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D E C I S I O N
of 2 July 1997

Case Number: T 0129/93 - 3.3.3

Application Number: 87118964.3

Publication Number: 0272676

IPC: C08G 63/18

Language of the proceedings: EN

Title of invention:
Wholly aromatic polyester and process for its production

Applicant:
Mitsubishi Chemical Corporation

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54, 84, 88(4)

Keyword:
"Novelty (yes) - disclaimer - valid priority"
"Claims - clear and supported by the description - (yes)"

Decisions cited:
G 0010/93

Catchword:
-

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0129/93 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 2 July 1997

Appellant: Mitsubishi Chemical Corporation
5-2, Marunouchi 2-chome
Chiyoda-ku
Tokyo (JP)

Representative: Wächtershäuser, Günter, Proof. Dr.
Patentanwalt
Tal 29
80331 München (DE)

Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 15 September 1992 refusing European patent application No. 87 118 964.3 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: C. Gérardin
Members: B. ter Laan
J. A. Stephens-Ofner

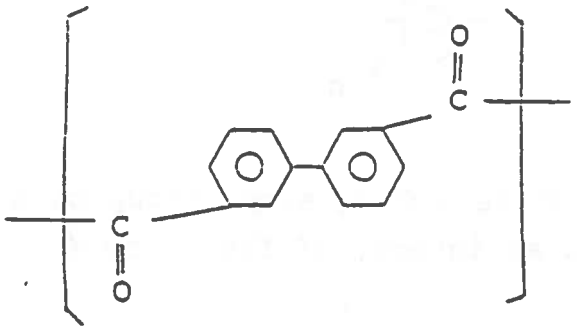
Summary of Facts and Submissions

I. European patent application No. 87 118 964.3, filed on 21 December 1987, claiming priority from an earlier application in Japan (JP 305299/86 of 23 December 1986), and published on 29 June 1988 under publication No. 0 272 676, was refused by a decision of the Examining Division of European Patent Office dated 15 September 1992.

The decision was based on a set of 16 claims, of which Claim 1 was filed on 23 July 1992 and Claims 2 to 16 were as originally filed. The claims read as follows:

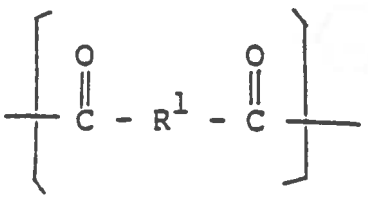
"1. A wholly aromatic polyester comprising:

(a) from 2 to 50 equivalent % of a 3,3'-biphenyldicarboxylic acid residue of the formula:



(1)

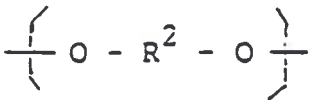
(b) from 0 to 50 equivalent % of a dicarboxylic acid residue of the formula:



(2)

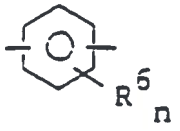
wherein R^1 is a bivalent aromatic hydrocarbon group or a group of the formula R^4-X-R^5 wherein each of R^4 and R^5 is a bivalent aromatic hydrocarbon group and X is an oxygen atom, a sulfur atom, a sulphonyl group, a carbonyl group, an alkylene group, an ester group or a direct bond, provided that R^1 is not a 3,3'-biphenyl group,

(c) from 10 to 52 equivalent % of a dioxy compound residue of the formula:

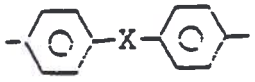


(3)

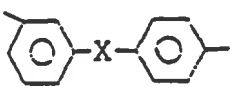
wherein R^2 is a group of the formula



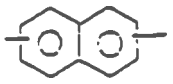
wherein R^6 is a C_1-C_4 alkyl group or a phenyl group and n is an integer of from 0 to 4,



wherein X is as defined above,

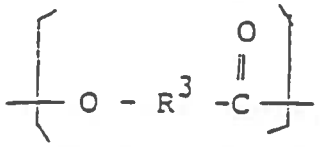


wherein X is as defined above, and/or



and

(d) from 0 to 80 equivalent % of an oxycarboxylic acid residue of the formula:



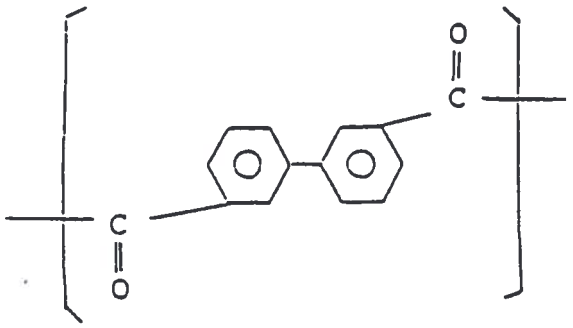
(4)

wherein R^3 is a bivalent aromatic hydrocarbon group or R^4-X-R^5 wherein R^4, R^5 and X are as defined above, whereby (a) + (b) + (c) + (d) = 100 eq%, and said polyester has a melt viscosity of at least 50 poise as measured at 275°C at 100 sec^{-1}.

Claims 7, 10 and 14 all referred to different preferred embodiments of Claim 1. They read:

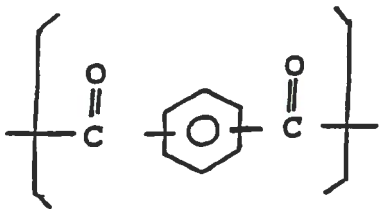
"7. The wholly aromatic polyester according to Claim 1, which comprises:

(a) from 2 to 40 equivalent % of a 3,3'-biphenyldicarboxylic acid residue of the formula:



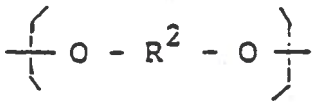
(1)

(e) from 5 to 45 equivalent % of an aromatic dicarboxylic acid residue of the formula:



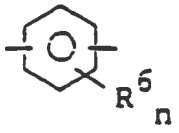
(5)

(c) from 12 to 45 equivalent % of an aromatic dioxy compound residue of the formula:

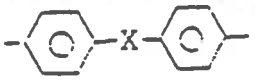


(3)

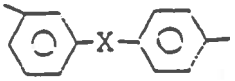
wherein R^2 is a group of the formula



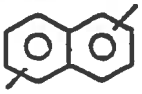
wherein R^6 is a C_1-C_4 alkyl group or a phenyl group and n is an integer of from 0 to 4,



wherein X is an oxygen atom, a sulfur atom, a sulphonyl group, a carbonyl group, an alkylene group, an ester group or a direct bond,



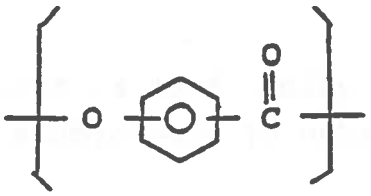
wherein X is as defined above, and/or



,

and

(f) from 10 to 75 equivalent % of an oxycarboxylic acid residue of the formula:

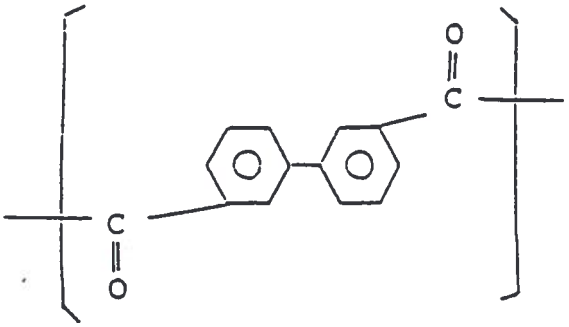


(6)

the total of the residues (a), (e) (c) and (f) being from 90 to 100 equivalent %.

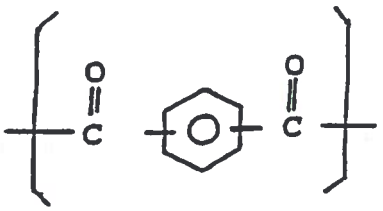
10. The wholly aromatic polyester according to Claim 1, which comprises:

(a) from 2 to 45 equivalent % of a 3,3'-biphenyldicarboxylic acid residue of the formula:



(1)

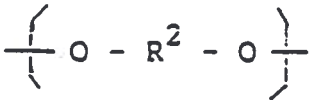
(e) from 5 to 48 equivalent % of an aromatic dicarboxylic acid residue of the formula:



(5)

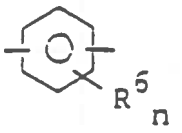
and

(c) about 50 equivalent % of an aromatic dioxy compound residue of the formula:

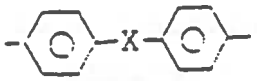


(3)

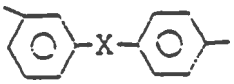
wherein R^2 is a group of the formula



wherein R^6 is a C_1-C_4 alkyl group or a phenyl group and n is an integer of from 0 to 4,



wherein X is an oxygen atom, a sulfur atom, a sulphonyl group, a carbonyl group, an alkylene group, an ester group or a direct bond,



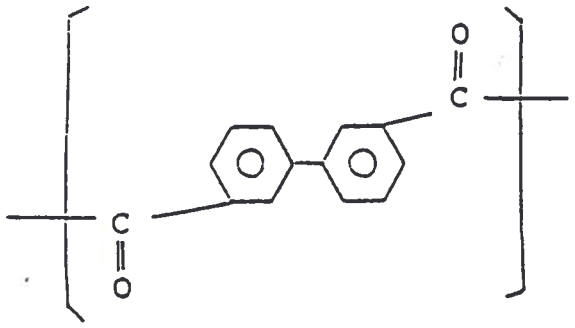
wherein X is as defined above, and/or

,

the total of the residues (a) and (e) being about the same equivalent % as the residue (c), and the total of the residues (a), (e) and (c) being from 90 to 100 equivalent %.

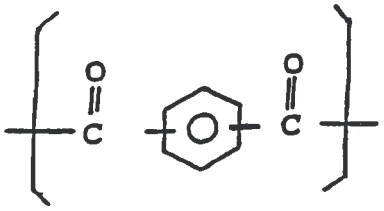
14. The aromatic polyester according to Claim 1, which comprises:

(a) from 5 to 45 equivalent % of a 3,3'-biphenyldicarboxylic acid residue of the formula:



(1)

(e) from 5 to 45 equivalent % of an aromatic dicarboxylic acid residue of the formula:



(5)

and

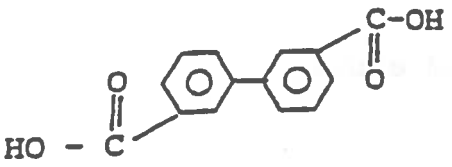
(c) about 50 equivalent % of an aromatic dioxy compound residue selected from the group consisting of a 2,2-bis(4-hydroxyphenyl)propane residue, a bis(4-hydroxyphenyl) sulphone residue, a bis(4-hydroxyphenyl)ether residue and a resorcinol residue, the total of the residues (a) and (e) being about 50 equivalent %, and the total of the residues (a), (e) and (c) being 100 equivalent %."

Claims 2 to 6, 8 and 9, 11 to 13 and 15 were dependent and referred to preferred embodiments of Claims 1, 7, 10 and 14, respectively.

Independent process Claim 16 read:

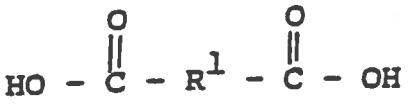
"A process for producing a wholly aromatic polyester, which comprises reacting

(i) 3,3'-biphenyldicarboxylic acid of the formula:



(7)

(j) an aromatic dicarboxylic acid of the formula:



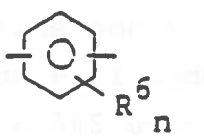
(8)

wherein R^1 is a bivalent aromatic hydrocarbon group or a group of the formula R^4-X-R^5 wherein each of R^4 and R^5 is a bivalent aromatic hydrocarbon group and X is an oxygen atom, a sulfur atom, a sulphonyl group, a carbonyl group, an alkylene group, an ester group or a direct bond, provided that R^1 is not a 3,3'-biphenyl group,

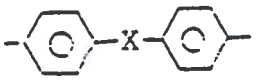
(k) an aromatic dihydroxy compound of the formula:

$$HO - R^2 - OH \qquad (9)$$

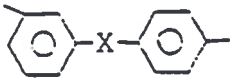
wherein R^2 is a group of the formula



wherein R^6 is a C_1-C_4 alkyl group or a phenyl group and n is an integer of from 0 to 4,



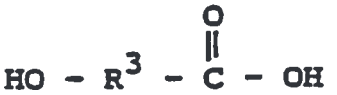
wherein X is as defined above,



wherein X is as defined above, and/or

and

(1) an aromatic hydroxy carboxylic acid of the formula:



(10)

wherein R^3 is a bivalent aromatic hydrocarbon group or R^4-X-R^5 wherein R^4, R^5 and X are as defined above, or their derivatives in the presence of an acid anhydride, followed by reaction at a temperature of from 100 to 400°C, while distilling off the acid and acid anhydride."

II. The Examining Division held that the claimed subject-matter was not novel. In particular, it was found that the present application was not entitled to its priority date of 23 December 1986 and that, although the filing date of D1 (EP-A-0 296 451, 14 June 1988) was later than that of the present application (21 December 1987), its priority date (26 June 1987)

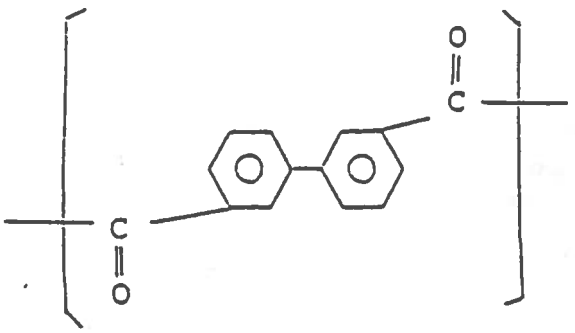
was earlier, so that it was a state of the art document according to Articles 54(3) and 54(4) EPC. As D1 disclosed compounds covered by the claims under consideration, it destroyed the novelty of the claimed subject-matter.

III. On 12 November 1992 a Notice of Appeal was lodged against that decision, together with payment of the prescribed fee. With the Statement of Grounds of Appeal filed on 15 January 1993, a new Claim 1, to be combined with original Claims 2 to 16, was filed by way of auxiliary request.

IV. After a communication from the Board, in which several objections were raised against the two sets of claims then on file, on 25 November 1996 a new set was filed, which claims were replaced by a set of 14 claims filed on 18 June 1997, to be replaced again by a set of 12 claims filed during the oral proceedings before the Board on 2 July 1997, which reads:

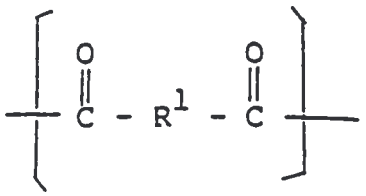
"1. A wholly aromatic polyester comprising:

(A) from 2 to 50 equivalent % based on the total of the aromatic components being 100 equivalent % of a 3,3'-biphenyldicarboxylic acid residue of the formula:



(1)

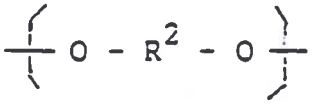
(B) from 0 to 50 equivalent % based on the total of the aromatic components being 100 equivalent % of a dicarboxylic acid residue of the formula:



(2)

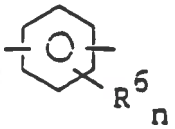
wherein R^1 is a bivalent aromatic hydrocarbon group or a group of the formula R^4-X-R^5 wherein each of R^4 and R^5 is a bivalent aromatic hydrocarbon group and X is an oxygen atom, a sulfur atom, a sulphonyl group, a carbonyl group, an alkylene group, an ester group or a direct bond, provided that R^1 is not a 3,3'-biphenyl group,

(C) from 10 to 52 equivalent % based on the total of the aromatic components being 100 equivalent % of a dioxy compound residue of the formula:

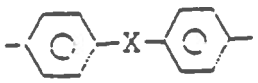


(3)

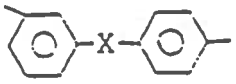
wherein R^2 is a group of the formula



wherein R^6 is a C_1-C_4 alkyl group or a phenyl group and n is an integer of from 0 to 4,



wherein X is as defined above,

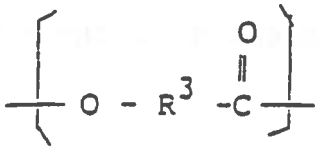


wherein X is as defined above, and/or

with the exception of a residue derived from phenylhydroquinone and 4,4'-dihydroxy-diphenyl ether;

and

(D) from 0 to 80 equivalent % based on the total of the aromatic components being 100 equivalent % of an oxycarboxylic acid residue of the formula:



(4)

wherein R^3 is a bivalent aromatic hydrocarbon group or R^4-X-R^5 wherein R^4, R^5 and X are as defined above, and said polyester has a glass transition temperature of at least 100 °C and a melt viscosity of at least 5 Pa/s (50 poise) as measured at 275°C

at 100 sec^{-1}.

2. The polyester according to Claim 1, wherein R^1 in the formula (2) is

3. The polyester according to Claim 1, wherein R^3 in the formula (4) is

4. The polyester according to Claim 3, wherein the residue (d) of the formula (4) is from 0 to 75 equivalent %.

5. The polyester as defined in Claims 2 and 3 jointly, which comprises:

(A) from 2 to 40 equivalent % of the residue of formula (1);

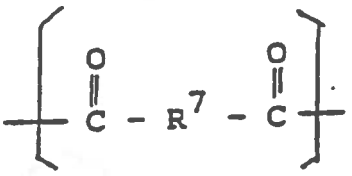
(B) from 5 to 45 equivalent % of the residue of formula (2);

(C) from 12 to 45 equivalent % of the residue of formula (3); and

(D) from 10 to 75 equivalent % of the residue of formula (4)

the total of the residues (A), (B), (C) and (D) being from 90 to 100 equivalent %.

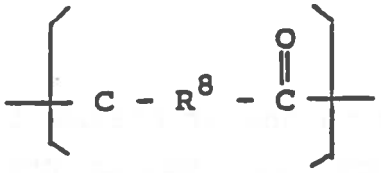
6. The polyester according to Claim 5 which further contains (E) up to 10 equivalent % of a residue of the formula:



(11)

wherein R^7 is R^1 with the exception of 

and/or of a residue of the formula



(12)

wherein R^8 is R^3 with the exception of  .

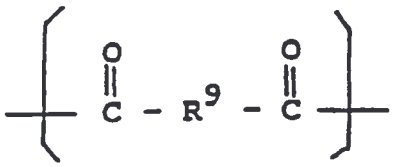
7. The polyester according to Claim 2, which comprises:

(A) from 2 to 45 equivalent % of the residue of formula (1);

(B) from 5 to 48 equivalent% of the residue of formula (2);

(C) about 50 equivalent % of the residue of formula (3) and

(F) from 0 to 10 equivalent % of a residue of the formula:



(13)

wherein R^9 is R^1 with the exception of 

the total of the residues (A) and (B) being about the same equivalent % as the residue (C), and the total of the residues (A), (B) and (C) being from 90 to 100 equivalent %, and the total of the residues A, B, C, F being 100 equivalent %.

8. The polyester according to one of Claims 1 or 5 which is optically an isotropic in a molten phase.

9. The polyester according to Claim 7 which has a glass transition temperature (Tg) of at least 120°C.

10. The polyester according to Claim 2, which comprises:

(A) from 5 to 45 equivalent % of the residue of formula (1);

(B) from 5 to 45 equivalent % of the residue of formula (2) and

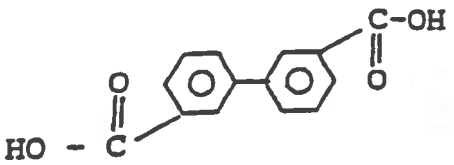
(C) about 50 equivalent % of an aromatic dioxy compound residue selected from the group consisting of a 2,2-bis(4-hydroxyphenyl)propane residue, a 2,2-bis(4-hydroxyphenyl) sulphone residue, and a resorcinol residue,

the total of the residues (A) and (B) being about 50 equivalent %, and the total of the residues (A), (B) and (C) being 100 equivalent %.

11. The wholly aromatic polyester according to Claim 14, which has a glass transition temperature (Tg) of at least 150°C.

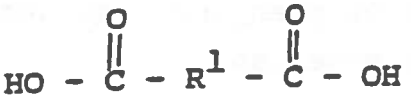
12. A process for producing a wholly aromatic polyester, which comprises reacting

(i) 3,3'-biphenyldicarboxylic acid of the formula:



(7)

(j) an aromatic dicarboxylic acid of the formula:



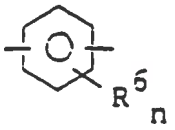
(8)

wherein R^1 is a bivalent aromatic hydrocarbon group or a group of the formula R^4-X-R^5 wherein each of R^4 and R^5 is a bivalent aromatic hydrocarbon group and X is an oxygen atom, a sulfur atom, a sulphonyl group, a carbonyl group, an alkylene group, an ester group or a direct bond, provided that R^1 is not a 3,3'-biphenyl group,

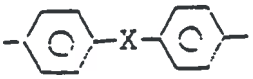
(k) an aromatic dihydroxy compound of the formula:

$$HO - R^2 - OH$$

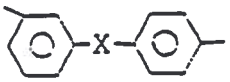
wherein R^2 is a group of the formula



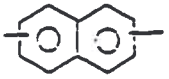
wherein R^6 is a C_1-C_4 alkyl group or a phenyl group and n is an integer of from 0 to 4,



wherein X is as defined above,

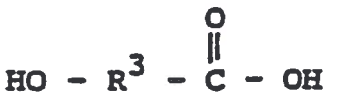


wherein X is as defined above, and/or



with the exception of phenylhydroquinone and 4,4'-dihydroxy-diphenyl ether and

(l) an aromatic hydroxy carboxylic acid of the formula:



(10)

wherein R^3 is a bivalent aromatic hydrocarbon group or R^4-X-R^5 wherein R^4, R^5 and X are as defined above, or their derivatives in the presence of an acid anhydride, followed by reaction at a temperature of from 100 to 400°C, while distilling off the acid and acid anhydride."

V. The Appellant argued that the compounds disclosed in D1 were now excluded from protection by means of a disclaimer. Therefore, the claimed-subject-matter was now novel. Although arguments for the presence of an inventive step were provided as well as additional experiments for support of those arguments were presented in the written proceedings, that issue was not further dealt with during the oral proceedings before the Board.

The Appellant requested that the decision of the first instance be set aside and that the application be remitted to the Examining Division for further prosecution on the basis of claims 1 to 12 as submitted during oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

Article 123(2) EPC

2. The wording of the claims does not give rise to any objections under Article 123(2) EPC for the following reasons.

2.1 Product Claim 1 is based upon Claims 1 and 6 as originally filed. The disclaimer derives from D1, page 3, line 28 (phenylhydroquinone) and example 8 (4,4'-dihydroxy-diphenyl ether). The relationship to the total of the aromatic components can be found on original description page 6, lines 20 to 21, page 7, lines 20 to 21 and page 9, lines 2 to 3 and 24 to 25 (page 6, lines 40 to 41 and 55 to 56 and page 7, lines 39 to 40 and 51 to 52 of the application as published).

Claims 2 to 4 correspond to original Claims 3 to 5.

As the result of its appendancy to Claims 2 and 3, Claim 5 corresponds in substance to original Claim 7. Claim 6 is based upon original Claim 9 and has been clarified in the sense that the presence of 0 equivalent % of component (E) is now excluded, which is consistent with the term "which further contains" originally present and with the meaning of Claim 5 (original 7).

Claim 7 is based upon original Claim 10, in which the optional compositional feature corresponding to the subject-matter of original Claim 13 has been incorporated; the total amount of components present derives from the combination of those two claims and is supported by the description, original page 20, lines 1 to 3 and page 23, lines 10 to 11 (page 12, lines 53 to 54 and page 14, lines 38 to 39 as published).

Claims 8 and 9 are based upon original Claims 2 and 8 and upon original Claim 12, respectively.

The basis for Claim 10 can be found in original Claim 14; it has been adapted to Claim 1.

Claim 11 is the same as original Claim 15.

2.2 Process Claim 12 refers to the process as disclosed in original Claim 16 and is amended in conformity with present Claim 1.

Clarity and support

3. The Board is satisfied that the claims comply with the requirements of Article 84 EPC as they are clear and are supported by the description. However, the description still needs some adaptation to conform to the amended claims.

3.1 In the claims the expression "equivalent %" is used to indicate the relative amounts of the various components of the polyester. In the Board's view, also in the light of the examples in which the amounts of aromatic components are expressed in moles and always add up to 100%, this term can only be understood as "mole %".

3.2 In Claim 7 the amounts of the residues (A) and (B) of the polyester are required to be about the same as the amount of residue (C). Since the latter is "about 50 equivalent %", this implies that the former together should be "about 50 equivalent %" as well, thus adding up to a total of 100 equivalent %. On the other hand, the total of residues (A), (B) and (C) can be from 90 to 100 equivalent %. From the optional presence of residue (F) in an amount of up to 10 equivalent % and the additional requirement that (A) + (B) + (C) + (F) must be 100 equivalent %, it can only be concluded that the residues (A) + (B) + (C) should always add up to 100 equivalent % unless residue (F) is present, in which case the latter requirement applies.

3.3 Following a communication from the Board the Appellant submitted arguments and comparative examples from which it appeared that the polyesters according to the application in suit were optically anisotropic (statement filed on 25 November 1996, point 1 and test Report 1). During oral proceedings, in order to make a distinction between essential and non-essential features, which would be of crucial importance for the

wording of the claims, the Board addressed the question of the general properties of the polyesters being claimed. In reply, the Appellant stated that in fact the polyesters according to Claim 1 were not limited to liquid crystalline polymers. This statement was in line with the description of the polyesters in the application, which only identifies the ranges of melt viscosity and glass transition temperature as essential features (page 8, lines 12 to 15 and page 16, lines 55/56), both of which are now present in Claim 1. Therefore, insofar as the claimed subject-matter now contains the features indicated in the description as being essential, it is supported by the description. However, the description still contains a number of examples referring to compounds not being claimed (e.g. Examples 14 and 17) and therefore is still in need of adaptation to the new claims.

3.4 Although the wording of the individual claims is adequately supported by the description, in view of the Appellant's statement about optical anisotropy, it also seems appropriate to consider the structure of the set of claims. Like the claims, the description also distinguishes three preferred embodiments characterized by different compositions in specific amounts:

- polyesters with optical anisotropy in the molten phase (page 8, line 19 to page 12, line 48) which is the subject-matter of present Claim 5,

- polyesters with a high modulus of elasticity (page 12, line 49 to page 15, line 7) which is the subject-matter of present Claim 7,

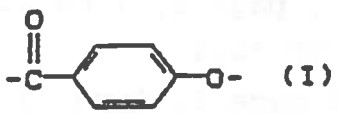
- polyesters with a well-balanced heat resistance and mouldability (page 15, lines 8 to 43) which is the subject-matter of present Claim 10.

If polyesters having optical anisotropy are nothing more than one out of three optional embodiments, any evidence in relation to that embodiment basically concerns only an aspect of Claim 1, not the single general technical concept underlying the application in suit. Therefore, the fact that the liquid crystalline properties do not belong to that single general technical concept may result in the question of unity of invention (Article 82 EPC) being raised.

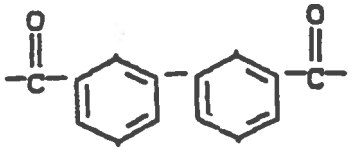
Novelty

4. The refusal for lack of novelty was based upon the conclusion that the application was not entitled to its priority date and that D1 therefore belonged to the state of the art within the meaning of Articles 54(3) and 54(4) EPC. However, a number of compounds falling under present Claim 1 was specifically disclosed in the priority document (see the paragraph bridging pages 5 and 6 of the English translation), so that there can be no doubt that the priority of at least those specific compounds is valid (Article 88(4) EPC). Therefore, in the present case, it first needs to be decided just how far D1 actually discloses the compounds now being claimed, and then the priority of those compounds needs to be verified.

4.1 D1 discloses wholly aromatic polyesters containing repeating residues of the formulae



(I)



(II)

and -O-Ar-O (III)

in which Ar consists of from 0 to 100 Mol-% 1,4-phenylene residues and of from 100 to 0 Mol-% of other bivalent aromatic residues containing 6 to 18 C-atoms, of which the chain-forming bonds are positioned coaxially or parallel in an amount of from 50 to 100 Mol-% and at an angle in an amount of from 50 to 0 Mol-% and which can be substituted by C_1-C_4-alkoxy groups or halogen atoms, provided that the mole ratio I/II is from 0,1 to 4,0, and the mole ratio II/III is from 0,95 to 1,05 (Claim 1). That very general disclosure of the dioxy compound (formula III), does not anticipate the claimed subject-matter, which requires the presence of much more specific dioxy compounds. However, specific compounds are mentioned in D1 on page 3, lines 23 to 30 and in the examples, of which only 4,4'-dihydroxydiphenyl, 1,4-, 1,5-, 2,6- and 2,7-dihydroxynaphthalene, methyl hydroquinone, resorcinol, Bisphenol-A, 4,4'-dihydroxydiphenylketone and 4,4'-dihydroxydiphenyl sulfide are covered by present Claim 1. Therefore, the question is, whether for those compounds a valid priority right exists.

4.2 That question can be answered positively in view of the disclosure on page 5, line 17 to page 6, line 16 of the English translation of the priority document, in particular page 6, line 10 (4,4'-dihydroxydiphenyl), page 5, lines 23 to 25 (1,4-, 1,5-, 2,6- and 2,7-dihydroxynaphthol), page 5, line 17 (methyl hydroquinone and resorcinol), page 5, line 25 (Bisphenol-A), page 6, line 10 (4,4'-dihydroxydiphenylketone) and page 6, line 13 (4,4'-dihydroxydiphenyl sulfide).

4.3 Therefore, the polyesters obtained from diphenols of formula (3) that are disclosed by D1 are entitled to the priority of 23 December 1986 (Article 88(4) EPC), so that for those compounds D1 is not state of the art within the meaning of Articles 54(3) and 54(4) EPC. The other compounds included by present Claim 1 are not disclosed by D1, so that D1 does not destroy the novelty of the claimed subject-matter.

4.4 As regards the polyesters derived from either phenylhydroquinone or 4,4'-dihydroxydiphenyl ether, which are explicitly envisaged in D1, the disclaimer of the corresponding residues from the definition of formula (3) overcomes any possibility of overlapping and ensures that the requirement of novelty is met.

5. Regarding D2 (EP-A-0 072 450) and D3 (EP-A-0 024 499), the first instance did not raise any novelty objections and, in the light of the disclosure of those two documents, the Board sees no reason to deviate from that opinion.

6. For those reasons, the claimed subject-matter is novel.

Inventive step

7. During the examination procedure (communication of 13 January 1992, point 3) the Examining Division invited the Appellant (then Applicant) to submit arguments supporting the presence of an inventive step vis-à-vis D2 and D3. Although the Appellant pointed at the examples and comparative examples of the application to show that the claimed subject-matter was not obvious, in the reasons for the Examining Division's decision no reference was made to the issue of inventive step. In these circumstances, the Board cannot deal with the merits of this issue by way of appeal for by so doing it would deprive the Appellant

from its right to two instances, one administrative, the other judicial (G 10/93, OJ EPO 1995, 172). Accordingly, the Board remits the case to the first instance for further prosecution pursuant to Article 111(1) EPC.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Examining Division for further processing of the claims submitted during oral proceedings.

The Registrar: The Chairman:

E. Görgmaier C. Gérardin