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DECISION
of 15 February 1995

Case Number: T 0572/92 - 3.3.3

Application Number: 87107757.4

Publication Number: 0248352

IPC: C08L 67/02

Language of the proceedings: EN

Title of invention:

Polyester-based shock-resistant compositions and process for their preparation

Applicant:

ENICHEM S.p.A.

Opponent:

-

Headword:

-

Relevant legal provisions:

EPC Art. 56

Keyword:

"Inventive step (no - effect unrelated to distinguishing features)"

Decisions cited:

-

Catchword:

-



Case Number: T 0572/92 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 15 February 1995

Appellant:

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Decision under appeal:

Decision of the Examining Division of the European Patent Office dated 13 November 1991 with written reasons posted on 20 January 1992 refusing European patent application No. 87 107 757.4 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: C. Gérardin
Members: P. Kitzmantel
M. Aúz Castro

Summary of Facts and Submissions

I. This appeal, which was filed on 19 March 1992, lies against the decision of the Examining Division issued orally on 13 November 1991 and posted on 20 January 1992 refusing European patent application No. 87 107 757.4, filed on 27 May 1987, published under the No. 0 248 352 and claiming the priority of 29 May 1986 from an IT application.

The appeal fee was paid together with the notice of appeal and the statement of grounds of appeal was filed on 26 May 1992.

II. The decision under appeal was based on Claim 1 of the main request filed on 6 November 1991, and on Claim 1 of the auxiliary request filed on 13 November 1991.

Claim 1 of the main request reads as follows:

- "Shock-resistant polymeric compositions comprising
- (a) from 55 to 98% by weight of poly(butylene terephthalate) having an intrinsic viscosity of 1.4 dl/g;
 - (b) from 1 to 35% by weight of butadiene rubber selected from butadiene/styrene/acrylonitrile and butadiene/styrene/methylmethacrylate terpolymers having a glass transition temperature (T_g) lower than 0°C and a crosslinking degree (gel content) of from 50 to 99% by weight; and
 - (c) from 1 to 10% by weight of an elastomeric copolyester."

The further Claims 2 to 5 of the main request were dependent on Claim 1 and corresponded to Claims 7 to 10 as originally filed (see Minutes of the Oral Proceedings, point 1).

Claim 1 of the auxiliary request differed from the main request in that the elastomeric copolyester was a specific polyester ether block copolymer, as defined in original Claim 7.

III. The decision under appeal held that both requests related to novel subject-matter which, however, did not involve an inventive step.

From the prior art disclosed in

(1) US-A-4 200 567

the compositions according to Claim 1 of the main request would be literally different by the selection of:

- (i) a poly(butylene terephthalate) ("PBT") having an intrinsic viscosity ("I.V.") of 1.4 dl/g;
- (ii) a rubbery terpolymer having a crosslinking degree (gel content) of 50 to 99% by weight; and
- (iii) the rubbery terpolymer having a $T_g < 0^\circ\text{C}$.

Considering the presence of polybutadiene blocks and crosslinking agents in the rubber component (multiphase composite interpolymer) used in the compositions according to (1), the above features (ii) and (iii) would, however, be implicit to and thus not distinguishing over the whole disclosure of (1). Even if regarded as distinguishing, these features could not contribute to an inventive step, because their critical

importance for the desired low temperature impact strength was unproved. Furthermore, a beneficial influence of the high I.V. of 1.4 dl/g of the PBT on the impact strength of the claimed compositions, even if recognized, would anyway have been expectable from the teaching in (1), namely that low I.V. polyesters were generally less responsive to impact modifiers.

According to the decision under appeal the same arguments applied also to the disclosure of document

(5) JP-A-61-42562 (English translation),

which differed from the claimed subject-matter by the same features (i), (ii) and (iii) and also stressed that good impact resistance at low temperatures - among other favourable properties - could be achieved.

Since the polyester ether copolymers used as elastomeric copolyesters according to the auxiliary request would be preferred constituents of the compositions disclosed in document (1), the decision under appeal held that this additional feature could not contribute to an inventive step either.

The request of the Appellant, submitted during the oral proceedings before the first instance, to continue the procedure in written form, was refused; it was held that there had been ample opportunity for the Appellant to present and argue its case.

IV. Together with the grounds of appeal the Appellant withdrew its previous auxiliary request and submitted as the basis of a new auxiliary request the following

Claim 1:

- "Shock-resistant polymeric compositions comprising
- (a) from 55 to 98% by weight of poly(butylene terephthalate) having an intrinsic viscosity of 1.4 dl/g;
 - (b) from 1 to 35% by weight of butadiene/styrene/methylmethacrylate terpolymer having a glass transition temperature (T_g) of -80°C and a crosslinking degree (gel content) of 94% by weight; and
 - (c) from 1 to 10% by weight of an elastomeric copolyester."

The arguments presented by the Appellant in support of its appeal may be summarized as follows:

While the decision under appeal correctly pointed out that the subject-matter of the main request was different over document (1) by the triple selection of I.V. of the PBT, cross-linking degree and T_g of the terpolymer, it was surprising - contrary to said decision - that thereby the desired high impact strength could be achieved. This must be concluded because (1), although disclosing an I.V. range of 0.5 to 1.4 dl/g, preferred low I.V. materials and, according to the evidence therein, did not suggest that higher Izod values of the compositions could be attained by enhancing the I.V. of the PBT. As to the degree of crosslinking, this feature amounted to a true selection not comprised by (1), and the same conclusion held also for the T_g-feature, which was not proved to be implicit to the disclosure of (1) by the statements in document

(12) Kirk-Othmer, Encyclopedia of Chemical Technology, 3rd Ed., Vol. 8., page 609 (1979).

Furthermore, the two Comparative Examples corresponding to Examples 42E and 46E of document (1), which had been

filed during the first instance procedure with letter of 3 January 1991, would clearly show a substantial improvement of the Izod values; the decision under appeal was wrong in disregarding these results on the ground that they did not identify the contribution to the Izod improvement of each individual inventive feature.

- V. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the main request or of the auxiliary request filed 26 May 1992.

A subsidiary request for oral proceedings was withdrawn on 13 October 1994.

Simultaneously, the Appellant requested the Board to "decide upon the pending appeal on the merits of this case."

Reasons for the Decision

1. The appeal is admissible.
2. *Article 123(2) EPC*

Claim 1 of the main request is based on Claim 1 as originally filed, on the worked Examples 2 to 4, 13 and 15 (PBT of I.V. of 1.4 dl/g), on page 5, lines 10 to 12 of the original description

(butadiene/styrene/acrylonitrile [ABS] and butadiene/styrene/methylmethacrylate [MBS] terpolymer) and on Claim 5 as originally filed ($T_g < 0^\circ\text{C}$ and gel content 50 to 99%).

With regard to the specific definition of the rubber component (b) in Claim 1 according to the auxiliary request, it is based on Examples 2 to 4, 13 and 15 of the original description.

These claims comply thus with the requirement of Article 123(2) EPC.

3. *Novelty*

It was acknowledged in the decision under appeal that the subject-matter of Claim 1 of the main request was novel over the cited prior art; the Board concurs with that opinion (for a more detailed analysis cf. section 4 of this decision).

Claim 1 of the auxiliary request is narrower in scope than Claim 1 of the main request and its subject-matter is therefore likewise novel.

4. *Inventive step, main request*

4.1 Document (1) discloses a synergistic impact modifier system for poly(alkylene terephthalate), especially for PBT in the I.V. range of 0.5 to 1.4 dl/g (determined in phenol/tetrachloroethane 60/40), consisting of

(A) a thermoplastic segmented copolyester elastomer and

(B) a multiphase composite interpolymer comprising a rigid thermoplastic phase polymerized in the presence of an elastomeric phase (cf. Claim 1; col. 2, lines 46 to 51).

More specifically, the interpolymer (B) comprises either a crosslinked rubbery acrylic or butadiene type first stage graftlinked to an outer rigid thermoplastic phase

(column 3, lines 29 to 54). The compositions used in Examples 42E and 42F, which comprise a) 55 parts of PBT having an I.V. of 0.9 dl/g; b) 7.5 respectively 10 parts of MBS core-shell polymer not identified by its physical parameters; and (c) 7.5 respectively 5 parts of a segmented copolyester elastomer sold under the trademark Hytrel 4056 (Example 42 (referring to Examples 41 and 40) in conjunction with Examples 22, 32 and 35) are particularly relevant. With regard to these particular compositions, which must be regarded as the closest state of the art, the claimed subject-matter differs by

- (i) a higher I.V. of 1.4 dl/g of the PBT; it is noted that, according to the original description of the application in suit (cf. page 3, lines 2 to 4 from the bottom), the I.V. was determined in a 1:1 phenol/tetrachlorethane mixture, not in a 60/40 mixture as in (1);
- (ii) a crosslinking degree (gel content) of the MBS being from 50 to 99% by weight; and
- (iii) a Tg of the butadiene/styrene/methylmethacrylate (MBS) being below 0°C (it is noted that according to page 6, 2nd paragraph of the application in suit the "main" Tg of the rubber component is meant).

4.2 It was argued in the decision under appeal that "according to general background knowledge" (supported by document (12)) the Tg feature (iii) would be an implicit characteristic of the MBS terpolymers used in (1) and that it was "doubtful" whether said MBS differed in its gel content (feature (ii)) from that according to present Claim 1. The appealed decision in its point 3.2, last paragraph, concluded therefrom that features (ii) and (iii) "are regarded as being inherent to the compositions used in (1)".

In the Board's opinion, irrespective of whether or not these allegations are factually correct, they are not conclusive from the arguments presented. Neither discloses document (12) a general rule for calculating the "main" Tg of the butadiene/styrene core of a core/shell polymer as used in (1) - which need not, as presumed in the appealed decision, necessarily be a block copolymer -, nor has it been made plausible that the amounts of divinyl benzene (DVB) and 1,3-butylene dimethacrylate (BGDMA) contained in the core/shell polymer according to Example 22 (used in the compositions of Examples 42E and 42F of (1)) must cause a gel content in the range of 50 to 99%; the reference in the appealed decision to page 6, lines 4 and 13 of the application in suit concerns acrylic rubbers and is of no consequence for the butadiene rubbers of present Claim 1.

The features (ii) and (iii) have therefore not been demonstrated to be within the disclosure of document (1).

4.3 Since the Appellant regards as the object of the alleged invention the provision of compositions endowed with enhanced shock resistance, it has to be examined whether the claimed compositions, in particular features (i) to (iii) thereof, really provide an improvement in that respect over the known compositions, i.e. whether it is proper to define the problem underlying the application in suit in such positive terms.

4.3.1 The Appellant's allegation of a surprisingly enhanced impact strength is based on its Comparative Examples 42E₁ and 46E to Examples 42E and 46E of document (1) filed with Appellant's letter of 3 January 1991:

	PBT amount/I.V.		core-shell polymer amount/type/Tg/gel-%		polyester amount/type
Claim 1 (appl.)	55-98%	1.4	1-35% ABS/ MBS	<0°	50-99 1-10%
42E (1)	55%	0.90	7.5% MBS (Ex.22)	? ?	7,5% Hytrel 4056
Comp. 42E	55%	1.4	7.5% MBS (KM653)	-80° 94	7.5% Hytrel*
46E (1)	50%	0.90	10% MBS (Ex.22)	? ?	10% Hytrel 4056
Comp. 46E	50%	0.90	10% MBS (KM653)	-80° 94	10% Hytrel*

ABS = acrylonitrile butadiene styrene rubber;
MBS = methyl methacrylate butadiene styrene rubber;
MBS (Ex.22) = butadiene-styrene/methyl methacrylate/styrene/methylmethacrylate rubber of Example 22 of (1);
MBS (KM653) = grafted butadiene-styrene-methacrylate terpolymer with a crosslinking degree of 94% and a Tg of -80°C;
Hytrel 4056 = segmented copolyester elastomer, not further defined in (1);
Hytrel* = segmented copolyester elastomer obtained from terephthalic acid, 1,4-butanediol and polyoxytetramethylene glycol of MW = 1000, content of rigid segments of 33%; in the third paragraph on page 6 of the grounds of appeal the Appellant states that, owing to the use of Hytrel*, which was a commercial product, its comparative examples would be truly comparable; neither in this statement, nor in the letter of 3 January 1991 submitting these comparative examples, the Appellant stated clearly that the copolyester elastomer used (Hytrel*) was

indeed Hytrel 4056; there remains thus some doubt that Hytrel* and Hytrel 4056 are identical, but for the purpose of this decision identity is assumed (however, the correctness or not of this assumption is without consequence for this decision).

According to the Appellant, by changing from Example 42E to Comparative Example 42E the Izod value (ASTM D-256) was enhanced from 45 J/m to 90 +/- 10 J/m; the Izod improvement of Comparative Example 46E over Example 46E was from 262 J/m to 400 J/m.

4.3.2 For the recognition of a technical effect being the consequence of a new technical feature or new technical features, it is necessary that this effect is proved to be caused by this (these) new feature(s) only (here features (i), (ii) and (iii) as set out in section 4.1 above); in comparative examples to the state of the art all other features should therefore be kept unchanged in order to justify the conclusion that it is the new feature(s) which is/are responsible for said technical effect.

4.3.3 These conditions are not fulfilled in the present case for the MBS component.

The MBS used in Examples 42E and 46E of document (1) is defined by the monomer weight ratios (butadiene-styrene//methyl methacrylate-divinyl benzene//styrene//methyl methacrylate-1,3-butylene dimethacrylate) in the different stages of its preparation by multistage emulsion polymerization (see Example No. 22 in the table on top of col. 5), but nothing is said in (1) concerning its crosslinking degree (feature (ii)) and its "main" Tg

(feature (iii)). It is therefore not possible to make any conclusion as to what extent these two properties of the MBS KM653 (grafted butadiene-styrene-methacrylate terpolymer with a crosslinking degree of 94% and a Tg of -80°C: see page 9, component (b) of Examples 1 to 4 of the original description of the application in suit) used in the Appellant's Comparative Examples 42E and 46E, separately or together, contribute to the demonstrated enhancement of the Izod impact strength. Such a conclusion would only be possible if the extent of the change of Tg and crosslinking degree were known and if the Izod impact improvement could clearly be imputed to this change.

In the present situation neither is the said extent of change known, nor would it be possible - due to the unknown chemical and physical characteristics (e.g. structure, particle size and proportion of elastomeric and rigid portions) of the MBS components to be compared - to recognize that such change of Tg and crosslinking degree, were it known, was the cause for the ascertained improvement of the Izod impact strength.

It is also not possible to attribute the Izod impact strength enhancing effect to the change of the I.V. of the PBT component from 0.9 dl/g in Examples 42E and 46E of (1) to 1.4 dl/g in the comparative examples, because the unspecified structural and property-wise difference between the MBS components used in the Examples and Comparative Examples 42E and 46E precludes any such conclusion.

4.3.4 Thus the effect of features (i), (ii) and (iii), individually or in combination, on the Izod impact strength of the compositions, if any, is unknown,

and the alleged enhancement of this property by these features cannot therefore be considered as technical solution of a corresponding problem. In particular it has not been demonstrated that, in consequence of features (i), (ii) and (iii), the impact resistance properties achieved with the claimed compositions went beyond the synergistic effect observed in document (1).

This means that the technical problem underlying the application in suit cannot be defined in the positive terms suggested by the Appellant (see point 4.3 above).

4.3.5 Under these circumstances the issue of inventive step boils down to the question whether the combined selection of features (i), (ii) and (iii) was an obvious **alternative** or not to the same features of the corresponding components as used in (1).

PBT having an I.V. of 0.5 to 1.4 dl/g was contemplated in (1) column 2, lines 46 to 51 and so was a certain degree of crosslinking (feature (ii)) of the butadiene terpolymer (cf. the use of divinylbenzene and 1,3-butylene dimethacrylate in the MBS according to Example 22 of (1)); in the absence of proof for a particular unexpected technical effect resulting from the use of PBT of an I.V. of 1.4 dl/g and/or of a MBS having a degree of crosslinking (gel content) within the range of from 50 to 99% by weight, these figures must be considered to be the result of an arbitrary choice from options which have been suggested in document (1). As to the use of a MBS terpolymer having a Tg below 0°C, it is textbook knowledge that almost all elastomers possess Tgs which are far below 0°C (see Encyclopedia of Polymer Science and Technology,

Vol. 5, page 433, Table 17 (1966): e.g. poly(butadiene-co-styrene) with a styrene content of 23% and 36%, respectively, has Tgs of -56°C and -38°C ; since the skilled person knows that the improvement of the impact strength of polymer compositions is linked to the incorporation of elastomeric domains, it was obvious that the terpolymer impact modifiers to be used in the present PBT compositions should have a main Tg below 0°C .

Consequently, the combination of

- (i) a PBT having an I.V. of 1.4 dl/g,
- (ii) a gel content of the butadiene rubber of from 50 to 99% by weight, and
- (iii) a Tg of the butadiene rubber of below 0°C

must be regarded as an obvious choice for one skilled in the art intending to formulate shock-resistant PBT compositions of characteristics similar to the ones disclosed in document (1).

The subject-matter of Claim 1 of the main request does therefore not involve an inventive step.

4.4

The same negative inventive step conclusion is arrived at when starting from document (5) which describes ternary compositions containing a linear saturated polyester which is a polytetramethylene terephthalate (PBT), a rubber reinforcing resin, and a polyester block copolymer. The specific definitions given for the rubber reinforcing resin and the polyester block copolymer (page 6/17, paragraph 2 to page 8/17, paragraph 1) show that both are within the terms of the application in suit; it follows that, like in the case of document (1), the prior art compositions according

to document (5) differ from the subject-matter of Claim 1 by the same features (i), (ii) and (iii). In the absence of evidence proving an unexpected improvement of the impact strength of the claimed compositions over those according to document (5), the problem underlying the subject-matter under consideration can again only be seen in the provision of further impact resistant PBT compositions. In view of the arbitrary character of the distinguishing features (i) and (ii), and of the obviousness of feature (iii) (cf. section 4.3 above) they cannot contribute to an inventive step; Claim 1 of the main request is therefore directed to an obvious alternative of the compositions according to document (5).

5. *Inventive step, auxiliary request*

5.1 Claim 1 of the auxiliary request differs from that of the main request by a restriction of the butadiene rubber component to MBS having a Tg of -80°C and a gel content of 94% by weight.

The problem to be solved by these features, with respect to documents (1) and (5), is the same as that for the main request, namely the provision of further PBT compositions having good Izod impact strength.

Since these new features correspond exactly to the properties of the MBS "KM653" used in the Appellant's Comparative Examples 42E and 46E, the conclusions drawn with respect to these examples in section 4.3 above are fully applicable to the subject-matter of the auxiliary request. It follows that the values of -80°C for the Tg and of 94% for the gel content of the MBS have not been shown to be

responsible for the demonstrated improvement of the impact strength. Their choice amounts therefore to an obvious, arbitrary selection from the options which the skilled person had at its disposal.


Consequently, the subject-matter of Claim 1 of the auxiliary request does not involve an inventive step either.

Order

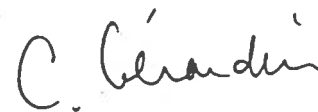
For these reasons it is decided that:

The appeal is dismissed.

The Registrar:


E. Görgmaier

The Chairman:


C. Gérardin

