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D E C I S I O N
of 28 November 1995

Case Number: T 0348/92 - 3.4.2

Application Number: 85304335.4

Publication Number: 0165077

IPC: B01D 71/68

Language of the proceedings: EN

Title of invention:
Sulfonated polysulfone composite semipermeable membranes and process for their production

Patentee:
NITTO DENKO CORPORATION

Opponent:
NORTH WEST WATER GROUP PLC

Headword:
-

Relevant legal provisions:
EPC Art. 56, 111(1)

Keyword:
"Inventive step - main request and auxiliary requests 1, 2, 3 and 5 - no"
"Auxiliary request 4 - remitted to Opposition Division"

Decisions cited:
-

Catchword:
-

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Case Number: T 0348/92 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 28 November 1995

Appellant: (Opponent)
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Decision under appeal: **Interlocutory decision of the Opposition Division of the European Patent Office dated 4 February 1992, posted 20 February 1992 concerning maintenance of European patent No. 0 165 077 in amended form.**

Composition of the Board:

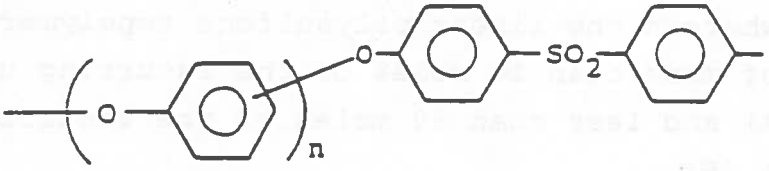
Chairman: E. Turrini
Members: C. Black
J.-C. Saisset

Summary of Facts and Submissions

I. The appeal lies against the decision of the Opposition Division to maintain European patent No. 0 165 077 (application No. 85 304 335.4) in amended form on the basis of claims and description as set out on EPO Form 2327.4 accompanying the decision.

II. Of these claims, independent claims 1 and 5 read as follows:

"1. A composite semipermeable membrane comprising a polysulfone series ultrafiltration support membrane on which is formed a film of a semipermeable membrane, characterized in that said semipermeable membrane is made of a partially sulfonated product of a polysulfone, which includes recurring units of the formula (A):



(A)

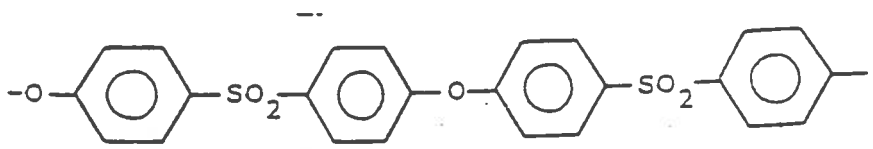
wherein n is an integer of 1 or more, and the sulfonated polysulfone has a logarithmic viscosity of 0.2 to 10 (measured at 30°C upon a solution of 0.5g thereof dissolved in 100 ml of N-methyl-2-pyrrolidone), and an ion-exchange capacity of 0.2 to 2.3 milli-equivalent/g.

"5. A process for producing a sulfonated polysulfone composite semipermeable membrane as claimed in any preceding claim, which comprises coating said polysulfone support membrane with a membrane-forming solution containing said sulfonated polysulfone, an alkylene glycol alkyl ether which may contain a small

amount of a non-protonic polar organic solvent, and a water-soluble and low volatile compound as an additive, and then evaporating off the solvent from the coated membrane-forming solution.

Dependent claims 3, 4 and 6 which are referred to in this decision reads as follows:

"3. A composite semipermeable membrane as claimed in claim 1 or 2, wherein the polysulfone is a copolymer which also contains a recurring unit of the formula (B):



(B)

"4. A polysulfone semipermeable membrane as claimed in claim 3, wherein the linear polysulfone copolymer is composed of more than 10 mole% of the recurring unit of formula (A) and less than 90 mole% of the recurring unit of formula (B).

"6. A process as claimed in claim 5, wherein the additive is at least one polyhydric alcohol, polyalkylene glycol, carboxylic or hydroxycarboxylic acid or salt of said acids.

III. The Opposition Division found that the claimed subject-matter was novel and involved an inventive step having regard to the disclosure in the following documents:

D1: EP-A-0 008 894
D4: Desalination, 41, 1 to 24 (1982)
D7: Office of Water Research and Technology, report 2001-20, August 1980

the other cited documents being deemed to be less relevant. The Division also found that the further ground of opposition under Article 100(b) EPC did not prejudice maintenance of the patent.

In particular it was argued that although D1 disclosed sulphonated polysulphones falling within the scope of those required by the patent in suit and indicated their possible use for the preparation of membranes, it was silent concerning the properties of the said polymers which would provide an incentive for the skilled person to substitute them for known sulphonated polysulphones in composite membranes.

IV. In addition to the above-mentioned documents D1, D4 and D7, the following documents have been considered during the appeal proceedings:

D3: Synthetic Membranes, Volume I, Desalination, pages 21, 305-307 and 324-326; ACS Symposium Series 153 (1981);
D5: GB-A-1 473 857;
D6: US-A-4 413 106;
D8: Pages 232-241 and 250-257 from file wrapper of US-A-4 818 387, corresponding with EP-A-0 165 077;
D9: US-A-4 054 707;
D10: Desalination, 18, 137-153 (1976);
D11: GB-A-1 495 887;
D12: Office of Water Research and Technology, report PB-167215 November 1980;
D14: NTIS Report No. PB-288 387 (dated March 1978)

V. Following an exchange of communications between the parties and the Board, oral proceedings were held. At the end of the oral proceedings the appellant (opponent) requested that the decision under appeal be aside and the patent revoked.

The respondent (patentee) requested that the appeal be dismissed and the patent maintained in amended form on the basis of claims according to a main request and five auxiliary requests presented at the oral proceedings.

VI. Claim 1 according to the main request corresponds to that set out in paragraph II above with the brackets enclosing the wording "measured...N-methyl-2 pyrrolidone" removed. Claim 1 according to auxiliary requests 1 and 2 incorporates the features of claims 3 and 4 respectively (see paragraph III above).

Claim 1 according to auxiliary request 3 has been reformulated as a product-by-process claim by incorporating the process features of claim 5.

Claim 1 according to auxiliary request 4 specifies the additive referred to in auxiliary request 3.

Claim 1 according to auxiliary request 5 specifies the thickness of the membrane in claim 1 of auxiliary request 1.

VII. The appellant's argumentation may summarised as follows:

At the priority date of the patent in suit, composite membranes consisting of a thin sulphonated polysulphone layer on a polysulphone support layer were known, as is evidenced by the various documents cited in the proceedings. To arrive at the subject-matter of claim 1 (main request) the skilled person has only to replace the known sulphonated polysulphone by those known from D1, and has every incentive to do so in view of the disclosed properties of the D1 sulphonated polysulphones and the statement in D1 that these are potentially useful as membrane materials, e.g. for ultra-filtration processes (sentence bridging pages 2 and 3).

The same considerations apply in respect of auxiliary requests 1 and 2.

Since the process steps introduced into claim 1 according to auxiliary request 3 are usual in the production of membranes, this claim is not inventive.

The additives which are the further distinguishing feature of claim 1 according to auxiliary request 4 have nothing in common with one another and it cannot be seen how these could support inventivity. In any case at least some of them are known from the cited prior art. The specified thickness of the membrane in claim 1 according to auxiliary request 5 is usual for composite membranes.

The gist of the respondent's counter-argumentation is as follows:

In the patent in suit the emphasis is on obtaining composite membranes with good chlorine and pH resistance and in the light of this the underlying problem is to be seen as the provision of such membranes while retaining the known desirable properties of high salt rejection etc.

D1 does not disclose composite membranes and gives no information about chlorine and pH resistance so that the skilled person has no incentive to replace the sulphonated polysulphane layer in known composite membranes by the polymers disclosed in D1.

Read as a whole, the other documents relied on by the appellant rather discourage the person of average skill in the art from further investigation of the use of sulphonated polysulphone on polysulphone in composite membranes. Chlorine resistance is either unsatisfactory

or not specified and more promising avenues are indicated (D4, D7, D12 and D14). On the other hand, later published documents demonstrate the superior properties of the claimed membranes, in particular high chlorine resistance over an extended period of time.

Reasons for the Decision

1. The appeal is admissible.

2. The Opposition Division, in point 2 of the reasons for the decision, found that the set of claims on which the decision was based met the requirements of Articles 123(2) and (3) EPC and the Board can agree with this finding. Moreover this applies also to the claims according to the requests at present under consideration, as will be evident from the indication of the basis of these claims in paragraph VI above in respect of the main request and auxiliary requests 1 to 4. The thickness of the membrane which is a feature of claim 1 according to auxiliary request 5 is to be found on page 5, lines 38 to 40 of the description of the granted patent.

3. *Novelty*

This is no longer an issue and will in any case be apparent from the discussion of inventive step.

4. *Inventive step*

4.1 Main request

In order to investigate inventive step it has first to be established what was state of the art at the priority date of the patent in suit. In this respect the Board,

in substantial agreement with the Appellant, is of the opinion that the field of the invention is relatively narrow and recent, and that there is a body of knowledge of which the average skilled person can be deemed to be aware, derivable from patents, articles in journals and research reports etc. rather than from text-books since this body of knowledge had not found its way to any great extent into text-books. In the present case the contents of documents D3 to D14 listed in paragraphs III and IV above has to be taken into consideration.

At this point it should be mentioned that the Board is now satisfied that the publication dates of all of these documents were before the earliest priority date of the patent in suit and this is no longer disputed by the respondent in view of the documentary evidence on file relating to the more contentious of these.

D3, dating from 1980, is a review article tracing the history of composite membranes. It refers to the original asymmetric membranes of Loeb and Sourirajan (1962) and the first cellulose acetate composite membranes followed by the first use of polysulphone as a preferred support material in 1966. It also emphasises the trend away from cellulosic membranes because of the biodegradability of the latter (pages 305 to 307).

D4 states on page 4 that a foremost objective has been the development of membranes with chlorine resistance and that to date (1981) this has not been achieved, as emphasised by the respondent. D4 goes on to refer to other desirable properties including thermal resistance, pH stability, resistance to compaction and bacterial attack and to the ideal of high productivity (usually described as high flux) coupled with high selectivity (or high rejection). As will be seen these represent conflicting requirements. On page 13 composite membranes

comprising sulphonated polysulphone on polysulphone are described, which were tested for the expected chlorine resistance. Disappointing results in this respect could be attributed at least in part to iron fouling, a problem also mentioned in D12. The respondent referred to the paragraph bridging pages 5 and 6 of D4 as directing the skilled person seeking chlorine resistance towards nitrogen bearing polymers (point 16 of response). In the Board's view this passage is only a statement of what some researchers have been doing, and later on page 6 it is stated that some membranes have shown irreversible degradation when exposed to chlorine. D5 describes composite membranes but is of less relevance for the present case.

D6 in Examples 6 and 8 discloses a composite hollow fibre membrane made by coating a porous polysulphone hollow fibre with sulphonated polyether sulphone. D7 also describes studies aimed at producing composite membranes which were resistant to chlorine and had acceptable flux and rejection values (see for example the abstract). Membranes comprising sulphonated polysulphone on polysulphone were selected because sulphonated polysulphone asymmetric membranes had already been shown to exhibit stability to chlorine, heat and pH (page 2).

The disadvantage of the cost of asymmetric membranes because of the cost of the sulphonation step is referred to (page 2). Composite membranes with the desired properties were obtained, but not reproducibly. An alternative approach, namely direct sulphonation of the surface of a polysulphone support, was abandoned because results were not encouraging and the degree of sulphonation could not be readily controlled.

D8 consists of extracts from the prosecution of the corresponding US patent and in the Board's opinion these are of only minor interest for the present case.

D9 relates to a novel sulphonated polyaryl-ether sulphone resin which find application in the manufacture of membranes. These may be composite membranes (column 3, lines 6 to 10) and the claimed polymers are stated to exhibit improved permeability in comparison with known sulphonated polysulphones.

D10 is an article dealing with sulphonated polysulphones and their application in the manufacture of membranes. It is mainly of interest in providing documentary evidence that the salt rejection efficiency increases with increasing exchange capacity (degree of sulphonation) whereas the flux, at least above a certain level decreases with increasing exchange capacity - see page 147.

D11 is a relatively old document (priority date May 1975) describing composite membranes wherein the active layer can be a sulphonates polysulphone - see e.g. claims 8, 9.

D12 is a report on investigations into seven chemical systems for preparation of hollow fibre composite membranes. These were subsequently narrowed down to three, of which one involved sulphonated polysulphones (page 2). Membranes with promising combinations of flux and rejection were obtained (page 19, first paragraph). However a module investigated for chlorine resistance fouled badly, apparently because of iron dissolved from the chlorine injection pump, so that the test was inconclusive.

D14 is a report on a research programme designed to develop chlorine-resistant composite membranes for sea-water desalination. Three types of polymer were investigated, of which sulphonated polysulphone was inferior to polypiperazine as regards salt rejection. It was however chlorine-resistant (page 2). The first sentence of the second paragraph on page 2 reads: "The most promising results on chlorine-resistant membranes have come through new approaches in the basic interfacial formation of polypiperazine-amide membranes." The respondent interprets this as meaning that the said membrane showed the best chlorine resistance. The Board does not agree with this interpretation and in its view the sentence would have been worded differently if this had been the intended meaning.

In D14 the approach was to sulphonate polysulphone until it became water-soluble, then to cross-link it **in situ** when coated on to a poly sulphone support. The cross-linked film was brittle and showed micro-cracks (page 21) and attempts to obviate this problem had only moderate success (pages 21 to 23). In the Board's opinion the skilled person would see no promise in this approach.

From the foregoing it can be derived that at the priority date of the patent in suit composite membranes consisting of polysulphone as the support layer and sulphonated polysulphone as the active layer were known and indeed this construction was one of the most preferred in view of the constituent layers.

The problem underlying the patent in suit can be seen as the general one of seeking to improve a known product by making a better one or making it more cheaply or more easily, This basic problem is the constant endeavour of

the average skilled person. The question to be answered is therefore whether the average skilled person, faced with the above-mentioned problem, would be led to replace the known sulphonated polysulphone layers by a layer produced from the sulphonated polysulphone layers known from D1.

Since D1 discloses the sulphonated product of a polysulphone having the repeating unit A required by claim 1 for the case that n equals 1 or of a polysulphone having the repeating units A and B required by claim 3, making such replacement would lead to the claimed subject-matter.

In the Board's view, in agreement with the appellant, the skilled person has every incentive to make this replacement.

In the first place D1, in the paragraph bridging pages 2 and 3, states that the hydrophilic polymers are potentially suitable as membrane materials, e.g. for ultrafiltration processes, e.g. desalination and removal of microorganisms, since they are not only hydrophilic in character but retain considerable strength even when containing a significant quantity of water. Secondly the polymers and copolymers in D1 are readily sulphonated with concentrated sulphuric acid instead of the more severe sulphonating agents disclosed in the prior documents and for the skilled person this is an indication that the cost disadvantage referred to in D7, page 2 may be overcome. Moreover the skilled person knows that the exchange capacity, therefor the degree of sulphonation, has to take into account the conflicting requirements of high flux and high rejection. D1 discloses that the degree of sulphonation can be readily adjusted by variation of the proportion of units A and B in the copolymer.

The remaining features of claim 1, that is, the logarithmic viscosity range and the ion-exchange capacity range, correspond to limits of operability, as is apparent from the description, page 3, lines 38 to 43 and 50 to 53 respectively and establishing these ranges falls within the competence of the average skilled person.

The respondent argues that the problem underlying the patent in suit should be seen as providing a composite membrane having improved chlorine and pH resistance while retaining the flux and retention properties of known membranes. D1 does not disclose composite membranes and says nothing about the chlorine and pH resistance of the disclosed sulphonated polysulhones, so that the skilled person has no reason to take its teaching into account.

In the Board's view however, since it has been shown to be obvious for the skilled person to replace the known sulphonated polysulphones by those disclosed in D1 in the knowledge that a composite membrane can thereby be produced more readily and cheaply with controlled ion-exchange capacity, the chlorine and pH resistance is to be seen as a bonus. In any case there is nothing surprising about the chlorine resistance. As pointed out above, D7 on page 2 refers to the desirable properties of sulphonated polysulphones used as asymmetric member materials, including stability to chlorine and extremes of pH. It is true that this has to be qualified in the light of D4 and D12. However the apparent sensitivity to chlorine disclosed in these documents is attributed at least in part to iron fouling resulting from iron dissolved from chlorine injection pumps.

The respondent also drew attention to D14 wherein composite membranes comprising sulphonated polysulphone as active layer are stated to be inferior to those using polypiperazine-amide membranes. This inferiority however can be attributed to the particular method employed, that is, sulphonating until the polysulphor is water-soluble then cross-linking the coated layer to impart water-insolubility. Moreover D14 bears the date 1978 and the fact that in D4, D7 and D12 investigations were continuing into the use of sulphonated polysulphone on polysulphone in 1980 and 1981 indicates that no prejudice arose from the disclosure in D14.

The respondent observed that in D7 the optimistic objectives were not reflected in the results obtained. As noted above, one approach in D7 was direct sulphonation of the surface of a polysulphone support and this was abandoned. It cannot be seen as an alternative approach which might have led the skilled person away from the solution of the patent in suit. In the case of composite membranes comprising sulphonated polysulphone on polysulphone there was a problem with reproducibility, but in the Board's experience reproducibility was a recurring problem in the membrane field whose solution usually involved rigid control of process parameters. In the Board's view the disclosure in D7 would not deter the skilled person from investigating further sulphonated polysulphone membranes. Finally the respondent cited three post-published documents with a view **inter alia** to demonstrating the superior chlorine resistance of the membranes according to the patent in suit. As indicated above the Board's opinion that the said chlorine resistance is not surprising. In any case in these documents comparison is made with cellulose acetate and interfacial polymerisation membranes but not with previously known sulphonated polysulphone membranes.

The subject-matter of claim 1 according to the main request therefore does not involve an inventive step.

Auxiliary requests 1 and 2

Claim 1 according to these requests relates to the use of the copolymers disclosed in D1 and to specific proportion ranges thereof which are disclosed **inter alia** in Example 5 thereof. The restrictions add nothing inventive to the subject-matter of claim 1 but fall rather within the competence of the skilled person seeking to adjust the ion-exchange capacity of the membrane material.

Auxiliary request 3

Claim 1 according to this request includes process features taken from granted claim 5. These features are usual in the manufacture of composite membranes. In particular 2-methoxyethanol, an example of an alkylene glycol alkyl ether, is a known solvent for sulphonated polysulphones to be coated on to a support - see D7 (abstract), D10 (page 143) and D11 (Example 1). The co-solvent is optional and serves to aid solution of the polymer (page 4, lines 36 to 40). This is a measure which falls within the competence of the skilled person. As to the requirement that the optional co-solvent is aprotic it is noted that dimethyl formamide is an example thereof (page 4, line 41) and is also a known solvent for sulphonated polysulphones (see e.g. D9, sentence bridging columns 3 and 4 and D10, page 143). For the average skilled person dimethyl formamide would therefore be a co-solvent of choice.

The claim also requires a water-soluble low volatile additive. Additives are common in film-forming polymer formulations and in the absence of any indication as to its role, the simple mention of the additive cannot support the presence of an inventive step.

Auxiliary request 5

Claim 1 according to this request differs from that according to the main request in that the thickness of the semipermeable membrane is specified as being in the range of 0.01µm to 5µm. D3 discloses a thickness range for the active layers of a composite membrane of 0.02µ to 5µm (page 306) and D9 a range of 0.1µm to 10µm (column 3, line 8) so that also this feature does not contribute to inventivity.

Auxiliary request 4

In comparison with auxiliary request 3, the additive is exemplified. At oral proceedings during the opposition proceedings a similar auxiliary request was filed - see Annex III to the minutes of the proceedings. However the Opposition Division, once it was minded to allow the patentee's main request, did not have to consider this request. The Board notes that the opponent had commented on the said additives in connection with claim 8 then under consideration (page 9 of the opposition grounds) and again in the communication dated 2 January 1992 (pages 17, 18) but from the file it appears that the Opposition Division did not have to address this question. The Board, having heard the opposing arguments of the parties, has concluded that the matter requires further investigation, and to this end, makes use of its power under Article 111(1) to remit the case to the Opposition Division for further prosecution so as to avoid a loss of instance for either party.

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 auxiliary request 4.

The Registrar: The Chairman:

P. Martorana E. Turrini