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Datasheet for the decision of 03 April 2009

Case Number: T 1934/07 - 3.3.07

Application Number: 01983289.8

Publication Number: 1349644

IPC: B01D 69/02

Language of the proceedings: EN

Title of invention:
Modified membranes

Applicant:
Siemens Water Technologies Corp.

Headword:
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Relevant legal provisions:
-

Relevant legal provisions (EPC 1973):
EPC Art. Art. 54, 84, 111(1), 123(2)
EPC R. 80

Keyword:
"Amendments - added subject-matter (yes) (Main Request)"
"Amendments - allowable (yes) (Auxiliary Request)"
"Novelty (yes) (Auxiliary Request)"
"Remittal (yes)"

Decisions cited:
-

Catchword:
-

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Case Number: T 1934/07 **-** 3.3.07

D E C I S I O N
of the Technical Board of Appeal 3.3.07
of 03 April 2009

Appellants: Siemens Water Technologies Corp.
181 Thorn Hill Road
Warrendale, PA 15086 (US)

Representative: Grünecker, Kinkeldey,
Stockmair & Schwanhäusser
Anwaltssozietät
Leopoldstraße 4
D-80802 München (DE)

Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 22 May 2007 refusing European application No. 01983289.8 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: S. Perryman
Members: G. Santavicca
B. ter Laan

Summary of Facts and Submissions

I. The appeal lies from a decision of the Examining Division refusing European patent application 01 983 289.8, which originates from International application PCT/AU01/01450 (publication N° WO 02/38256) concerning "Modified Membranes".

II. The application as filed contains 69 claims, the independent claims thereof reading as follows:

"1. A porous polymeric membrane formed from a blend of a polymeric membrane forming material and a polymeric reactivity modifying agent adapted to modify the surface active properties of the porous polymeric membrane relative to a porous polymeric membrane formed from the polymeric membrane forming material alone."

"35. A method of preparing a porous polymeric membrane wherein the polymeric reactivity modifying agent is blended in the surface active porous polymeric membrane by incorporation into the bulk material."

"56. A method of modifying the surface of a porous polymer membrane including:
i) blending a polymeric reactivity modifying agent with a polymeric membrane forming material and
ii) forming a modified membrane."

"62. A porous polymeric membrane when formed by a method of any one of claims 35 to 56."

"64. A blend of a membrane forming polymer and a compatible second polymer, said second polymer being

capable of chemical modification after membrane formation.".

III. The decision under appeal was based on a set of amended Claims 1 to 60 filed with letter dated 6 March 2007, Claim 26 reading as follows:

"26. A porous polymeric membrane formed from a blend of a polar polymer as a polymeric membrane forming material and poly(alkyl vinyl ether/maleic anhydride).".

According to that decision:

(a) D8 (US-A-4 707 266) disclosed the use of a polyamide as a membrane forming polymer, which was cast together with a copolymer of methyl vinyl ether and maleic anhydride (Gantrez(TM)), to modify the surface properties;

(b) polyamide was known to be a polar polymer, thus fell under the definition given in Claim 26;

(c) the term "blend", in Claim 26, did not exclude that, at least to a certain extent, some bond was formed by further reaction between the polar polymers of the blend;

(d) therefore, the subject-matter of Claim 26 lacked novelty;

(e) the application was thus refused.

IV. The applicants lodged an appeal against that decision. In their statement setting out the grounds of appeal, the appellants submitted a new Main Request as well as an Auxiliary Request.

In a communication in preparation for the oral proceedings, the Board drew attention to the amendments

made to the claims, in particular in relation to the requirements of Articles 84 and 123(2) EPC.

In response to the communication of the Board, the appellants submitted, on the one hand, copies of a number of dictionaries and standards, to show that the meaning of the terms mentioned in the claims was known and well recognised in the art, and certain particulars taken from product details of Gantrez(TM) ES 225 and MS 955 (letter dated 5 March 2009). On the other hand, the appellants replaced the requests then on file with a fresh Main Request and First to Fifth Auxiliary Requests.

Claim 1 of the Main Request reads as follows (the amendments to Claim 64 as filed are indicated as follows: deletions in strikethrough, additions in bold):

"1. A blend of a**n inert polar polymeric** membrane forming ~~polymer~~ **material** and a compatible ~~second~~ polymer **that is poly (alkyl vinyl ether/maleic anhydride)**, ~~said second polymer being~~ **wherein the poly (alkyl vinyl ether/maleic anhydride) is** capable of chemical modification after membrane formation.".

V. Oral proceedings took place on 3 April 2009. After a discussion on the allowability of the amendments in the claims of the Main Request, in particular of the term "polar inert", the appellants submitted a set of 16 amended claims as the sole Auxiliary Request, replacing all of the previous auxiliary requests.
The only independent claim of the Auxiliary Request reads as follows (the amendments to Claim 1 as filed

are indicated as follows: deletions in strikethrough, additions in bold):

"1. A porous polymeric membrane formed from a blend of a polymeric membrane forming material **that is PVDF or a polysulfone selected from the group consisting of polysulfone per se polyether sulfones, polyaryl sulfones, polyalkyl sulfones and polyaralkyl sulfones,** and a polymeric reactivity modifying agent adapted to modify the surface ~~active~~ properties of the porous polymeric membrane relative to porous polymeric membrane formed from the polymeric membrane forming material alone, **said agent being poly(alkyl vinyl ether/maleic anhydride), wherein the reactivity modifying agent is added to the polymeric membrane forming material before the membrane is cast**.".

VI. The appellants essentially argued as follows:

Main Request

(a) The amendments were based on the application as filed so that the amended claims fulfilled the requirements of Article 123(2) EPC.

(b) The terms such as "inert", "polar", "surface active" and "blend" were clear, as shown in the documents submitted, such as the excerpts of dictionaries. Also, the term "inert polar" was clear. As to "blend", the description contained a specific definition because the invention relied on the discovery that copolymers of alkyl vinyl ether and maleic anhydride (Gantrez$^{(TM)}$) were miscible with polar polymers such as

polyvinylidenedifluoride (PVDF) and polysulfones, so that the casting dope was a blend.

(c) Hence, the amended claims of the Main Request were allowable.

Auxiliary Request

(a) The amendments were based on the claims of the application as filed so that the amended claims fulfilled the requirements of Article 123(2) EPC.

(b) The terms to which objections had been raised, such as "inert polar" and "surface active", had been removed from the claims, in line with the claims as filed, so that the amended claims were also clear (Article 84 EPC).

(c) As to novelty:

D8 disclosed the reaction of a polyamide membrane forming polymer with a membrane surface modifying copolymer of maleic anhydride with methyl vinyl ether (Gantrez(TM) S-97) to form a cross-linked surface layer. Since Gantrez(TM) S-97 was a fully hydrolized copolymer, it was immiscible with the polyamide membrane forming polymer, so that no blend could be formed. Hence, D8 did not disclose the claimed combination of a membrane forming polymer such as PVDF or polysulfone [which were inert to, i.e. did not react] with a poly(alkyl vinyl ether/maleic anhydride) as a reactivity modifying agent, let alone that the membrane was cast from a blend.

The membrane disclosed by D9 contained a specific combination of layers such as a porous support, an adhesive layer and a diffusive polymeric layer, which specific combination was not cast from one polymeric blend.

None of the membranes disclosed by any of the further documents cited in the examination proceedings, namely D1 (EP-A-0 463 627), D2 (WO-A-99/59707), D3 (EP-A-0 911 073), D4 (US-A-5 531 900), D5 (EP-A-0 430 082) and D6 (WO-A-99/01207), were made from a blend of PVDF or polysulfone as the membrane forming polymer with, as a reactivity modifying agent, a poly(alkyl vinyl ether/maleic anhydride).

D7 was identical to D3.

(d) The subject-matter of Claim 1 was thus novel having regard to D8 or to any of D1 to D6 and D9.

(e) The claimed subject-matter involved an inventive step over D8 as the closest prior art document even if combined with any of D1 to D6 and D9.

(f) Thus, a European patent should be granted.

VII. The appellants requested that the decision under appeal be set aside and that the case be remitted to the first instance for further prosecution on the basis of the claims of the Main Request submitted on 5 March 2009 or of the Auxiliary Request submitted at the oral proceedings on 3 April 2009.

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 *inter alia* comprises the amendment "polar inert" in the definition of the membrane forming polymer.

2.1.2 The expression "polar inert" as such is neither mentioned in the claims as filed nor described elsewhere in the application as filed.

2.1.3 As regards the term "inert", the description mentions:

(a) that the polymeric membrane forming material should preferably be of low reactivity relative to the reactivity modifying agent (page 6, lines 10 and 11); and,

(b) that the polymeric membrane forming material may in some cases desirably be inert (page 6, lines 11 and 12).

From the above passages it can be gathered that, in general, a higher reactivity was not excluded and that the choice of an inert polymeric membrane forming material was desirable in some cases, which however have not been described further.

2.1.4 As to the term "polar", it is mentioned in several parts of the description concerning e.g. the copolymer

of methyl vinyl ether and maleic anhydride (Gantrez(TM)) to indicate, respectively:

(a) the polar moiety represented by the maleic anhydride as well as the polar polymeric free acid obtained after dissolution in e.g. water (page 11, lines 1 and 8 to 10); or,

(b) that the copolymer was miscible with polar polymers such as polysulfones and PVDF (page 12, lines 20 and 21); and,

(c) that the known mixtures of two polymeric membrane forming materials which were not miscible, and thus required stabilization by cross-linking, i.e. by a chemical reaction, were not true "blends" as in the present application.

From the above passages it can be gathered that PVDF and polysulfones are polar polymers, which fact was however known and is not in dispute, and that the mixtures containing two cross-linked (i.e. reacted) membrane forming polymers were not blends. However, these passages do not disclose a generic inertness between polar polymers, not necessarily PVDF or polysulfone, and modifying agents (not necessarily a second membrane forming material) such as a poly(alkyl vinyl ether/maleic anhydride), so that they give no guidance on what is to be understood by "inert polar" now put forward in the claims.

2.1.5 It follows from the above that the expression "inert polar" has never been associated with any membrane forming material in combination with any poly(alkyl vinyl ether/maleic anhydride), as defined in Claim 1 of the Main Request, so that it has no direct and unambiguous basis in the application as filed (Article 123(2) EPC). Nor can any clear meaning be

attributed to the ***prima facie*** self contradictory expression "inert polar" (Article 84 EPC).

2.2 Therefore, Claim 1 of the amended claims of the Main request does not fulfil at least the requirements of Articles 84 and 123(2) EPC.

2.3 The Main Request is not allowable.

Auxiliary Request

3. *Amendments*

3.1 Compared to Claim 1 as filed, Claim 1 of the Auxiliary Request comprises the following amendments, each having a basis in the indicated passages of the application as filed:

3.1.1 "that is PVDF or a polysulfone selected from the group consisting of polysulfone per se polyether sulfones, polyaryl sulfones, polyalkyl sulfones and polyaralkyl sulfones". These features were defined in Claims 2 and 5 as filed, both referring to Claim 1 as filed;

3.1.2 "said agent being poly(alkyl vinyl ether/maleic anhydride)". This feature was defined in Claim 12 as filed, which referred to Claim 1 as filed;

3.1.3 "wherein the reactivity modifying agent is added to the polymeric membrane forming material before the membrane is cast". This feature is a process feature that was defined as such in Claim 37 as filed. Although Claim 37 as filed concerns a preferred embodiment of the method of preparation as defined in Claim 35 as filed, the

latter gives a generic definition of the modified membrane according to Claim 1 as filed by its method of preparation.

3.1.4 "surface ~~active~~ properties". This deletion is based not only on the wording of Claims 7 ("the reactivity modifying agent modifies the reactivity of the membrane surface to ...) and 56 as filed ("A method of modifying the surface ...") but also on many passages of the description as filed (page 4, lines 22 and 23; page 7, lines 1 and 12 to 14; page 8, lines 11 and 12; page 9, lines 16 to 24; page 14, line 5), from which it is apparent, on the one hand, that the term "surface active" indeed simply means the surface of the membrane, and on the other hand, that it particularly refers to those groups (anhydrides) of the modifying agent incorporated into the membrane (poly(alkyl vinyl ether/maleic anhydride)) that are susceptible of chemical modification. Since that modifying agent is defined in Claim 1, it is inevitably suitable for any of the disclosed, desirable modifications of the properties of the surface of the membrane.

3.2 As regards the further claims of the Auxiliary request:

(a) Claims 2 to 4 are, respectively, identical to Claims 2, 5 and 6 as filed.

(b) Claim 5 has been amended because the index "n" (indication of the number of repeating units) is undefined (Article 84 EPC), so that the formula of the repeating unit of the intended copolymer was merely a graphic definition of the name of that copolymer. Hence, that formula has simply been replaced by the name of the intended copolymer. The mention of the name of the copolymer instead

of its generic formula does not add any subject-matter.

(c) Claims 6 to 16, respectively, apart from minor modifications (deletion of "active", as in Claim 1) are identical to Claims 19-27, 31 and 32 as filed.

3.3 Therefore, the amended claims of the Auxiliary Request all have a direct and unambiguous basis in the application as filed (Article 123(2) EPC).

3.4 The terms and the claims that had been objected to by the Board in the communication in preparation for the oral proceedings have been removed. The Board has no reason to raise any objections under Article 84 EPC.

3.5 The amendments carried out are appropriate and necessary for overcoming the ground for refusal, as will be apparent from the following (Rule 80 EPC).

3.6 Therefore, the Auxiliary Request is formally allowable.

4. *Novelty*

4.1 The sole ground for refusal in the decision under appeal was the lack of novelty of the subject-matter then claimed (Point III, *supra*) having regard to the disclosure of D8.

4.2 D8 concerns a process for preparing a surface modified, skinless, hydrophilic, microporous, alcohol-insoluble polyamide membrane with controlled pore surface properties, capable of reacting or interacting in a controlled manner with (a) particulate matter in a fluid, (b) non-particulate matter in a fluid, or (c)

both (a) and (b) and that is readily wetted by water, which comprises:

(1) preparing a casting solution comprised of (A) a casting resin system comprised of (a) an alcohol-insoluble polyamide resin having a ratio CH_2 : NHCO of methylene CH_2 to amide NHCO groups within the range of from about 5:1 to about 7:1, and (b) a water-soluble, membrane surface modifying polymer having functional polar groups selected from the group consisting of hydroxyl, carboxyl, and amino or a non-reacting combination thereof, and a molecular weight of 20,000 or greater, and (B) a solvent system in which said casting resin system is soluble;

(2) inducing nucleation of said casting solution by controlled addition of nonsolvent for said casting resin system under controlled conditions of concentration, temperature, addition rate and degree of agitation to obtain a visible precipitate of casting resin system particles, thereby forming a casting composition;

(3) spreading said casting composition on a substrate to form a thin film thereof on the substrate;

(4) contacting and diluting the film of said casting composition with a liquid nonsolvent system for said casting resin system comprised of a mixture of solvent and nonsolvent liquids, thereby precipitating said casting resin system from said casting composition in the form of a thin, skinless, hydrophilic, surface modified, microporous, polyamide membrane with controlled surface properties;

(5) washing said membrane to remove solvent; and

(6) drying said membrane (Claim 1).

In particular, the polyamide resin can be poly hexamethylene adipamide (Claim 4), poly-ε-caprolactam (Claim 5) or polyhexamethylene sebacamide (Claim 6) and the membrane surface modifying polymer can *inter alia* be a fully hydrolyzed copolymer of maleic anhydride with methyl vinyl ether (Claim 16).

D8 also concerns a surface modified, skinless, hydrophilic, microporous, alcohol-insoluble polyamide membrane derived from an alcohol-insoluble hydrophobic polyamide resin having a ratio CH_2 : NHCO of methylene CH_2 to amide NHCO groups within the range of from about 5:1 to about 7:1, said membrane having an integral microstructure and characterized by
(1) the surface properties thereof being substantially controlled by functional polar groups of a membrane surface modifying polymer having a molecular weight of 20,000 or greater, said functional polar groups selected from the group consisting of hydroxyl, carboxyl, and amino, or a non-reacting combination thereof,
(2) said membrane surface modifying polymer being homogeneously distributed in said membrane, and
(3) having the capability of reacting or interacting in a controlled manner with (a) particulate matter in a fluid, (b) non-particulate matter in a fluid, or (c) both (a) and (b) (Claim 26).

4.2.1 Claim 1 according to the Auxiliary Request, compared to Claim 1 underlying the decision under appeal, contains several new features, in particular the definition of the membrane forming polymer being PVDF or polysulfone.

4.2.2 Since the membrane forming polymer disclosed by D8 is a particular polyamide, the membrane defined in Claim 1 of the Auxiliary Request is structurally different from that disclosed by D8, and thus it is novel (Article 54 EPC).

4.3 A further document (D9, US-A-3 556 305) has been cited by the Examining Division in their last communication (dated 13 July 2006) but has not been dealt with in the decision under appeal.

4.3.1 D9 concerns a membrane suitable for use in ultrafiltration, reverse osmosis, and other such separation processes, said membrane comprising:

1. a porous substrate formed of a highly anisotropic polymeric membrane having
   a. a barrier layer at one surface thereof comprising micropores from 15 to 1,000 angstroms in diameter and having a thickness from about 0.1 to 5 microns and
   b. a macroporous support layer;
2. an adhesive polymeric layer having a maximum thickness of about 1,500 angstroms coated on said substrate; and
3. a diffusive polymer or gel-like film bonded to said barrier layer of said porous substrate by said adhesive layer (Claim 1).

4.3.2 The adhesive layer can consist of an alkyl vinyl ether polymer or copolymer (Claim 3). The diffusive film can be polyvinyl alcohol or a mixture of polyvinyl methyl ether with a copolymer of methyl vinyl ether with maleic anhydride (Claim 4). The porous substrate can be formed of a polysulfone polymer (Claim 6).

4.3.3 The examples of D9 illustrate, respectively:

(a) A preparation method in which an anisotropic membrane of Polymer 360(TM) (a polysulfone, see D9, column 5, lines 61 to 70) is soaked in an aqueous solution containing poly(vinyl methyl ether) (Gantrez(TM) M-155) to form an adhesive coated anisotropic membrane, through which, subsequently, an aqueous solution comprising poly(vinyl alcohol) is filtered to form a thin diffusive membrane of poly(vinyl alcohol) over the adhesive layer (Example 1).

(b) A preparation method essentially as in Example 1, in which however an aqueous solution comprising poly(methyl vinyl ether/maleic anhydride) (Gantrez(TM) AN-139)) is filtered through the adhesive coated anisotropic membrane to form a thin diffusive membrane of poly(methyl vinyl ether/maleic anhydride) over the adhesive layer (Example 2);

(c) The preparation procedure of Example 2 followed by a filtration test with a different solution (Example 3).

4.3.4 Hence, D9 concerns a tripartite membrane comprising an anisotropic porous substrate, an ultrathin adhesive layer formed over the porous substrate and a thin diffusive membrane formed over the adhesive layer and bound to the substrate by the adhesive layer (Column 1, lines 60 to 65).

4.3.5 Claim 1 according to the Auxiliary Request, compared to Claim 1 underlying the decision under appeal, contains several new features, in particular the definition of a

step of the preparation process of the membrane, according to which the reactivity modifying agent, i.e. the poly(alkyl vinyl ether/maleic anhydride), is added to the membrane forming polymer that is PVDF or polysulfone before the membrane is cast. This process step implies that the claimed membrane is cast from a single polymer solution or dope, called "blend" in the present application, which casting forms an integral membrane that is different from composite membranes consecutively cast from more than one polymer solution to form a layered membrane, as in D9.

4.3.6 Since the membrane disclosed by D9 is a tripartite membrane, i.e. a layered or composite membrane, the "integral" membrane defined in Claim 1 of the Auxiliary Request is structurally different from that disclosed by D9. Also, the layered membrane disclosed by D9 does not necessarily comprise a copolymer of poly(alkyl vinyl ether/maleic anhydride). Thus, the claimed membrane is novel also having regard to D9 (Article 54 EPC).

4.4 As regards the other documents cited by the Examining Division in their first communication (31 August 2005), the Board has no reason to take a different position, for the following reasons.

(a) The membrane of D1 (EP-A-0 463 627) is made of PTFE and a hydrophilic agent that is a highly functionalized PTFE (Claim 1).

(b) That of D2 (WO-A-99/59707) comprises PVDF and a hydrophilic polymer that can comprise polyvinyl pyrrolidone (PVP) (Claims 19 and 24), as well as, as a wetting agent, hydroxypropylcellulose (HPC) or a surfactant (page 7, lines 17-19).

(c) The membrane disclosed by D3 (EP-A-0 911 073) is made of polysulfone or polyethersulfone and contains a hydrophilic acrylate polymer (Claim 1).

(d) That of D4 (US-A-5 531 900) is made of PVDF and contains a positively charged organic phosphonium compound, i.e. vinyltriphenyl phosphonium bromide, and, grafted thereto, an acrylate or methacrylate (Claim 1).

(e) The membrane of D5 (EP-B-0 430 082) comprises a hydrophobic fluorinated hydrocarbon polymer such as PVDF and a hydrophilic cross-linked polymer such as hydroxy alkyl acrylate (Claims 1, 4 and 5).

(f) The membrane disclosed by D6 (WO-A-99/01207) comprises a film that is made of a reaction product of (a) a complex of PVDF with calcined α-alumina particles, and (b) a hydrophilic polymer (Claim 1) such as polyvinyl alcohol (Examples).

None of these documents thus disclose the polymeric membrane forming material and the particular reactivity modifying agent now claimed.

4.5 Therefore, the claimed subject-matter is novel having regard to each of those documents.

5. *Remittal*

5.1 The sole ground for revocation in the decision under appeal was the lack of novelty having regard to D8.

5.2 Claims 1 to 16 of the Auxiliary Request filed at the oral proceedings before the Board overcome that ground as well as the objections raised by the Board under Articles 84 and 123(2) EPC.

5.3 As inventive step was not considered in the decision under appeal, the Board, in the exercise of its discretion under Article 111(1) EPC, considers it appropriate to remit the case to the Examining Division for further prosecution.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance for further prosecution on the basis of Claims 1 to 16 of the Auxiliary Request submitted during the oral proceedings on 3 April 2009.

The Registrar: The Chairman:

C. Moser S. Perryman