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D E C I S I O N
of 22 October 1998

Case Number: T 0927/95 - 3.3.3

Application Number: 82107642.9

Publication Number: 0073036

IPC: C08L 77/00

Language of the proceedings: EN

Title of invention:
Toughened polyamide blends

Patentee:
E. I. Du Pont De Nemours and Company

Opponent:
BASF Aktiengesellschaft, Ludwigshafen
Bayer AG, Leverkusen Konzernverwaltung RP Patente Konzern
EMS - Inventa AG

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (no) - obvious combination of known features
with known effect"

Decisions cited:
T 0551/89

Catchword:
-



Case Number: T 0927/95 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 22 October 1998

Appellant 1:
(Opponent 03)

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Other party:
(Opponent 02)

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Respondent:
(Proprietor of the patent)

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Decision under appeal:

Interlocutory decision of the Opposition Division
of the European Patent Office posted 27 September
1995 concerning maintenance of European patent
No. 0 073 036 in amended form.

Composition of the Board:

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

Summary of Facts and Submissions

- I. Mention of the grant of European patent No. 0 073 036 in respect of European patent application No. 82 107 642.9, filed on 20 August 1982, claiming priority from an earlier application in the USA (295018 of 21 August 1981), was announced on 6 May 1987 on the basis of twelve claims, Claim 1 reading as follows:

"A blend containing

(I) 99-60% by weight of thermoplastic polyamides comprising complementally

(a) about 20 to 90 percent by weight, based on total polyamide, of a thermoplastic semicrystalline polyamide of film-forming molecular weight,

(b) about 80 to 10 percent by weight, based on total polyamide, of a thermoplastic amorphous polyamide obtained from at least one aromatic dicarboxylic acid containing 8-18 carbon atoms and at least one diamine selected from the class consisting of

- i) 2-12 carbon aliphatic straight-chained diamine,
- ii) 4-18 carbon branched aliphatic diamine, and
- iii) 8-20 carbon cycloaliphatic diamine containing at least one cyclohexane moiety,

and wherein optionally, up to 50 weight percent of the amorphous polyamide may consist of units obtained from lactams or ω -aminoacids containing 4-12 carbon atoms, or from polymerization salts of aliphatic dicarboxylic acids containing 4-12 carbon atoms and aliphatic diamines containing 2-12 carbon atoms, with complementally

(II) 1-40% by weight of blend of a toughening agent having a maximum tensile modulus of 55,160 N/cm² (80,000 psi) determined according to ASTM D-638, and that is an

organic polymer of film-forming molecular weight which imparts to Component a) a notched Izod value at least 50% greater than the notched Izod value of component a) alone when 20% by weight of the toughening agent is present with Component a)."

Claims 2 to 9 referred to preferred embodiments of the blend of Claim 1.

Claim 10 was directed to a process for preparing the blend of Claim 1 and Claims 11 and 12 referred to processes for preparing articles from the blend of Claim 1.

II. On 2 February 1988, 4 February 1988 and 4 February 1988 three Notices of Opposition against the granted patent were filed, in which the revocation of the patent in its entirety was requested on the grounds set out in Article 100(a) EPC.

III. By an interlocutory decision delivered orally on 13 July 1995 and issued in writing on 27 September 1995, the Opposition Division held that there were no grounds of opposition prejudicing the maintenance of the patent in amended form, i.e. on the basis of Claim 1 filed as auxiliary request on 13 July 1995 and Claims 2 to 4, 7 and 10 to 12 as granted, the amendments being:

- (1) the definition of the polyamide of Component a) as being polyamide 66,
- (2) the method of determination of the maximum tensile modulus of the toughening agent II of Component b),
- (3) the method of determination of the notched Izod value of Component a) and

(4) the toughening agent II of Component b) as being selected from

- (i) polymers of ethylene, at least one α -olefin of 3 to 6 carbon atoms, and at least one unsaturated monomer taken from the class consisting of branched, straight chain and cyclic compounds having from 4 to 14 carbon atoms and having at least one additional nonconjugated unsaturated carbon-carbon bond, in which said polymer has grafted thereto an unsaturated monomer taken from the class consisting of a) α,β -ethylenically unsaturated dicarboxylic acids having from 3 to 8 carbon atoms, and derivatives thereof taken from the class consisting of monoesters of alcohols of 1 to 29 carbon atoms, b) anhydrides of the dicarboxylic acids, c) the metal salts of the dicarboxylic acids and the monoesters of said dicarboxylic acid having from 0 to 100 percent of the carboxylic groups ionized by neutralisation with metal ions,

- (ii) polymers of ethylene and unsaturated monomers taken from the class consisting of (a) α,β -ethylenically unsaturated carboxylic acids (preferably) having from 3 to 8 carbon atoms, and derivatives of the acid taken from the class consisting of a) monoesters of alcohols of 1 to 29 carbon atoms, b) dicarboxylic acids and anhydrides of the dicarboxylic acids and c) metal salts of the monocarboxylic, dicarboxylic acids and monoesters of the dicarboxylic acid having from 0 to 100 percent of the carboxylic acid groups ionized by neutralization with metal ions, or

(iii) organic polymers which comprises a first phase polymerized from monomers comprising 50 to 99.9 parts by weight alkyl acrylate wherein the alkyl group contains one to 15 carbon atoms, butadiene or substituted butadiene; 0 to 40 parts by weight of other ethylenically unsaturated monomers; 0 to 5 parts by weight of a polyethylenically unsaturated cross-linking monomer; and 0 to 5 parts by weight of a graft-linking monomer; and a final rigid phase thermoplastic stage containing amine-reactive carboxylic acid groups and polymerized from monomers comprising 1 to 50 parts by weight of a copolymerizable carboxylic acid, 50 to 99 parts by weight of a member selected from the group consisting of alkyl methacrylates, styrenes, acrylonitrile, methacrylonitrile and olefins that when homopolymerized, form polymers having a heat distortion temperature greater than about 20°C; 0 to 49 parts by weight of another acrylic monomer; and 0 to 40 parts of another copolymerizable ethylenically unsaturated monomer."

IV. The above decision was essentially based upon the following documents:

D1: DE-A-1 769 040 and

D5: DE-B-2 622 973.

The Opposition Division held that the amendments in the definitions of the crystalline polyamide and the toughening agent had overcome the objection of lack of novelty and that, consequently, the only point at issue was the question of the presence of an inventive step.

In that respect, D5 was considered as the starting point to achieve the object underlying the invention: polyamide based moulding compositions which exhibited high toughness, solvent resistance and dimensional stability. D5 disclosed compositions of polyamide, amongst which polyamide 66 (Nylon-6,6), and a toughening agent. In the light of the information contained in D1, from which it was clear that replacing part of polyamide 66 by an amorphous polyamide did not result in increased toughness, it was not obvious for the skilled person to do so in order to improve the impact resistance of the compositions according to D5.

- V. An appeal was lodged against that decision on 7 November 1995 by Opponent III (hereinafter Appellant 1) and on 24 November 1995 by Opponent I (hereinafter Appellant 2), the appeal fees being paid simultaneously in both cases. The Statements of Grounds of Appeal were filed on 6 February 1996 and 12 January 1996, respectively.

The Proprietor of the patent (Respondent), who had filed an appeal on 29 November 1995 together with payment of the prescribed fee, did not file a Statement of Grounds for Appeal. His submission of 22 August 1996, in which he confirmed that the Proprietor had lost interest in the further prosecution of the initial main request (to set aside the decision under appeal and to maintain the patent as granted), was a proper Counterstatement of Appeal containing detailed arguments in favour of the maintenance of the patent as amended before the Opposition Division. On 23 September 1998 the Respondent informed the EPO that he would not attend the oral proceedings scheduled on 22 October 1998 and asked for a decision on the merits of the case.

Opponent II, who did not play an active role in the appeal proceedings, but as a party to the proceedings under Article 107 EPC had been duly summoned to the hearing, informed the EPO by letter of 5 October 1998, that he would not attend the oral proceedings and also asked for a decision on the merits of the case.

VI. The Appellants, in their written submissions as well as during the oral proceedings argued essentially that D1 was the closest state of the art and that it was obvious to improve the impact strength of the mixture of Nylon-6,6 and amorphous polyamide disclosed therein by adding a toughener, as was done in D5. The presence of a surprising synergistic effect was not proven, as demonstrated by a number of additional experiments. Even when starting from D5, which was directed to blends of polyamides and toughening polymers, as the closest prior art document, it was, in the light of the teaching of D1, obvious to consider ternary mixtures based on a combination of amorphous polyamide with semi-crystalline polyamide for improvement of impact strength as an alternative for the usual impact modifiers.

VII. The Respondent contended that D5 was the proper starting point since the object of the patent in suit was to provide polyamide compositions having the highest possible impact strength. As it could not be concluded from D1 that the addition of amorphous polyamide to a mixture of Nylon-6,6 and toughener would improve the impact resistance to the extent shown in the examples of the patent in suit, a surprising synergistic effect was present and the claimed subject-matter was inventive.

VIII. The Appellants requested that the decision under appeal be set aside and the patent be revoked.

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

Article 123 EPC

2. The Board concurs with the Opposition Division that the amendments do not result in subject-matter extending beyond the content of the application as originally filed, not do they extend the protection conferred, so that the requirements of Article 123(2) and (3) EPC are met.

Novelty

3. The Opposition Division as well as the parties agreed that the claimed subject-matter as amended during the proceedings before the first instance was novel and the Board sees no reason to deviate from that view.

Closest document

4. The patent in suit concerns toughened polyamide blends which, according to the specific definition of the thermoplastic semi-crystalline polyamide introduced in the opposition proceedings, must comprise a polyamide 66 of film-forming molecular weight. Such blends are described in both D1, which the Respondent held to represent the closest state of the art, as well as in

D5, which the Appellants deemed to be the proper starting point. The question which of the two documents was the closest, was elaborately discussed in writing as well as during the oral proceedings before the Board.

- 4.1 D1 describes thermoplastic polyamide compositions consisting of mixtures of 1 to 99 parts per weight of a partially crystalline polyamide based on ω -amino acids or ω -lactams or derived from aliphatic dicarboxylic acids and straight chain aliphatic diamines, and 99 to 1 parts per weight of an amorphous polyamide based on aromatic dicarboxylic acids and 2,2,4- and/or 2,4,4-trimethylhexamethylene diamine or based on aromatic dicarboxylic acids and straight chain aliphatic diamines with 6 to 12 carbon atoms (Claim 1).

The essence of the teaching of D1 is that the properties of partially crystalline polyamides, which in the light of the enlisted polymers should be understood to be aliphatic polyamides, can be improved by the addition of amorphous polyamide, thus avoiding the disadvantages of the usual inorganic filling materials, such as a lowered impact strength and increased abrasiveness (page 1 and page 2, paragraph 2). The semi-crystalline/amorphous polyamide mixtures do not possess the disadvantages of the individual polyamide components, such as unfavourable melt viscosity and high abrasiveness. On the contrary, they show an improved rigidity and dimensional stability of the moulded products. In particular, products made out of those mixtures are harder, more resistant to breaking, more heat resistant and have an increased impact strength. The latter shows an optimum at a proportion of the amorphous polyamide of 40 to 60 weight % of the mixture (page 4, second full paragraph).

The examples illustrate that property for several blends of a semi-crystalline aliphatic polyamide (Nylon-6 in Examples 1 to 5; Nylon-6,6 in Examples 6 to 10; Nylon-6,10 in Examples 11 to 15; Nylon 11 in Examples 16 to 20; Nylon 12 in Examples 21 to 25; and Nylon-6,6 in Examples 26 to 30) with an amorphous polyamide. Although the examples directed to Nylon-6,6 do not show the optimum impact strength at 40 to 60 weight % amorphous polyamide indicated before, in all cases the impact strength of the samples containing both the amorphous polyamide and the semi-crystalline polyamide is increased. Therefore, the general teaching of D1 is that mixing an amorphous polyamide with a semi-crystalline one improves a number of its properties, in particular the impact strength.

4.2 D5 describes toughened multiphase thermoplastic compositions with 60 to 99% by weight polyamide matrix resin having a number average molecular weight of at least 5,000, and from 1 to 40% by weight of a dispersed phase of a specified straight-chain or branched polymer having a particle size in the range of from 0.01 to 1,0 μm , containing adhesive sites to the polyamide resin and having specific mechanical properties (Claim 1). The incorporation of the disperse phase polymer, which can be an elastomer or a non-elastomeric thermoplastic polymer (column 5, line 63 to column 8, line 38), aims at improving the impact resistance of polyamide compositions (column 2, line 59 to column 3, line 11). The polyamide matrix can be made of a semi-crystalline or an amorphous polyamide (column 5, lines 11 to 40). In practice, the polyamides regarded as suitable encompass a broad class of polymers, e.g. aliphatic and aromatic polyamides as well as terpolyamides and copolyamides. Thus, even if Nylon-6,6 is explicitly enlisted and actually used in numerous examples, it can by no means be regarded as an essential ingredient of the compositions disclosed in

this citation. On the contrary, it is clear that other features, in particular the granularity and tensile modulus of the toughening agent, have a major influence on the properties of the toughened polyamide compositions (column 5, lines 42 to 59).

In accordance with the general teaching of the citation, the compositions exemplified in D5, which refer to Nylon-6,6 (Examples 1 to 115 and 137 to 163, Comparative Examples A to S and Y to DD), amorphous polyamide (Examples 131 to 133 and Comparative Example W) and other polyamides (Examples 116 to 130 and 134 to 136, Comparative Examples T to V and X) all exhibit high notched-impact strength values compared to the polyamide without the toughening agent.

In conclusion, the teaching of D5 is that adding certain types of toughening agents to a relatively broad class of polyamides, be they amorphous or semi-crystalline, improves their impact strength.

- 4.3 In the light of the disclosures of D1 and D5 (points 4.1 and 4.2 above), it is clear that, whilst both D1 and D5 address the problem of impact strength improvement of polyamide compositions, D1 is specifically concerned with semi-crystalline polyamide, including Nylon-6,6 and should, consequently, be regarded as the closest state of the art.

Inventive step

5. Following the amendments in Claim 1 the patent in suit relates to toughened polyamide blends containing Nylon-6,6 as the thermoplastic semi-crystalline polyamide.

- 5.1 Although articles moulded from blends of semi-crystalline and amorphous polyamides of D1 are said to have good rigidity and dimensional stability, to be harder, more resistant to breaking, more heat resistant and to have an increased impact strength as compared to its individual components (whole page 1), these properties were to some extent impaired by the presence of various customary additives, such as fillers, flame retardants and stabilizers. In particular, the blends of Nylon-6,6 and amorphous polyamide did not show any consistent tendency towards toughening, as characterized by notched Izod measurements.
- 5.2 In view of this shortcoming, the technical problem underlying the patent in suit may thus be seen in the improvement of the impact strength of mixtures of Nylon-6,6 and amorphous polyamide.
- 5.3 According to the patent in suit that problem is to be solved by compositions additionally containing an impact modifier or toughening agent as defined in Claim 1.
- 5.4 It was not disputed by the parties, and confirmed by all examples on file, that the claimed compositions indeed have an improved impact resistance over the compositions of D1, so that the problem is effectively solved.
6. It remains to be decided whether the claimed subject-matter was obvious having regard to the documents on file.

According to the general teaching of D5, the addition of a polymer toughening agent to a semi-crystalline or an amorphous polyamide results in improved impact strength. In the light of that teaching the skilled person would not hesitate to add a toughening agent to

a blend of semi-crystalline polyamide and amorphous polyamide in the expectation that the impact resistance would increase. Even if an unexpected additional or synergistic effect would occur, this would merely amount to the scale of the effect surpassing the skilled person's anticipations, not to a completely unforeseen event. The extent of the effect does not contribute anything to the creative effort of the skilled person and therefore cannot make an obvious measure inventive (T 551/89 of 20 March 1990, not published in OJ EPO).

7. The situation would not be different if D5 were used as the starting point. D5 discloses mixtures of polyamides with toughening agents, which are said to have a good impact resistance. The problem to be solved could then be defined as a further improvement of the mechanical properties of those blends. The examples in the patent indeed demonstrate such improvement, which, however, in the light of the general teaching of D1 (see point 3.1 above), was to be expected, especially since it is explicitly stated that the mixtures of D1 can also contain all the usual additives (page 4, first full paragraph). Also if the improvement in impact strength is not consistent, as demonstrated by the additional experiments filed by the Appellants, and the problem should be reformulated to merely providing an alternative to the known mixtures of semi-crystalline polyamide and toughening agent, the teaching of D1 provides a clear incentive for the skilled person to add an amorphous polyamide to the mixture of semi-crystalline polyamide and toughening agent as disclosed in D1, with the composition now being claimed as the result.

The specification of the amorphous polyamide and the toughening agent which can be part of the composition according to Claim 1 does not render the claimed subject-matter inventive either, since i) no special effects attributable to those specific compounds have been demonstrated and ii) those compounds have been explicitly mentioned in either of D1 and D5.

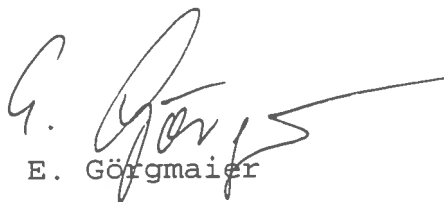
8. Whatever the starting point, therefore, the claimed subject-matter must be regarded as an obvious combination of features known from the prior art, which, as a consequence, does not involve an inventive step.

Order

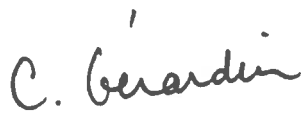
For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The patent is revoked.

The Registrar:


E. Görgmaier

The Chairman:


C. Gérardin

