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D E C I S I O N
of 15 September 1999

Case Number: T 0927/97 - 3.3.3

Application Number: 93912102.6

Publication Number: 0633911

IPC: C08L 67/02

Language of the proceedings: EN

Title of invention:
Polyester/polyamide blend having improved flavour retaining property and clarity

Applicant:
Eastman Chemical Company

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56

Keyword:
"Novelty - implicit disclosure (no)"
"Inventive step - (yes) after amendment"

Decisions cited:
-

Catchword:
-

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Case Number: T 0927/97 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 15 September 1999

Appellant: Eastman Chemical Company
100 North Eastman Road
Kingsport
TN 37660
ETATS-UNIS D'AMERIQUE

Representative: Wibbelmann, Jobst, Dr., Dipl.-Chem.
Wuesthoff & Wuesthoff
Patent- und Rechtsanwälte
Schweigerstrasse 2
81541 München
ALLEMAGNE

Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 27 March 1997 refusing European patent application No. 93 912 102.6 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: C. Gérardin
Members: B. ter Laan
A. Lindqvist

Summary of Facts and Submissions

I. European patent application No. 93 912 102.6, based on the international application No. PCT/US93/03056, filed on 31 March 1993, claiming priority of 2 April 1992 from an earlier application in the USA (US 862027), and published on 14 October 1993 under No. WO 93/20147, was refused by a decision of the Examining Division of the European Patent Office dated 27 March 1997. That decision was based on a set of five claims which were held not to be novel in view of D1 (US-A-4 837 115).

Claim 1 read:

"A polyester composition having an improved flavour retaining property comprising:

(A) 98.0 to 99.5 weight percent of a polyester which comprises

(1) a dicarboxylic acid component comprising repeat units from at least 85 mole percent terephthalic acid; and

(2) a diol component comprising repeat units from at least 85 mole percent ethylene glycol; based on 100 mole percent dicarboxylic acid and 100 mole percent diol; and

(B) 2.0 to 0.05 weight percent of a polyamide selected from the group consisting of low molecular weight partially aromatic polyamides having a number average molecular weight of less than 12,000 and an I.V. of less than 0.8 dl/g, low molecular weight aliphatic polyamides having a number average molecular weight of less than 7,000 and an

I.V. of less than 0.8 dl/g, and combinations thereof, provided that the polyamide is prepared from a diamine and either a diacid or the ester form of a diacid, wherein the combined weights of (A) and (B) total 100 percent."

Dependent Claims 2 to 4 referred to preferred embodiments of the composition according to Claim 1.

Independent Claim 5 read:

"A process for reducing the acetaldehyde concentration in a heat-molded polyester article which comprises dry-blending a thermoplastic polyester with 0.05 to 2.0 weight percent, based on the polyester, of a polyamide selected from the group consisting of low molecular weight partially aromatic polyamides having a number average molecular weight of less than 12,000 and an I.V. of less than 0.8 dl/g, low molecular weight aliphatic polyamides having a number average molecular weight of less than 7,000 and an I.V. of less than 0.8 dl/g, and combinations thereof, provided that the polyamide is prepared from a diamine and either a diacid or the ester form of a diacid, melt-kneading the composition and thermoforming the melt-kneaded composition into an article."

II. On 15 May 1997 a Notice of Appeal was lodged against that decision, together with payment of the prescribed fee. With the Statement of the Grounds of Appeal, filed on 28 July 1997, the Appellant submitted a new set of five claims, in which the polyamide was defined more broadly than in Claim 1 upon which the Examining Division had based its decision. By a further letter

dated 20 November 1997, a declaration by one of the inventors was filed in support of the arguments given in the Statement of the Grounds of Appeal.

III. During oral proceedings held on 15 September 1999, after the Board had pointed out several objections under Article 84 EPC, the Appellant submitted a new set of five claims as the sole request.

Claim 1 reads:

"A polyester composition comprising:

(A) 98.0 to 99.5 weight percent of a polyester which comprises

(1) a dicarboxylic acid component comprising repeat units from at least 85 mole percent terephthalic acid; and

(2) a diol component comprising repeat units from at least 85 mole percent ethylene glycol, based on 100 mole percent dicarboxylic acid and 100 mole percent diol; and

(B) 2.0 to 0.05 weight percent of a polyamide having terminal amino groups said polyamide being selected from the group consisting of low molecular weight partially aromatic polyamides having a number average molecular weight of less than 12,000 and an I.V. of less than 0.7 dl/g, low molecular weight aliphatic polyamides having a number average molecular weight of less than 6,000 and an I.V. of less than 0.8 dl/g, and combinations thereof, wherein the combined weights of (A) and (B) total 100 percent, said inherent

viscosity (I.V.) being measured at 25°C using 0.50 grams of polymer per 100 ml of a solvent consisting of 60% by weight phenol and 40% by weight tetrachloroethane, and said polyamide being formed from isophthalic acid, terephthalic acid, cyclohexanedicarboxylic acid, *meta*- or *para*-xylylenediamine, 1,3- or 1,4-cyclohexane(bis)methylamine, aliphatic diacids with 6 to 12 carbon atoms, aliphatic amino acids or lactams with 6 to 12 carbon atoms, aliphatic diamines with 4 to 12 carbon atoms."

Claims 2 to 4 are directed to preferred embodiments of the composition of Claim 1.

Claim 5 refers to a process for reducing the acetaldehyde concentration in a heat-moulded polyester article, which comprises dry-blending a polyester composition as defined in Claim 1, melt-kneading the composition and thermoforming the melt-kneaded composition into an article.

IV. The Appellant's arguments submitted in writing and during oral proceedings can be summarised as follows:

(i) Regarding the wording of the claims, the requirements of Article 123(2) EPC were met since the amendments, which aimed at a more specific definition of the polymers, were all supported by the application as originally filed.

(ii) As to novelty, D1 differed in the polyamides

used, which were film-forming, whereas the polyamides used in the present compositions had too low molecular weights to have that capability. Therefore, the claimed subject-matter was novel.

(iii) As regards inventive step, D1 required the polyamides to be "film-forming", that is, to have a high molecular weight. The molecular weights of the polyamides used in Examples 4 and 10 of D1 could not be established either by the terminal amino group method since the polyamide was not linear, or by the viscosity method since a relative viscosity of less than 1 made no sense. The possibility to use low molecular weight compounds also mentioned by D1 explicitly excluded polyamides. Therefore, it taught away from the use of low molecular weight polyamides, as now required. Moreover, D1 did not contain any indication of the surprising reduction of acetaldehyde in combination with an improved transparency, as demonstrated in the examples. Hence, the claimed subject-matter was inventive.

The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 5 as filed during the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

Wording of the claims

2. The amendments to the claims are in conformity with the requirements of Article 123(2) EPC.

2.1 Claim 1 of the main request differs from the one as originally filed in

(a) a lower upper limit for the number average molecular weight and the inherent viscosity of the polyamide; supported by original page 7, lines 22 to 24 and 26 to 28 of the original description;

(b) the indication of the method of measuring the inherent viscosity; specified on original description page 13, lines 15 to 18; and

(c) the definition of the polyamide; disclosed on original description page 8, lines 1 to 7.

2.2 Claim 2 is based on original Claims 6 and 8.

2.3 Claim 3 corresponds to original Claim 7.

2.4 Claim 4 corresponds to original Claim 8.

2.5 Claim 5 finds its counterpart in original Claim 25, adapted to the amended Claim 1.

3. The present wording of the claims also satisfies the requirements of Article 84 EPC.

First, the characterization of the low molecular weight

polyamides by the number average molecular weight as well as by the inherent viscosity corresponds to the dual definition used in the description.

Secondly, the introduction of the inherent viscosity of the polyamides together with an indication of the method used to measure that parameter results in a more specific definition of these polymers, which also contributes to clarity.

Novelty

4. D1 describes a heat-moulded or draw-formed plastic vessel composed of a thermoplastic polyester composition comprising (A) a thermoplastic polyester having ethylene terephthalate units and (B) 3×10^{-7} to 10.0% by weight, based on the polyester, of a thermoplastic polyamide having a terminal amino group at a concentration of 0.05 to 50 millimoles per 100 g of the resin (column 2, lines 11 to 18). According to another embodiment, the thermoplastic polyester (A) may be combined with (C) 5×10^{-7} to 3.0% by weight, based on the polyester, of a primary amino group-containing compound having a molecular weight of at least 100, except polyamide (column 2, lines 22 to 28).

The concentration of amino groups is described as essential to reduce the acetaldehyde concentration caused by thermal decomposition of the polyester, which would affect the flavour and fragrance retaining property of the composition and, thereby, its suitability as constituent material of vessels in contact with food (column 1, lines 11 to 18; column 2, line 29 to column 3, line 5; column 10, lines 41 to

68).

4.1 According to its general definition the polyester is a thermoplastic polyester having ethylene terephthalate units (Claim 1). In practice, it is either a polyester composed mainly of ethylene terephthalate units or a copolyester of terephthalic acid/isophthalic acid/ethylene glycol/bis(2-hydroxyethoxy)benzene (column 3, lines 40 to 46). Further comonomers may be used in so far as they do not affect the inherent properties of polyethylene terephthalate; to that end at least 80 mole% of the acid component should be terephthalic acid and, similarly, at least 80 mole% of the diol component should be ethylene glycol (column 3, lines 48 to 66). This corresponds to the definition of the polyester according to the application in suit.

The polyamide component should contain terminal amino groups at a concentration of 0.05 to 50 millimoles per 100 g of the polyamide resin (column 4, lines 41 to 44). Its molecular weight is however not particularly critical, so far as it has a film-forming property; a relative viscosity of 0.4 to 4.5 is preferred (column 6, lines 36 to 41). Three classes of polyamides are specified:

(i) homopolyamides and copolyamides comprising amide recurring units according to a given formula (column 5, line 8 to column 6, line 18) encompassing the kind of polyamide mentioned in Claim 1 of the application in suit,

(ii) substantially linear co-condensation polyamides comprising 35 to 50 mole% of specified

dicarboxylic acid component units, 35 to 50 mole% of specific diamine component units and 0 to 30 mole% of units of a specified aminocarboxylic acid component (column 6, lines 19 to 28) and

(iii) polymerized fatty acid polyamides obtained by copolycondensing a polymerized fatty acid formed by polymerizing an unsaturated fatty acid (such as linolic or linoleic acid) with a diamine (column 6, line 29 to 34).

4.2 The primary amino group-containing compound (C) which can be used as an alternative to the polyamide (B), contains preferably an amino group concentration of 0.05 to 3000 meq. Various compounds can be used as such (column 6, line 58 to column 8, line 34).

4.3 The examples describe various combinations of:

- polyethylene terephthalate having an inherent viscosity of 0.75 dl/g,

- polyethylene terephthalate having an inherent viscosity of 0.65 dl/g,

- copolyester of terephthalic acid/isophthalic acid at a molar ratio of 70/30 and ethylene glycol/bis(2-hydroxyethoxy)benzene at a molar ratio of 95/5, having an inherent viscosity of 0.69 dl/g,

- copolyester of terephthalic acid/isophthalic acid at a molar ratio of 40/60 and ethylene glycol/bis(2-hydroxyethoxy)benzene at a molar

ratio of 80/20, having an inherent viscosity of 0.55 dl/g,

- polycapramide having a relative viscosity of 3.42 and a terminal amino group concentration of 1.637 millimoles/100 g,

- polyhexamethylene adipamide having a relative viscosity of 1.27 and a terminal amino group concentration of 4.429 millimoles/100 g,

- poly-m-xylylene adipamide having a relative viscosity of 2.24 and a terminal amino group concentration of 0.471 millimoles/100 g,

- polymerized fatty acid polyamide having a relative viscosity of 0.52 and a terminal amino group concentration of 31.08 millimoles/100 g,

- polyoxyethylene diamine of molecular weight 8,000 and an amino group concentration of 25.0 meq/100 g,

- spiroacetal diamine with a molecular weight of 274.4 and an amino group concentration of 729/meq/100 g and

- spiroguanamine with a molecular weight of 434.2 and an amino group concentration of 921 meq/100 g.

Bottles were made from polyester compositions described in the examples, and these were tested. All the examples show an effective reduction of acetaldehyde

concentration in the products made out of the exemplified blends (Tables 1 to 4, 6 and 8).

4.4 From the above it can be seen that the polyamides described in D1 should be of film-forming molecular weight. Accordingly, in all the examples except Examples 4 and 10, the polyamides used have molecular weights well above those required by present Claim 1. The type of polyamide used in Examples 4 and 10 - polymerized fatty acid polyamide - is not included in the definition of polyamides according to present Claim 1, so that for that reason the blends of those examples are outside the scope of Claim 1 regardless of any molecular weight values. Therefore, the claimed subject matter is novel.

Problem and solution

5. The application in suit concerns a polyester/polyamide blend having improved flavour retaining property and clarity.

5.1 Such compositions were known from e.g. D1, which the Board, like the Examining Division and the Appellant, considers to be the closest state of the art.

5.2 Although the compositions described in D1 are said to have low residual acetaldehyde concentration and good transparency, these properties were still capable of improvement.

5.3 According to the description of the application in suit, the object of the invention is to reduce residual acetaldehyde contained in a polyethylene terephthalate

based polyester, to improve the flavour and fragrance retaining property and to provide polyester/ polyamide blends having superior clarity as well as excellent mechanical properties (page 4, line 8 to 23). However, from the examples (Tables I to VI) it appears that only the acetaldehyde concentration of the blends and the haze have been measured, so that no evidence is available regarding the other properties. Therefore, the technical problem underlying the application in suit has to be reformulated on a less ambitious basis as to provide a polyester composition having an improved reduction of acetaldehyde concentration as well as improved transparency.

5.4 According to the application in suit that problem is to be solved by a polyester/polyamide composition in which the polyamide has a specific composition and a low molecular weight, as defined in Claim 1.

5.4.1 The examples and comparative examples in the application (Tables I to VI) show that the two aspects of the above-defined problem are effectively solved. In particular, it has been shown that the compositions according to Claim 1 have an improved reduction of acetaldehyde concentration (Examples 1 to 4 vs. Examples 5 and 6, Example 7 vs. Example 8, Examples 10 to 13 vs. Examples 14 and 15, Examples 17 to 19 vs. Examples 20 to 22) and lower haze (Examples 10 and 12 vs. Example 15, Examples 17 to 19 vs. Examples 20 to 22) as compared to compositions containing polyamides having molecular weights that fall outside the scope of present Claim 1.

Obviousness

6. It remains to be decided whether the claimed subject-matter is obvious having regard to the documents on file.

6.1 Although D1 is mainly concerned with the reduction of the concentration of acetaldehyde in polyester compositions, there is no information about the influence of the individual features of the polymer components on the acetaldehyde residual ratio. Faced with the above defined problem the skilled person could thus only rely on the experimental results provided in the examples.

6.2 Whether one considers compositions derived from polyesters within the terms of the application in suit, e.g. polyethylene terephthalate, or from the specific copolyesters, in all the examples except Examples 4 and 10, the polyamides used have molecular weights higher than the ones required by present Claim 1. The molecular weights of the polyamides used in Examples 1 to 3 and 7 to 9 can be calculated from the terminal amino-group concentration to be 21,000 or higher. By the same method, the molecular weights of the polyamides of Examples 4 and 10 would amount to about 3100, which is within the range indicated in present Claim 1. However, the Appellant stated that this method was not applicable to such polyamides since they were not linear. Also, the relative viscosity could not be used for calculation of the molecular weights, because it should be above 1 according to its definition, so that the value indicated was not meaningful.

The Board considers these arguments to be sufficiently plausible to conclude that the molecular weights of the polyamides used in Examples 4 and 10 are not directly and unambiguously derivable from these examples. In view of the requirement of D1 that the polyamides used should be film-forming, implying a high molecular weight, the Board is satisfied that D1 contains no teaching to use lower molecular weight polyamides.

6.3 Even the alternative embodiment (cf. point 4.3 above), according to which the polyester can be mixed with various amino compounds of low molecular weight would not provide an incentive to operate in accordance with the requirements of the application in suit, since polyamides are explicitly excluded from the definition of compounds (C) as primary amino group-containing compounds with a molecular weight of at least 100 (column 2, lines 22 to 28).

6.4 D1 is thus clearly limited to the use of film-forming polyamides and thereby, although no specific reason is given for the exclusion of lower molecular weight polyamides, teaches away from the use of such compounds. In the Board's view, there would be no reason to depart from that explicit teaching in the expectation of a further reduction of residual acetaldehyde concentration and improved transparency.

6.5 Therefore, D1 does not render obvious the subject-matter of Claim 1.

6.6 For the above reasons, the Board comes to the conclusion that the subject-matter of Claim 1 involves an inventive step.

7. Since Claim 1 is allowable, the same goes for dependent Claims 2 to 4, the patentability of which is supported by that of Claim 1.

The above considerations also apply to Claim 5 since its subject-matter is based on the same combination of features as Claim 1.

8. Although the claims satisfy the requirements of the EPC, a patent cannot be granted according to the Appellant's request in the absence of a description incorporating the substantial amendments to the claims. The case is therefore remitted to the Examining Division for adaptation of the description.

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 with the order to grant a patent on the basis of Claims 1 to 5 as submitted during the oral proceedings and a description yet to be adapted.

The Registrar: The Chairman:

E. Görgmaier C. Gérardin