followed by immersion into a non-solvent medium for subsequent gelation and/or complete solvent removal by diffusion as in the patent in suit.

Accordingly, when the average skilled person interprets the teaching of D1 in the light of his general knowledge in the art as reflected in D3, he is aware that casting dopes may be emulsoid or two-phase and have been brought to such a state by the addition of non-solvent. D3 does not state any limit as to the amount of non-solvent that may be added (common sense dictates that it should not cause precipitation of the polymer) so that the Board is not persuaded on the information before it that there was any prejudice against adding non-solvent to the extent that the casting dope was brought close to the spinodal.

Accordingly feature (a), while it might arguably contribute to novelty, is not seen as importing a distinction into the subject-matter of Claim 1 which would contribute to the inventivity of the claimed process as compared with what is disclosed in D1 taking into account the disclosure of D3.

The Appellant argues that D1 only refers to solutions. However it is apparent that the word solution is sometimes used rather loosely, for example D3 refers on page 116 to the emulsoid nature of the solutions and on page 121 to the colloidal properties of multicomponent polymer solutions. The Board notes also that in the patent in suit, Example III refers to a turbid solution, Example IV to "this solution" previously stated to be cloudy, Examples VII and VIII again to "this solution" (the casting dope) and also the part of the Comparative Example relating to the invention refers to casting a solution. Therefore little significance can be attributed to the use of the word "solution" in D1.

6. Feature (b) requires that the membrane is formed by spinodal decomposition. This is somewhat dubious as a technical feature seeking to define the claimed process, being rather an attempt to explain the mechanism by which the membrane is formed. In any case the description of the patent in suit states at column 3, lines 48 to 52 that the mechanism of phase separation **may** change to the so-called spinodal decomposition in which periodic and network structures **may** form without nucleation, and in the paragraph bridging columns 4 and 5 that, while the process of solidification is not yet fully understood, **it is**

believed that the mechanism of solidification involves spinodal decomposition etc., and that it is also **believed that** this is favoured by bringing the casting dope close to its spinodal. The feature is therefore based on a conjecture. Apart from bringing the casting dope close to the spinodal, no process parameters are specified which ensure spinodal decomposition and in this respect feature (b) adds nothing by way of definition to feature (a).

Moreover, it was already known from D4 (see pages 202 and 214) that spinodal demixing, which is synonymous with spinodal decomposition, had a role to play in determining membrane structure, so that in the present case no contribution to inventivity can be seen in recognising this.

Finally, in respect of feature (b) the Board notes that in D1, column 5, lines 27 to 31, it is stated that at polymer concentrations between 10% and 40%, it frequently happens that both polymer-rich and polymer-poor phases are continuous - that is, a polymer-rich matrix containing an interconnecting array of voids filled with polymer-poor liquid is produced. Such a structure appears to the Board to be more representative of spinodal demixing than nucleation and growth and it could be argued that the skilled person, aware of D4, would recognise this. However the Board has not required to rely on this passage in its decision.

7. Feature (c) requires that the concentration of the polymer in the polymer dope is high enough to produce a coherent membrane yet low enough to form substantially

all reticulated structure within said porous support. This feature is in substance repeated in D1, column 10, lines 38 to 41, except that D1 does not refer here to a reticulated structure. However the patent in suit is based on an application which was divided out of the application granted as EP-B-0 036 315, the proceedings of which gave rise to appeal T 493/89. In point 8 of the reasons for the decision the Board concluded that from the weight of the evidence it was persuaded that membranes having a porous skin and a reticulated structure belonged to the state of the art, and the Board sees no reason to depart from this conclusion. Accordingly feature (c) is seen as being formulated in terms of a known result to be achieved and again does not contribute to the inventivity of Claim 1, the more so because the polymer concentration in the casting dope disclosed in D1 (5 to 20% - see column 8, lines 66 to 68) embraces that disclosed in the Examples of the patent in suit (5 to 10%).

8. Feature (d) requires that the skin has an average pore diameter of from about 0.05 µm to about 3 µm. This range overlaps that disclosed in D1, namely 0.001 to 1 µm (see Claim 1 and the corresponding description). No invention can be seen in extending this range farther into the microfiltration range.

9. As to feature (e), this finds a direct counterpart in D1, column 6, lines 46 to 54. In this respect the Appellant sought to distinguish the membrane produced by the claimed process from that according to D1 in that the latter showed only a single level of asymmetry whereas the former showed a high level of asymmetry in depth. The quoted passage in the Board's view

contradicts this, stating that the pore size is much the smallest in the skin and grows rapidly as the distance from the skin increases. In any case feature (e), like feature (d) is not a process feature, and these features do not contribute to a clear definition of the invention in the absence of corresponding process features by which the stated result might be achieved.

10. Other distinctions sought to be drawn at the oral proceedings between the claimed process and the disclosure in D1, namely the very rapid kinetics of the membrane formation, the selectivity, permeability, throughput and good physical properties of the formed membrane and the fact that it can be used with the coarse side facing the feed are not features of the claimed process or reflected in the wording of the claim.

11. The various features (a) to (e) have been thus shown to contribute nothing individually to the inventivity of Claim 1 and in the Board's view it will be clear from the foregoing that this also applies to their combination.

The subject-matter of Claim 1 is therefore not seen as involving an inventive step over the disclosure of D1 when common general knowledge in the art, as evidenced by D3, is taken into account.

12. Claims 2 to 6 fall because they are dependent on a claim found not to be allowable. No feature of these claims was moreover made the subject-matter of an auxiliary request.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

P. Martorana E. Turrini