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File Number: T 493/89 - 3.4.2  
Application No.: 81 301 074.1  
Publication No.: 0 036 315  
Title of invention: Anisotropic membranes

Classification: B01D 13/04

D E C I S I O N  
of 26 February 1991

Proprietor of the patent: MEMTEC AMERICA CORPORATION

Opponent: 1) Enka AG  
2) Hoechst AG  
3) Sartorius GmbH  
4) Fresenius AG

Headword:

EPC Article 56

Keyword: "Inventive step - denied"

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Headnote



Case Number : T 493/89 - 3.4.2

**D E C I S I O N**  
of the Technical Board of Appeal 3.4.2  
of 26 February 1991

**Appellant :**  
(Proprietor of the patent)

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**Decision under appeal : Decision of Opposition Division of the European Patent Office dated 30 May 1989 revoking European patent No. 0 036 315 pursuant to Article 102(1) EPC.**

**Composition of the Board :**

**Chairman :** E. Turrini  
**Members :** C. Black  
C.V. Payraudeau

## Summary of Facts and Submissions

- I. European patent No. 0 036 315 was granted on the basis of European patent application No. 81 301 074.1.
- II. The patent was opposed by the Respondents on the grounds that its subject-matter was not novel or inventive and its description insufficient.
- III. The patent was revoked by decision of the Opposition Division on the ground that its subject-matter did not involve an inventive step.
- IV. An appeal was lodged against this decision.
- V. Oral proceedings were held, attended by the Appellant (Patentee) and three of the Respondents (Opponents 01, 02 and 03). The fourth Respondent (Opponent 04) had notified the Board of his intention not to attend the proceedings.
- VI. At the end of the proceedings the appellant maintained the request set out in the grounds of appeal, that is, that the decision under appeal be set aside and that the patent be maintained in amended form on the basis of Claims 1 to 7 submitted as main request with letter dated 4 October 1989 (main request),  
Claims 1 to 7 submitted as subsidiary request 1 with letter dated 4 October 1989 (1st auxiliary request),  
Claims 1 to 6 submitted as subsidiary request 2 with letter dated 4 October 1989 (2nd auxiliary request),  
Claims 1 to 7 submitted as subsidiary request 3 with letter dated 4 October 1989 (3rd auxiliary request).

The Appellant further requested that the proceedings be continued in writing in the case that the article Ultrafiltration, Alan S. Michaels, Progress in Separation

and Purification, Volume 1, Interscience Publishers (1968) or the book "Synthetic Polymeric Membranes" by Kesting (1971) McGraw Hill Company were to be used in an adverse decision.

The first of these document was introduced in Respondent 03's communication dated 14 February 1991, the second by Respondent 03 during the oral proceedings.

The Respondents (including Respondent 04 in the written submissions) requested that the appeal be dismissed. Respondent 03 further requested that if the Appellant's experimental report dated 8 January 1991 and the accompanying photographs were to be taken into account in an adverse decision, the proceedings should be continued in writing.

In addition to the above-mentioned documents, the following are referred to in this decision:

US-A-3 615 024 (Michaels)

K. Kock's Dissertation "Untersuchungen über den Bildungsmechanismus asymmetrischer Membranen für die umgekehrte Osmose" (Kock) (1975)

Research and Development Progress Report No. 359 (October 1968) United States Department of the Interior (North Star)

US-A-3 988 245 (Wang).

VIII. Claim 1 according to the main request reads as follows:

"An asymmetric integral polymeric membrane comprising a skin and porous support, characterised in that the

membrane has a bulk porosity greater than 70%, the skin contains pores of average diameter 0.005 to 3.0  $\mu\text{m}$ , and the support comprises a reticulated structure containing pores which gradually increase in size from the skin to the opposite face of the support, the average pore diameter of the pores at said opposite face being from 10 to 20000 times the average pore diameter of the skin pores."

Claim 1 according to the first auxiliary request reads as follows:

"A polymeric membrane useful as an ultrafilter or microporous membrane comprising a porous support, characterised in that the membrane is an asymmetric membrane having an integral skin and has a bulk porosity greater than 70%, the skin containing pores of average diameter 0.005 to 3.0  $\mu\text{m}$ , and the support comprises a reticulated structure containing pores which gradually increase in size from the skin to the opposite face of the support, the average pore diameter of the pores at said opposite face being from 10 to 20000 times the average pore diameter of the skin pores."

Claim 1 according to the second auxiliary request reads as follows:

"A polymeric membrane useful as an ultrafilter or microporous membrane comprising a porous support, characterised in that the membrane is an asymmetric membrane having an integral skin and has a bulk porosity greater than 70%, the skin containing pores of average diameter 0.005 to 3.0  $\mu\text{m}$ , and the support comprises a reticulated structure containing pores which gradually increase in size from the skin to the opposite face of the support, the average pore diameter of the pores at said

opposite face being from 10 to 20000 times the average pore diameter of the skin pores, wherein when the membrane is disposed in a fluid flow with the support upstream, the volume of fluid passed through said membrane by the time the rate of flow through said membrane is reduced to 50% of the initial value is at least twice the volume of fluid which would be passed through the same membrane if it had its skin upstream."

Claim 1 according to the third auxiliary request reads as follows:

An asymmetric integral polymeric membrane comprising a skin and porous support, characterised in that the membrane has a bulk porosity greater than 70%, the skin contains pores of average diameter 0.005 to 3.0  $\mu\text{m}$ , and the support comprises a reticulated structure containing pores which gradually increase in size from the skin to the opposite face of the support, the average pore diameter of the pores at said opposite face from being 10 to 20000 times the average pore diameter of the skin pores, the water permeability (cm/min psi) of the membrane being at least 0.5 at 0.05  $\mu\text{m}$ , 4 at 0.2  $\mu\text{m}$  and 7.4 at 0.4  $\mu\text{m}$  pore size, with a coefficient of variation of up to 50%."

IX. The gist of the Appellant's argumentation is as follows:

The person of average skill in the art cannot derive from the teaching of Michaels a membrane having all of the features required by Claim 1. Michaels invariably starts from a casting solution and not from a metastable dispersion as in the process which leads to the claimed membrane, and the product is therefore a membrane having a microporous skin supported on a granular macroporous layer having finger-like voids, or a non-porous skin on a

reticulated support structure. Any modification of Michaels, for example taking into account the teaching of Kock, leads to the same result. Kock is in any case directed to the investigation of reverse osmosis membranes, therefore concerns a different technical field, and any more general teaching contained therein is saying no more than Michaels; a combination of the teaching of Michaels and Kock therefore does not go beyond either of the documents. North Star is also concerned with reverse osmosis membranes, admittedly in particular with supports for the membrane proper. The experimental report accompanying the letter of 8 January 1991 shows that the membranes functioning as support structures and prepared according to the North Star teaching, are different from the claimed membranes.

The argumentation of the Respondents in the written and oral proceedings may be summed up as follows:

Process features largely relied on by the Appellant in the written proceedings cannot be used to defend a claim to a product per se. The designation of the porous support as reticulated does not serve to distinguish the asymmetric membrane according to Claim 1 of the patent in suit from the prior art membranes, in particular as disclosed in Michaels, Kock and North Star.

#### Reasons for the Decision

1. The appeal is admissible.
  
2. The Board is satisfied that no objection under Article 123(2) or (3) arises in respect of the main request and the first and second auxiliary requests. As regards the third auxiliary request, the added feature,

relating to the water permeability of the membrane, is taken from examples I, II and IV. The validity of the generalisation is open to question, particularly since example I uses a different polymer from the other two. However since the appeal does not succeed for other reasons, this need not be gone into further.

3. The Opposition Division concluded that the subject-matter of Claim 1 of the main request then under consideration was novel; this has been disputed in the appeal proceedings, in particular by 02. However it appears appropriate in the present case to deal with novelty, not in isolation, but together with inventive step.
4. What purports to distinguish the membranes according to the patent in suit from the prior art is the combination of a reticulated structure for the support portion of the asymmetric membrane and the specific range of pore diameters of the skin portion thereof. In the Appellant's view this combination is not known. A reticulated structure is known for symmetric membranes, but in the case of asymmetric structures having a porous skin, the support is granular with finger-like intrusions. Any modification of process conditions, e.g. as taught in Michaels, so as to achieve a reticulated structure results at the same time in a substantially non-porous skin.
5. The Board is not persuaded that this is the case. It is true that Michaels, or any other English language document cited in the proceedings, does not use the term "reticulated". Michaels however discloses asymmetric membranes having a skin containing pores of diameter from 0.001 to 1 micrometers, this range overlapping that required by Claim 1 at the ultrafiltration end. According to column 3, lines 30 to 34, the integral support comprises a much more coarsely porous polymer structure.

In the context, much more coarsely porous appears to mean in comparison with the skin. Column 4, lines 17 to 20, states that by proper selection of solvent and diluent, a variety of polymer-gel structures of controllable porosity and pore-fineness can be obtained for a given polymer, and goes on to refer to the porosity and pore-fineness of the skin (lines 39, 40) and the grossly microporous structure consisting of large interconnected voids (lines 49, 50). On lines 54, 55 however the substructure is described as coarsely microporous. Column 5, lines 47 et seq. refers to the desirability of uniformity of pore size at the thin barrier layer (skin). The passage from column 5, line 52 to column 6, line 17 concludes that if highly porous, highly permeable, molecularly permselective membranes are to be produced by the precipitation process being discussed, it is necessary and desirable that the polymer selected should have a high glass transition temperature, or high level of crystallinity, or both. In column 6, lines 43 to 55, Michaels goes on to say that a structure will be produced which will be a highly anisotropic microporous membrane whose pore size varies with distance from the skin, being smallest in the skin and growing rapidly as the distance from the skin increases. The remainder of column 6 explains the conditions under which a non-porous skin may be formed and which are therefore presumably to be avoided when a porous skin is desired. Later on, in column 9, it is stated that the pore-structure of the membranes can be further modified by the addition of a solution-modifier, and/or by further moderate increases in the temperature of the casting and wash operations, and/or by changes in polymer concentration in the casting dope, the solution-modifiers giving rise to tighter or looser membranes. Also, if a given casting solution yields an impermeable membrane a decrease in polymer concentration usually results in a permeable microporous membrane (column 10, lines 36 to 43).

6. What kind of structure therefore is Michaels describing?  
It is true that "grossly microporous substructure consisting of large interconnected voids" might be describing the finger structure, but Michaels never refers to such a structure. Any other reference in Michaels appears less likely to be describing a finger structure and in particular column 6, lines 43 to 55 describes a structure which would seem to fall within the scope of the term reticulated.
  
7. During the opposition proceedings, the patentee submitted with the communication dated 28 July 1987 a document, the Simonetti Declaration, which purported to demonstrate that membranes according to the patent in suit were not obtainable by following the Michaels teaching. However Simonetti's experiments covered only a small part of the Michaels teaching, using only three polymers of which none were employed in the specific examples in Michaels and not investigating the effect of additives such as non-solvents on the casting solution. This document is therefore not seen as demonstrating that the Michaels membranes cannot have a reticulated structure.
  
8. It is true that Michaels refers consistently to a casting solution, whereas the membranes according to the patent in suit are obtained when a metastable dispersion is used for casting. Michaels does however refer to the addition of non-solvent to the casting solution (column 10, lines 48 to 56) and then states that any clarification of the casting dope need not be so complete as to cause the elimination of a Tyndall effect in a casting dope containing inorganic salts. Column 10, line 71 to column 11, line 2 points to the desirability of keeping the casting dope agitated between preparation and use and

suggests keeping it on a ball mill. This would seem to be an unlikely way of keeping a simple solution agitated and moreover is not unlike the treatment suggested in the patent in suit, column 5, lines 6 to 12, wherein segregated phases may be converted to the metastable state by redispersing, e.g. by agitation. The foregoing, while not in itself conclusive, does cast doubt on the Appellant's assertion that Michaels starts only from solutions and that the person of average skill reading Michaels would not start from a dispersion.

9. Kock refers to asymmetric membranes having a sponge structure ("Schwammstruktur"), therefore a reticulated structure and distinguishes such a structure from a finger structure. Any intermediate structure between sponge and finger structure may be formed - see e.g. pages 19, 44\* and 79. Kock does not use the term granular to describe the structure of the membrane but only, at one point (in respect of photograph 9b, page 20), to describe the membrane wall. It is true, as argued by the Appellant, that Kock is mainly concerned with reverse osmosis membranes (see the title of the dissertation) but there are many references to other types. Section 2.3.3 (page 12 et seq.) describes the use of asymmetric membranes having a skin and porous substructure in ultrafiltration and reverse osmosis, their suitability for these uses residing in the fact that the pores have their smallest radius at the surface, and as shown schematically in Fig. 3, the radius increases with distance from this surface. The final paragraph on page 13 also indicates that asymmetric membranes can function as porous membranes or non-porous membranes (Löslichkeitsmembran) and that any intermediate condition is possible. This is in keeping with the common general knowledge in the membrane filtration art that the operating ranges of the various types of membranes overlap

- see for example Table 2 on page 8 of Kock. Moreover pages 36, 37 of Kock disclose a membrane having a reticulated substructure (third paragraph in Fig. 17) and which filters Bacitracin (Table 6) therefore is in the ultrafiltration range. Kock also indicates, in the paragraph bridging pages 75 and 76, that the sponge structure is often the desired structure, a fact which would seem to be self-evident. Finally Kock gives a lot of information as to the conditions under which sponge or finger structures may be formed.

10. From the weight of the evidence of the disclosures in Michael and Kock the Board is persuaded that membranes having a porous skin and reticulated substructure belonged to the state of the art at the priority date of the patent in suit. The pore diameter range of the skin stated in Claim 1 corresponds to the size cut-off ranges for ultra- and microfiltration and are in part known from Michaels. In the opposition and appeal proceedings it has not been disputed that the bulk porosity required by Claim 1 is usual for ultra- and microfiltration membranes. Accordingly the subject-matter of Claim 1, while arguably novel, is not seen as involving an inventive step.
  
11. The disclosure in North Star, and the corresponding submissions of the parties, tends to confirm this conclusion. North Star is indeed concerned with reverse osmosis, but describes membranes which form a support for the reverse osmosis membranes proper in a composite assembly. The support membranes are asymmetric - cf. the description and electron micrographs of the front surface and back surface on pages 40 to 43, the pore diameter increasing from not more than 100 angstroms (0.01 micrometer) at the front surface to 7500 angstroms (0.75 micrometer) at the back surface. North Star also discloses that by adding non-solvent to the casting

solution, or by cooling the casting solution, support films which were improved in reverse osmosis performance were obtained. These are measures also employed in preparing the membranes according to the patent in suit; it is true however that North Star does not mention that metastable suspensions are cast.

The Respondent 02 (Hoechst) during the opposition proceedings submitted an experimental report (accompanying communication of 25 January 1988), in which two membranes were prepared according to North Star. REMs of cross-sections of these membranes were very similar to those of the patent in suit (Simonetti declaration). The Appellant, in an experimental report accompanying the communication of 8 January 1991, sought to refute this. The Hoechst experiments were repeated, as were other examples from North Star and the membranes obtained were compared with membranes according to the patent in suit as regards structure, flow rate, flow decay and pore size. Certainly some of the North Star membranes showed the finger structure and the Hoechst membranes were not identical to those of the patent in suit. Nevertheless the Hoechst membranes appeared to have a reticulated structure. Moreover from the flow rate information, the Hoechst membranes were applicable in the ultrafiltration range and those according to the patent in suit in the microfiltration range. (It will be recalled that the membranes according to Claim 1 cover both ultrafiltration and microfiltration.) Moreover this suggests that the Hoechst membranes had a more finely porous structure, so that like was not being compared with like. It appears to the Board that starting from a metastable dispersion is not a prerequisite for obtaining membranes with a porous skin and a reticulated substructure, but does give some guarantee that such a structure will be obtained. This however does not justify a claim to the membrane itself.

12. In the response of 8 January 1991, filed on 9 January 1991, the Appellant submitted that the term granular as used in the patent in suit and in the earlier proceedings was probably not appropriate to describe the prior art membranes having a structure which was not reticulated; the Appellant suggested that the prior art structures had a honeycomb structure consisting of closed cells with permeable walls. The Board can agree with the Respondents that this is speculative. No document apart from Wang suggests such a structure and the weight of the evidence is against it. The Wang structure appears to arise from the choice of a specific polymer (polyvinyl formal).
13. The Appellant, in the response of 8 January 1991, also suggested that Kock led away from the subject-matter of Claim 1, but this was in respect of process parameters and not the membrane per se.
14. Claims 2 to 5 according to the main request fall because they are appendant to a main claim which is not allowable. Moreover the Board can see no feature in these claims which would confer inventivity on the subject-matter of Claim 1. Claim 6, although formally independent, is a claim to a method of using the membrane according to Claims 1 to 5 which is a consequence of the membrane structure, and Claim 7 falls with Claim 6.
15. The same considerations apply to Claim 1 according to the first auxiliary request, which merely adds that the membrane is useful as an ultrafilter or microporous membrane.
16. Claim 1 according to the second auxiliary request adds to Claim 1 according to the first auxiliary request the feature relating to flow rate beginning "wherein" (see paragraph VIII above). This feature is either an automatic

result of the claimed structure of the membrane, or a restriction, in comparison with the first auxiliary request, to membranes having desired or advantageous flow rate parameter. In neither case can the feature be seen as contributing to an inventive step, and no arguments were submitted by the Appellant in this respect.

17. Claim 1 according to the third auxiliary request adds to Claim 1 according to the main request the feature relating to the water permeability. The same considerations apply to this feature as to the added feature of the second auxiliary request, and this request also is not allowable.

18. The same applies to the remaining claims of the auxiliary requests as to the corresponding claims of the main request, and in any case none of these claims was defended by the Appellant.

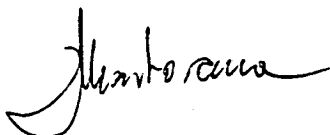
18. In coming to its decision, the Board did not require to consider the documents objected to by the Appellant (see paragraph VI above) therefore it was not necessary to consider whether the proceedings should be continued in writing.

Order

For these reasons, it is decided that:

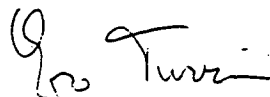
The appeal is dismissed.

The Registrar



P. Martorana

The Chairman



E. Turrini

BB P