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Application No.: 81 305 170.3

Publication No.: 0 051 471

Title of invention: Tough thermoplastic nylon compositions and
processes for preparing them

Classification: C08L 77/00

D E C I S I O N
of 24 October 1990

Proprietor of the patent: Monsanto Company

Opponent: BASF Aktiengesellschaft

Headword:

EPC Article 56

Keyword: "Inventive step (affirmed)"

Headnote

Summary of Facts and Submissions

I. The grant of European patent No. 51 471 in respect of European patent application 81 305 170.3, filed on 30 October 1981 claiming two US priorities of 3 November 1980 and 30 September 1981 was announced on 5 June 1985 (cf. Bulletin 85/23).

II. A notice of opposition was filed by the Appellant on 25 January 1986 requesting revocation of the patent based on Art. 100(a). Originally there was also an objection under Art. 100(b) EPC, which objection was later withdrawn. The opposition was supported by

- (1) US-A-3 796 771 (considered during examination)
- (2) Technisches Merkblatt Ultramid^(R)-E-Marken, BASF, September 1971.

During the opposition proceedings further documents were discussed, including

- (3) EP-A-3 126 (state of the art acknowledged on page 2, line 29 of the patent in suit) and
- (4) The description of a polybutadiene latex designated FR-S 2004 (publication date unknown).

III. To overcome the objections raised, a new set of claims was submitted by the Patentee consisting of 19 claims, wherein the only independent Claims 1 and 16 read as follows:

"1. A multiphase thermoplastic composition consisting essentially of at least 55 parts by weight of a polyamide matrix resin of number average molecular weight in the range of 5000 to 30.000 and up to 45 parts by weight of a multiphase core/shell polymer comprising 50 to 90 parts by weight of a crosslinked elastomer core and 10 to 50

parts by weight of a rigid thermoplastic polymer shell of glass transition temperature at least about 35°C comprising from 1 to 25 parts by weight of an interpolymerized ethylenically unsaturated carboxylic acid monomer, from 20 to 80 parts by weight of interpolymerized styrene, from 0 to 79 parts by weight of an interpolymerized C₁ to C₈ alkyl acrylate or methacrylate and from 0 to 45 parts by weight of interpolymerized acrylonitrile or methacrylonitrile based on 100 parts by weight of shell polymer, wherein the multiphase core/shell polymer has a core of weight average particle diameter of at least 0.3 μm and a rigid shell of average thickness of at least 0.025 μm, and wherein the amount of core/shell polymer is sufficient that said elastomer core is at least 10 percent by weight of said composition.

16. A process of preparing a toughened multiphase thermoplastic composition which comprises:

1. preparing an aqueous emulsion of an elastomeric core polymer of weight average particle size in the range of 0.3 to 0.8 μm,
2. graft polymerizing on the elastomeric core a rigid shell comprising from 1 to 25 parts by weight of an interpolymerized ethylenically unsaturated carboxylic acid monomer, from 20 to 80 parts by weight of interpolymerized styrene, from 0 to 79 parts by weight of a C₁ to C₈ alkyl acrylate or methacrylate and from 0 to 45 parts by weight of interpolymerized acrylonitrile or methacrylonitrile per 100 parts by weight of rigid shell, wherein the rigid shell has a glass transition temperature of at least 35°C and is of average thickness at least 0.025 μm and wherein the weight ratio of core to shell is in the range of 9:1 to 1:1,
3. recovering the multiphase core/shell polymer from the aqueous emulsion; and

4. melt blending the multiphase core/shell polymer with an amount of a polyamide of number average molecular weight in the range of 5000 to 30.000, such that the composition consists essentially of at least 55 parts by weight of the polyamide and up to 45 parts by weight of the multiphase core/shell polymer and the amount of said elastomer core is at least 10 percent by weight of the composition."

- IV. In an interlocutory decision issued 16 September 1988 the Opposition Division held that the claimed subject-matter on the basis of the aforementioned set of claims was new and inventive, for the following reasons.

As to novelty, none of the cited documents disclosed an amount of at least 10 percent by weight of the elastomer core in combination with the other features of Claim 1. As to inventive step, they held that the distinguishing features of the claim resulted in a reduction to zero of the brittle failure in the multiaxial driven dart test, in combination with a considerable improvement of the impact strength. To arrive at the claimed solution to this problem could not have been obvious in view of the cited prior art.

The request by the Appellant to adjourn the oral proceedings, so as to afford him an opportunity to file further evidence for lack of novelty having regard to the latex particle size in Example 17 of (1), was refused on the ground that such evidence would have been irrelevant to their decision.

- V. A notice of appeal together with payment of the prescribed fee was received on 15 November 1988. A statement of grounds of appeal was submitted on 11 January 1989.

In their written submissions and during oral proceedings held on 24 October 1990, the Appellant argued essentially as follows:

- (a) The claims contained subject-matter which extended beyond the content of the application as filed (Art. 123(2) EPC).

- (b) Disregarding the inadmissibility of the term "elastomer core" instead of "elastomer", the subject-matter of Claim 1 was not novel since it was disclosed in Example 17 of (1). Moreover, the provisions of Art. 56 EPC were not met since for a man skilled in the art it would have been obvious to improve the impact strength of thermoplastic polymers by the addition of elastomers in such amounts that a balance between different physical properties would be achieved.

The Appellant maintained his objection under Article 113 EPC formally made by him during the oral proceedings before the Opposition Division.

- VI. The Appellant requests that the decision under appeal be set aside and the patent be revoked. Moreover, he requests reimbursement of the appeal fee.

The Respondent requests that the appeal be dismissed.

- VII. The Respondent did not attend the oral proceedings, of which intention he had given advance notice. At the end of the oral proceedings the Chairman announced the Board's decision.

Reasons for the Decision

1. The appeal is admissible.
2. The admissibility of the amendments
 - 2.1 Claim 1 as granted is supported by original Claims 1 and 3.
 - 2.2 Claims 1, 2, 6, 12, 16, 17 and 18 in the set of claims under appeal differ from the corresponding claims of the originally filed ones and those of the patent as granted in that the term "about" preceding various numerical values has been deleted. Since the said values themselves were disclosed in the originally filed document and in the patent specification as granted, this amendment complies with the provisions of Art. 123(2) EPC.
 - 2.3 Moreover, Claim 1 differs from the originally filed and the granted Claim 1 in that, at its end, there is added the phrase "and wherein the amount of core/shell polymer is sufficient that said elastomer core is at least 10 percent by weight of said composition".

Support for this is found on page 11, lines 8 and 9 of the originally filed document (= page 5, lines 15 to 17 of the patent specification). While these passages do not contain appropriate literal disclosure (since "elastomer" and not "elastomer core" is mentioned therein), a man skilled in the art would not read the indicated lines in isolation, but rather in context with, at least, the complete paragraph bridging pages 10 and 11 of the originally filed document (corresponding to page 5, lines 10 to 18 of the granted patent specification) with the result that he would interpret "elastomer" in the sense of "elastomer core".

- 2.4 As a consequence of the afore-mentioned restriction, the 99% upper limit for the polyamide and the 1% lower limit for the core/shell polymer previously contained in Claim 1 have become redundant, so that the ranges "of 55 to 99" and "1 to 45" could be replaced by "at least 55" and "up to 45", respectively.
- 2.5 Process Claim 16 has merely been brought into line with product Claim 1 so that the above will equally apply.
- 2.6 The Board is, therefore, satisfied that the present claims meet the requirements of Art. 123(2) EPC.
- 2.7 Since the above discussed amendments do not extend the protection conferred by the claims of the patent in suit, they also comply with Art. 123(3) EPC.
3. The only documents in appeal proceedings dealing with thermoplastic polyamide compositions comprising as additive a multiphase core/shell polymer are (1) and (3). Document (2) concerns polyamides and some physical properties thereof. The late-submitted document (4) relates to the composition and properties of a polybutadiene trade product. In the absence of any proof that it actually is a pre-published document, it is disregarded as cited prior art under Art. 114(2) EPC. Its contents are, however, taken into consideration as evidence for the nature of the trade product FR-S 2004 (see below).

Following the finding of the Opposition Division, the Board accepts (1) as the closest prior art.

The most relevant part of document (1) is Example 17, which discloses a composition of nylon type C, i.e.

nylon 6, with a relative viscosity of 2.50 (see lower part of column 12), corresponding to a molecular weight of below 18 000 (cf. Table 7 on page 14 of (2)) and a core/shell polymer with a core/shell ratio of about 1/1 having a polybutadiene core (on the basis of the trade product FR-S 2004 (document (4)) and a shell comprising 400 parts by weight styrene and 100 parts by weight acrylic acid. The elastomer content disclosed in Example 17 is about 6.25%, and is thus lower than the minimum content of 10% as claimed in the patent in suit.

As far as the particle size is concerned, numerous arguments were produced to demonstrate that the polybutadiene latex of (4) has a particle size of at least 0.3 μm . These arguments clearly showed that particle sizes in the technical field concerned were either indicated on an average weight or on an average number basis, leading to different figures. The patent in suit specifies the particle size as calculated on a weight basis. Both documents (4) and (1) are totally silent on the basis on which the particle size has been calculated, see Example 17 and column 9, lines 13 to 18 of (1). The Board cannot, therefore, accept that Example 17 of (1) teaches the use of core/shell polymers wherein the particle diameter of the core corresponds to the core diameter as claimed in the patent in suit.

4. In the light of this prior art, the problem underlying the patent in suit may thus be seen in providing a uniform dispersion of a core/shell polymer in a polyamide having low molecular weight, leading to a considerable improvement of the impact strength in combination with a reduction of the brittle failure to zero despite of the low viscosity of the polyamide at the blending temperature.

5. According to the patent in suit, this problem is solved by a composition of a polyamide and a core/shell polymer with an elastomer core content of at least 10% by weight, a core with the weight average particle diameter of at least 0.3 μm , a rigid shell of an average thickness of at least 0.025 μm and comprising 50 to 90 parts by weight of core and 10 to 50 parts by weight of shell.

In view of the results given in Table 2 of the patent in suit, the Board is satisfied that the above defined technical problem is solved, since a comparison of Examples 10 to 13, the compositions of which differ in particle size (0.3 versus 0.13 μm), shows that the selection of a value of at least 0.3 μm is a factor that is responsible for high impact strength.

The influence of the core/shell ratio on impact strength is also shown in Examples 1 to 7, of which Examples 6 and 7 have a core/shell ratio of 1/1.4 and 1/1.8 instead of 9/1 to 1/1. That an elastomer core content of at least 10% by weight of composition leads to a brittle failure of zero is also demonstrated by Examples 4, 5, 9, 10 and 11.

6. Having examined the prior art, the Board is satisfied that the subject-matter of Claim 1 was not disclosed in any single document thereof, since an amount of at least 10% by weight of elastomer core in combination with the other features of the claimed compositions was nowhere specified. Since the issue of novelty was not contested during appeal proceedings, there is no need to give more detailed reasons for this finding.
7. It remains to decide whether the subject-matter of the patent in suit involves an inventive step with regard to the teaching of the cited documents.

From column 9, lines 12 to 18 of (1) a man skilled in the art would learn that "particle size is not of narrow significance" to that invention, but may "range from as low as about" 0.05 to about 0.3 μm or even more. Moreover, (1) is silent upon the influence of core/shell ratio, shell thickness and elastomer core content on the physical properties of such polyamide compositions. Thus, from (1) alone it would not be obvious how to solve the problem underlying the patent in suit.

The only other document in the proceedings dealing with polyamide compositions comprising a core/shell polymer is (3). On page 4, lines 13 to 34 thereof, it is taught that known acrylic core/shell impact modifiers, when blended with higher molecular weight nylon, lead to materials having improved impact strength but very poor flow; on the other hand, when blended with lower molecular weight nylon, they yield materials having good flow but no improvement in impact strength. To get the optimal combination of improved impact strength and high melt flow, (3) recommends to produce a blend of the core/shell modifier and a high molecular weight nylon and to mix such blend with a low molecular weight nylon. That teaching cannot provide any incentive to use a core/shell modifier having the characteristics specified in the patent in suit.

Hence, the Board is satisfied that it would not have been obvious for a person skilled in the art to arrive at the claimed solution of the existing problem. The said solution does, therefore, involve an inventive step.

Claim 16 relates to a process for preparing the new and inventive compositions of Claim 1 and is thus equally patentable within the meaning of Art. 52 EPC.

The subject-matter of the dependent claims are further embodiments of the subject-matter of Claims 1 and 16 and are, thus, likewise patentable.

8. Reimbursement of appeal fees

Rule 67 EPC states that the reimbursement of appeal fees shall be ordered where the Board of Appeal deems an appeal to be allowable, if such reimbursement is equitable by reason of a substantial procedural violation. In the present case, since the appeal is not allowable, the request for reimbursement must be rejected and the question whether or not there was a substantial procedural violation based on Art. 113 EPC as argued by the Appellant need not be decided.

Order

For these reasons, it is decided that:

1. The appeal is dismissed.
2. The case is remitted to the Opposition Division with the order to maintain the patent on the basis of the claims held allowable in the decision of 20 June 1988, and with the description amended to accord with those claims.

The Registrar:


E. Görgmaier

The Chairman:


F. Antony