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Datasheet for the decision of 28 June 2007

Case Number: T 0571/05 - 3.3.07

Application Number: 98931794.6

Publication Number: 0994747

IPC: B01D 67/00

Language of the proceedings: EN

Title of invention:
Highly asymmetric ultrafiltration membranes

Applicant:
PALL CORPORATION

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54, 123

Keyword:
"Amendments - allowable (yes) (Main Request)"
"Interpretation of the term "substantially free of macrovoids" in Claim 1"
"Novelty - (no) (Main Request, First to Third Auxiliary Requests)"

Decisions cited:
-

Catchword:
-

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Case Number: T 0571/05 **-** 3.3.07

D E C I S I O N
of the Technical Board of Appeal 3.3.07
of 28 June 2007

Appellant: PALL CORPORATION
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted on 14 December 2004 refusing European application No. 98931794.6 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: S. Perryman
Members: G. Santavicca
F. Rousseau

Summary of Facts and Submissions

I. The appeal lies from a decision of the Examining Division refusing European patent application 98 931 794.6, which originates from International application PCT/US98/13910 (publication N° WO 99/02246) claiming a priority of 8 July 1997. Independent Claims 1 and 18 as filed read as follows:

"1. An asymmetric integral polymer membrane for ultrafiltration, comprising:

a skin face, an opposite face, and a porous support between said skin face and said opposite face, said skin face having skin pores, said skin pores generally having a diameter of sufficient size to permit passage of a liquid therethrough while excluding passage of a macromolecule therethrough, said porous support comprising a substantially continuous reticular mass defining interconnecting flow channels communicating with said skin pores, said flow channels defining support pores having diameters generally increasing gradually in size from said skin face to said opposite face, to an average diameter at said opposite face of at least about 20 times the diameter of said skin pores, and said porous support being substantially free of voids materially larger in diameter than said gradually increasing support pores."

"18. A method of preparing an asymmetric integral polymer membrane for ultrafiltration, comprising the steps of:

providing a casting dope comprising a hydrophobic polymer, a solvent, and a nonsolvent, in ratios

sufficient to form a homogeneous solution or a colloidal dispersion;

casting said dope to form a thin film;

coagulating said thin film in a quench bath; and

recovering an ultrafiltration membrane having an asymmetry ratio of at least about 20, said membrane being substantially free of macrovoids.".

II. The decision under appeal was based on amended Claims 1 to 11 enclosed in the applicants' letter dated 2 August 2004 but received on 29 October 2004 by facsimile, as the Main (and sole) Request, Claim 1 reading as follows (emphasis added by the Board to show the amendments compared to Claim 1 as filed):

"1. An asymmetric integral **sulfone** polymer membrane for ultrafiltration, comprising:

a skin face, an opposite face, and a porous support between said skin face and said opposite face, said skin face having skin pores, said skin pores generally having a diameter of sufficient size to permit passage of a liquid therethrough while excluding passage of a macromolecule therethrough, said porous support comprising a substantially continuous reticular mass defining interconnecting flow channels communicating with said skin pores, said flow channels defining support pores having diameters generally increasing gradually in size from said skin face to said opposite face, to an average diameter at said opposite face of at least about 20 times the diameter of said skin pores, and said porous support being substantially free of **macrovoids**."

According to the decision, the presence of macrovoids in the membrane obtained by the appellants upon the repetition of Example VII of D1 was not sufficient to demonstrate that a membrane substantially free of macrovoids could not be prepared in the whole range of casting compositions, casting temperatures and quench temperatures disclosed in D1. Hence, the membrane defined in Claim 1 lacked novelty over the disclosure of D1. Since the additional features in dependent Claims 2 to 11 either were known from D1 or did not produce any special effects justifying the presence of an inventive step, the application was to be rejected.

III. The applicants lodged an appeal against that decision (notice received on 14 February 2005). In the statement setting out the grounds of appeal, received on 14 April 2005, the appellants requested to grant a patent on the basis of the claims then on file.

Then, in reply to a communication in preparation for oral proceedings, in which the Board had inter alia made comments on the basis and the clarity of Claim 1, in particular on the clarity of the terms "macrovoids" and "substantially free", the appellants submitted some papers and extracts from the Internet concerning the term "macrovoid" as well as three sets of amended Claims 1 to 8 as First, Second and Third Auxiliary Request, respectively (facsimile of 15 June 2007). Claim 1 according to these Auxiliary Requests reads respectively as follows (emphasis added by the Board to show the amendments to the claims as filed):

First Auxiliary Request

"1. An asymmetric integral **sulfone** polymer membrane for ultrafiltration, comprising:

a skin face, an opposite face, and a porous support between said skin face and said opposite face, said skin face having skin pores, said skin pores generally having a diameter of sufficient size to permit passage of a liquid therethrough while excluding passage of a macromolecule therethrough, said porous support comprising a substantially continuous reticular mass defining interconnecting flow channels communicating with said skin pores, said flow channels defining support pores having diameters generally increasing gradually in size from said skin face to said opposite face, to an average diameter at said opposite face of at least about 20 times the diameter of said skin pores, and said porous support being free of **macrovoids**."

Second Auxiliary Request

"1. An asymmetric integral **sulfone** polymer membrane for ultrafiltration, comprising:

a skin face, an opposite face, and a porous support between said skin face and said opposite face, said skin face having skin pores, said skin pores generally having a diameter of sufficient size to permit passage of a liquid therethrough while excluding passage of a macromolecule therethrough, said porous support comprising a substantially continuous reticular mass defining interconnecting flow channels communicating with said skin pores, said flow channels defining support pores having diameters generally

increasing gradually in size from said skin face to said opposite face, to an average diameter at said opposite face of at least about 20 times the diameter of said skin pores, and said porous support being substantially free of voids materially larger in diameter than said gradually increasing support pores."

Third Auxiliary Request

"1. An asymmetric integral **sulfone** polymer membrane for ultrafiltration, comprising:

a skin face, an opposite face, and a porous support between said skin face and said opposite face, said skin face having skin pores, said skin pores generally having a diameter of sufficient size to permit passage of a liquid therethrough while excluding passage of a macromolecule therethrough, said porous support comprising a substantially continuous reticular mass defining interconnecting flow channels communicating with said skin pores, said flow channels defining support pores having diameters generally increasing gradually in size from said skin face to said opposite face, to an average diameter at said opposite face of at least about 20 times the diameter of said skin pores, and said porous support being free of voids materially larger in diameter than said gradually increasing support pores."

IV. Oral proceedings took place on 28 June 2007, in the absence of the appellants, as announced by facsimile of 27 June 2007, in compliance with Rule 71(2) EPC.

V. The appellants had argued as follows in writing:

(a) It was an essential feature of the invention that the porous support of the asymmetric membrane for ultrafiltration was substantially free of macrovoids.

(b) The term "macrovoids" was a term of art, as shown by the papers and extracts from the Internet.

(c) D1 did not mention any absence of macrovoids in the membranes disclosed.

(d) Examples V, VII and VIII of D1, in which polysulfone was used, were repeated by Dr I-fan Wang. The membrane obtained from the repetition of Example VII was suitable for ultrafiltration but contained macrovoids, as shown in the SEM image submitted with letter dated 2 August 2004. In contrast, the membranes from the repetition of Examples V and VIII were suitable for only microfiltration, in line with the statement in the application as filed, and not for ultrafiltration. That these two examples showed no macrovoids was thus irrelevant.

(e) Therefore, D1 neither explicitly nor implicitly disclosed ultrafiltration membranes that were free of macrovoids.

(f) In view of the repetition of Example VII of D1, it could not be argued that the absence of any comments on macrovoids in D1 amounted to an

implicit disclosure that the ultrafiltration membranes of D1 were free of macrovoids.

(g) Since the only example of D1 concerning polysulfone ultrafiltration membranes had macrovoids, the skilled person would conclude that the polysulfone ultrafiltration membranes would all suffer from the same problem, i.e. that D1 did neither address nor solve that problem.

(h) In fact, since, when using the method of D1, the skin of the membrane formed quickly, that skin slowed down the non-solvent diffusion into the membrane, so that macrovoids were formed. Instead, in the invention defined in the claims under appeal, a particular solvent system was used, which comprised a Lewis acid such as propionic acid and a Lewis base such as N-Methyl-Pyrrolidone (NMP) in the form of a complex, which complex was readily dissociated by an aqueous medium, thus forming a macrovoid-free structure.

(i) Therefore, the claimed subject-matter was novel over the disclosure of D1.

(j) This had also been acknowledged by the USPTO by the allowance of two US cases, corresponding to the application under appeal, over documents equivalent to D1.

VI. The appellants had requested in writing that the decision under appeal be set aside and that a European patent be granted on the basis of the Main Request underlying the decision under appeal or on the Basis of

the Claims of the First, Second or Third Auxiliary Request as set out in the facsimile of 15 June 2007.

Reasons for the Decision

1. The appeal is admissible.

Main Request

2. *Amendments*

2.1.1 Compared to Claim 1 of the application as filed, Claim 1 according to the Main Request underlying the decision under appeal comprises the following amendments (emphasis added by the Board):

(a) "integral **sulfone** polymer";

(b) "said porous support being substantially free of **macrovoids**" (instead of the wording "said porous support being substantially free of voids materially larger in diameter than said gradually increasing support pores" used in the application as filed).

2.1.2 The remaining dependent claims concerning the membrane have been adapted to new Claim 1 in view of the deletion of the additional features of Claims 3-4 and 6-9, which concerned method aspects.

2.1.3 As regards the claims as filed concerning the method of manufacture of the membrane, they have been cancelled.

2.1.4 The first amendment mentioned (sulfone polymer) is disclosed in Claim 4 as filed, which is dependent on Claim 1 via Claim 3, which specifies the composition used in the process of preparation of the membrane.

2.1.5 The second amendment (substantially free of macrovoids) has a basis as such in independent Claim 18 as filed, which concerns the method of preparing an asymmetric integral polymer membrane.

2.1.6 Therefore, the amended claims of the Main Request fulfil the requirements of Article 123(2) EPC.

2.2 *The invention defined in Claim 1*

2.2.1 Claim 1 of the Main Request ***inter alia*** contains the term "macrovoids" in the amended feature "substantially free of macrovoids".

2.2.2 The Board does not dispute that "macrovoids" is a term of art. The question is that it has not been shown that the term of art "macrovoids" means a clear, definite and well recognised range of void sizes in the context in which it is used, i.e. preparation and use of membranes for ultrafiltration.

2.2.3 As regards the term "macrovoids", the application as filed defines the "macrovoids" as being "large voids", which "are finger-like projections in the support structure that generally do not communicate with the pores in the skin surface" (page 2, lines 13 to 19). However, the application as filed does not disclose the size range meant nor any methods by which it should be measured. Hence, the question "how large should a void

be to be a macrovoid" has no definite answer from the application as filed.

2.2.4 If the term "macrovoids" were taken as equivalent to the definition present in Claim 1 as filed, namely "voids materially larger in diameter than said gradually increasing support pores", further questions would arise, such as:

(a) Where is the gradually increasing support pore to be measured? At its maximum section? Since Claim 1 as filed does not define any average maximum pore diameter (neither on the skin surface, nor on the opposite face), the maximum pore diameter might be so large that only "**mega**voids" are excluded?

(b) When is a large void "**materially** larger" in diameter than said gradually increasing support pores? Might it need to be twice as large, or thrice as large or even larger?

(c) As regards the feature "substantially free", what is the volume, or number, or density, or whatever else amount of macrovoids which can be tolerated, i.e. what is not substantial? And what is the method or the protocol that has to be used for determining with certainty whether that substantiality of freedom from macrovoids is fulfilled? Or should a particular effect thereof, such as the performance of the membrane be evaluated? And if so how?

2.2.5 There is no answer to those questions, either in the application as filed or in the arguments and evidence put forward in writing by the appellants.

2.2.6 In the absence both of precise definitions of "substantially free" and "macrovoids" and an indication of how the presence of macrovoids is to be determined, and what numerical values so determined should not be exceeded, the skilled reader is at best left with the impression that an undesirably large number of undesirably large voids should not be present. The skilled reader is left to exercise his own subjective judgement as to what is meant.

2.2.7 The further guidance is provided to the skilled reader concerning the disadvantages if macrovoids are present. According to the application under appeal as filed, since fluid entering a macrovoid was trapped and could not be filtered, macrovoids added to the membrane's resistance to fluid flow, leading to undesirably low flow rates without any concomitant benefit in effectiveness of filtration. Therefore, while a macrovoid-ridden support structure might provide mechanical stability to the skin, that configuration did not result in optimal UF membrane performance (page 2, lines 15 to 19). Furthermore, the application under appeal mentions that "without macrovoids, the dead space within the membrane was significantly reduced if not eliminated, and flux rates were improved over prior MF membranes" (page 3, lines 2 and 3). Finally when commenting the results of Examples 1 to 8, the application under appeal inter alia mentions that the ultrafiltration membranes exemplified had high flow rates, such as 0.658 and 0.395 cm/min psi (Examples 1

and 2), and even a selective 30k membrane had a flow rate higher than 0.153 cm/min psi (page 13, lines 25 to 33). These high flow rates were the consequence of the high asymmetricity of the membrane and of the absence of macrovoids in the membranes (page 14, lines 1 and 2). According to Claim 16 as filed, the minimum flow rate should be 0.125 cm/min psi.

2.2.8 The reader could thus assume that the term "substantially free of macrovoids", if anything, could only have a functional meaning as given in the above passages in the application as filed.

3. *Novelty*

3.1 D1 discloses an improved highly asymmetric polymeric membrane comprising a skin and a porous support, said skin containing pores which have an average pore diameter of from about 0.005 to about 3.0 microns and said support comprising a reticulated structure which contains pores which have pore sizes ranging from about 10 to about 20,000 times as large as the average pore diameter of the pores of said skin, said membrane having a bulk porosity greater than about 70% (Claim 1).

3.2 The polymer of the membrane can be polyarylsulfone (Claims 2, 15 and 18). When a polysulfone polymer is used to prepare the membrane, the concentration of said polymer in the polymer-dope can be from about 6 to about 13 by weight of the casting dope (Claim 19).

3.3 Hence, D1 explicitly defines ultrafiltration polysulfone polymer membrane having all of the features of Claim 1 under appeal but contains no information as

to whether its porous support is "substantially free of macrovoids".

3.4 D1 does not mention whether or not macrovoids are present in the porous support, neither for microfiltration nor for ultrafiltration membranes. However, in the absence of any mention of defects such as macrovoids in D1, it cannot be assumed that defects are got. This question of fact has to be decided on the basis of all of the elements disclosed in D1.

3.5 In particular, according to D1, a significant advantage of the membranes was their high fluid permeability, particularly for small pore sizes. This was believed to be the result of the very high asymmetry so that the reticulated parts of the membrane offered a relatively low resistance to fluid flow as compared to the finely porous skin. For example, a membrane prepared according to D1 as described in its Example VI (comprising polysulfone and polyamide) had a pore size of 0.01 µm (hence, it was suitable for ultrafiltration) and a flow rate of 0.9 cm/min psi, one hundred times higher than that of commercial membranes then available having that pore size (D1, page 16, lines 12 to 23).

3.6 Since the UF membranes disclosed by D1 are as asymmetric as are the UF membranes claimed in the application under appeal, and since the flow rates of the UF membranes disclosed by D1 are, if not better (0.9 cm/min psi), as high as those exemplified in the application under appeal, it must be concluded in view of the above that the UF membrane disclosed by D1 must possess all of the features of the membranes defined in the claims under appeal, including the "substantial

freedom of macrovoids" in the support of the ultrafiltration membranes.

3.7 It follows from the above that the mere stating in the statement setting out the grounds of appeal (point 11) that, as illustrated in the SEM filed on August 2004, in the membrane obtained from the reproduction of Example VII of D1 "macrovoids are clearly present" cannot be compared clearly to "substantially free of macrovoids", and thus is not sufficient to distinguish over D1.

3.8 Consequently, the claimed subject-matter is not novel over the disclosure of D1.

Auxiliary requests

4. Each of the First, Second and Third Auxiliary Request contains a definition of what voids have to be excluded, either by the term "free of macrovoids" or by the feature present in Claim 1 as filed "materially larger than".

4.1 Apart that those definitions for the sought-for exclusion lack definiteness, as explained above (points 2.2, ***supra***), use of different terms meant by the appellant to have the same meaning cannot change the reasoning on lack of novelty over D1.

4.2 In view of the above the claimed subject-matter as claimed has been found to read on the membranes disclosed by D1 (points 3, ***supra***).

4.3 Therefore, each of First, Second and Third Auxiliary Request fails for the reasons given in connection with the Main Request.

5. Consequently, no European patent can be granted on those requests.

6. In view of this decision the Board need not decide whether the claims of the Auxiliary Requests are formally allowable.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

D. Sauter S. Perryman