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File Number: T 266/90 - 3.3.3

Application No.: 82 106 946.5

Publication No.: 0 072 480

Title of invention: A high impact polyamide composition

Classification: C08L 77/00

D E C I S I O N  
of 30 April 1991

Proprietor of the patent: Asahi Kasei Kogyo Kabushiki Kaisha

Opponent: Imperial Chemical Industries PLC

Headword:

EPC Article 56

Keyword: "Inventive step (no) - no inventive selection"

Headnote



Case Number : T 266/90 - 3.3.3

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.3  
of 30 April 1991

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**Decision under appeal :** Interlocutory decision of the Opposition Division  
of the European Patent Office dated  
29 November 1989, posted on 19 January 1990  
concerning maintenance of European Patent  
No. 0 072 480 in amended form.

**Composition of the Board :**

**Chairman :** F. Antony  
**Members :** C. Gérardin  
M. Aúz Castro

Summary of Facts and Submissions

I. The mention of the grant of the patent No. 72 480 in respect of European patent application No. 82 106 946.5 filed on 31 July 1982 and claiming priority of 4 August 1981 from an earlier application in Japan, was published on 22 January 1986 on the basis of seven claims.

Claim 1 read as follows:

"A high impact polyamide composition comprising a polyamide (A) and an ethylene ionomer resin (B) characterized in that it further comprises an elastomeric ethylene copolymer (C); that the weight proportion of the polyamide (A) relative to the total of (A), (B) and (C) is 50 to 95%, the weight proportion of the ethylene ionomer resin (B) relative to the total of (A), (B) and (C) is 1 to 45%, and the weight proportion of the elastomeric ethylene copolymer (C) relative to the total of (A), (B) and (C) is 0.5 to 40%; and that the ethylene ionomer resin (B) and the elastomeric ethylene copolymer (C) respectively have melt indexes ( $F_b$ ) and ( $F_c$ ) satisfying the inequalities:

$$\begin{aligned} 3 \geq F_b \geq 0.1 \\ 10 \geq F_c \geq 1 \\ 50 \geq F_c / F_b \geq 2 \end{aligned}$$

said melt indexes  $F_b$  and  $F_c$  each being given in terms of a value (g/10 min) as measured at a cylinder temperature of 190°C with a load of 2160 g in accordance with ASTM-D 1238."

Claims 2 to 7 were directed to preferred compositions according to the main claim.

II. On 1 October 1986 the Opponent filed a notice of opposition against the grant of the patent on the grounds that the requirements of Article 100(a) EPC, more specifically of Article 56 EPC, were not met. These objections, which were elaborated and emphasised in a later submission as well as during oral proceedings, were based essentially on the following document:

(3) GB-A-1 552 352.

III. By an interlocutory decision of 29 November 1989, with written reasons issued on 19 January 1990, the Opposition Division held that there were no grounds for opposition to the maintenance of the patent in amended form on the basis of a new set of five claims filed on 12 May 1987. In the new main claim it was specified that component (C) was "selected from the group consisting of a copolymer of ethylene and an alpha-olefin containing 3 to 8 carbon atoms and a polymer of ethylene, an alpha-olefin containing 3 to 8 carbon atoms and a diene monomer".

In that decision it was first stated that ternary compositions comprising components (A), (B) and (C) within the meaning of the patent in suit were not disclosed in document (3), so that novelty was acknowledged.

Examples 71 and 72 of that citation, wherein component (C) was respectively an ethylene/isobutyl acrylate copolymer and an ethylene/vinyl acetate copolymer, were regarded as the closest state of the art. In view of the experimental evidence submitted by the Patentee, showing that such ternary compositions were inferior to the claimed compositions in terms of heat resistance, mouldability,

surface appearance and Izod impact strength, the requirement of inventive step was also met.

- IV. The Opponent (Appellant) thereafter filed a notice of appeal against that decision on 23 March 1990 and paid the prescribed fee at the same time. In the statement of grounds of appeal filed on 16 May 1990 the Appellant argued that the incorporation of a rubber additive acting as a diluent was well known in the art to improve the low temperature impact performance of polyamide compositions; in this regard, the choice of an ethylene/propylene copolymer or of an EPDM rubber as component (C) was not inventive. Further, the alleged improvement in mouldability and impact strength had not been demonstrated; it followed that the claimed compositions could at most be regarded as equivalent to those known from document (3), which finding did not involve an inventive step.
- V. During oral proceedings held on 30 April 1991 emphasis was put more particularly on the composition according to Example 113 of document (3), which comprised a polyamide matrix, an ethylene/ethyl hydrogen maleate copolymer, and an ethylene/propylene copolymer, and appeared consequently as more relevant than the two compositions according to Examples 71 and 72 considered hitherto. This led the Patentee (Respondent) to file a new Claim 1 to be considered as auxiliary request, wherein it was additionally specified that the ethylene ionomer resin (B) comprised "ethylene monomer units, ethylenically unsaturated carboxylic acid ester monomer units, ethylenically unsaturated carboxylic acid monomer units and ethylenically unsaturated carboxylic acid metal salt monomer units;

the molar proportion of said ethylene monomer units relative to the total of the ethylene monomer units, ethylenically unsaturated carboxylic acid ester monomer units, ethylenically unsaturated carboxylic acid monomer units and ethylenically unsaturated carboxylic acid metal salt monomer units being 90 to 97%;

and wherein the number of moles of ethylenically unsaturated carboxylic acid ester monomer units ( $\alpha$ ), the number of moles of ethylenically unsaturated carboxylic acid monomer units ( $\beta$ ) and the number of moles of ethylenically unsaturated carboxylic acid metal salt monomer units ( $\gamma$ ) satisfy the inequalities:

$$0 \leq \frac{\alpha}{\alpha + \beta + \gamma} \leq 0.6$$

$$0.1 \leq \frac{\gamma}{\alpha + \beta + \gamma} \leq 0.7 \text{ "}$$

VI. In support of the patentability of the claimed subject-matter the Respondent argued mainly along the line of a selection invention. By contrast to the broad disclosure of document (3), the subject-matter of Claim 1 as amended during the opposition procedure as well as that of Claim 1 as submitted during oral proceedings should be regarded as attempts to optimise the balance of properties achieved in that prior art. For that purpose, the three components had to be present in specific amounts and the melt indexes of components (B) and (C) had to satisfy three inequalities; in Claim 1 submitted as auxiliary request the monomer units other than ethylene in component (B) additionally had to be within specific ranges satisfying two inequalities.

VII. The Appellant requested that the decision under appeal be set aside and the patent be revoked.

The Respondent requested that the appeal be dismissed or, alternatively, that the patent be maintained on the basis of Claim 1 submitted during oral proceedings and Claims 2 to 4 corresponding to Claims 3 to 5 of the main request.

#### Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.
2. The current wording of the claims does not give rise to any objections under Article 123 EPC.

Claim 1 according to the main request results from the combination of Claim 1 as granted and originally filed with Claims 4 and 5 as granted and originally filed. As far as dependent Claims 2 to 4 are concerned, they correspond to Claims 2, 3 and 6 as granted and originally filed; further, Claim 5 corresponds to Claim 7 as granted, which in turn derives from a combination of Claims 7 and 9 as originally filed, with their numbers and, where appropriate, appendancies adjusted.

Regarding the set of claims to be considered as auxiliary request, Claim 1 results from the incorporation of Claims 2, 4 and 5 as granted and originally filed into Claim 1 as granted and originally filed. Dependent Claims 2 to 4 correspond to Claims 3 to 5 according to the main request, with their numbers and, where appropriate, appendancies adjusted, and are thus allowable for the same reasons.

3. The patent in suit concerns a high impact polyamide composition. Such compositions are known from document (3), which the Board, like the Opposition Division, regards as the most relevant document. That citation deals with a toughened multi-phase thermoplastic composition defined in general terms as comprising 60 to 99 percent by weight of one phase containing a polyamide matrix resin, and 40 to 1 percent by weight of a least one other phase comprising particles of at least one polymer having sites which adhere to the polyamide matrix resin (Claims 1 and 3). Whereas the Opposition Division considered the ternary compositions mentioned in Examples 71 and 72, wherein the third polymer component is respectively an ethylene/isobutyl acrylate copolymer and an ethylene/vinyl acetate copolymer, as the closest state of the art, the Board finds it more appropriate to start from the ternary composition described in Example 113. This approach was also followed by the parties during oral proceedings. According to that example (page 15, Table 1-B; page 32, Table 3), a polymer referred to as polymer 64 is incorporated into a polyamide matrix; that product is defined as a blend of 64 grams of an ethylene/ethyl hydrogen maleate random copolymer containing 90 weight percent ethylene monomer units and 10 weight percent ethyl hydrogen maleate monomer units, and 256 grams of an elastomeric ethylene/propylene copolymer (page 20, lines 79 to 109). Although it is emphasised throughout that disclosure that such thermoplastic compositions exhibit generally a desirable combination of properties, the specific balance of mouldability and toughness properties cannot be regarded as optimal.

In the light of this prior art shortcoming, the technical problem underlying the patent in suit can thus be seen in providing a polyamide composition having an optimised balance of mouldability and toughness properties.

According to the patent in suit this problem is solved in the main request by (i) neutralizing part of the carboxylic groups of the random copolymer, and (ii) defining specific conditions which the melt indexes of the random copolymer and the elastomer have to fulfil, as well as additionally in the case of the auxiliary request (iii) defining quantitative requirements between the number of moles of ester, acid and metal salt in the ionomer resin.

Although no evidence has been provided that the combination of features (i) and (ii) in the case of the main request, or of features (i), (ii) and (iii) in the case of the auxiliary request, represents an effective solution to the above defined technical problem, the Board has no reason to cast doubts on the claimed improvement. Nor has the Appellant expressed any reserve in this respect.

4. Although it may be debatable whether the claimed subject-matter according to either of the two requests is even novel with regard to the teaching of Example 113 interpreted in the light of the whole content approach, the Board did not deem it appropriate to raise that issue for the first time during oral proceedings, all the more so as the Appellant explicitly acknowledged novelty.
5. It still remains to be examined whether the claimed subject-matter involves an inventive step with regard to the whole teaching of document (3).
- 5.1 Feature (i) reflects the fact that part of the carboxylic groups of the random copolymer has been neutralised, i.e. that component (B) is an ionomer resin.

The list contained in document (3) of polymers suitable for toughening polyamide compositions encompasses many ethylene random copolymers as well as metal salts thereof, and further mixtures of such compounds (page 4, line 79 to page 5, line 44). From the subsequent experimental data in that citation it appears that neutralisation has a major influence on properties like melt flow and toughness, and that consequently the skilled man knows how to control these parameters. Here specifically, the influence of neutralisation on melt flow can be seen in Table 1-B (pages 9 to 17), wherein the values of that parameter before and after neutralisation are given; in all the cases where the two values are indicated, one observes a marked decrease in melt flow after neutralisation.

Further, the influence of the degree of neutralisation on toughness can be seen in Examples 36 to 41 (pages 26/27, Table 3), wherein the same copolymer of ethylene/methyl acrylate/monoethyl ester of maleic anhydride in the proportions 42/54/4 has been neutralised at 0, 10, 25, 50, 75 and 100%; it clearly appears that the Notched Izod values increase with neutralisation. Similarly, the comparison between Examples 13 and 14 on page 25, Table 3, reveals the same trend caused by the presence of the ionomer group in the former composition.

These results show that the mere neutralisation of the random copolymer both lowers the melt flow value, whereby mouldability is improved, and increases toughness; it represents thus an obvious solution to the above-defined technical problem.

- 5.2 As noted above, the polyamide composition according to Example 113 of document (3) comprises an ethylene/ethyl hydrogen maleate random copolymer and an ethylene/propylene elastomer, the latter corresponding to the first alternative for component (C) in Claim 1 of the patent in

suit. The same combination of random copolymer and elastomer is also mentioned on page 5, lines 33 to 35. Similar combinations comprising such a random copolymer and an ethylene/propylene/1,4-hexadiene elastomer, containing thus a ternary elastomer corresponding to the second alternative for component (C) in the patent in suit, are described as equally suitable (page 5, lines 13 to 17; page 23, lines 21 to 27). Emphasis is put particularly on the wide toughening capability of such ternary elastomers, whose molecular weight as well as ratios of ethylene to propylene and diene content may vary within broad ranges (page 22, lines 120 to 127).

Such ternary elastomers represent thus an obvious alternative to the binary elastomer mentioned in Example 113; unexpected advantages resulting from their use in combination with an ionomer component as toughening agents of polyamides have not been put forward by the Respondent. It follows that this compositional feature cannot contribute to the inventiveness of the solution claimed in the patent in suit.

- 5.3 As far as feature (ii) is concerned, document (3) specifies that polymers liable to be combined with polyamides should generally have a melt flow range of more than 0.1 to 1000 g/10 min, preferably 0.5 to 100 g/10 min (page 5, lines 64 to 71). In the case of ethylene/propylene/ diene elastomers the melt flow range would be suitably 0.5 to 400 g/10 min. Regarding the melt flow of random copolymers, it appears from the experimental data in Table 1-B that most values indicated on pages 9 to 12 fall within the range required for (F<sub>b</sub>) in Claim 1 of the patent in suit.

During oral proceedings the Respondent underlined the contrast between the broad ranges of melt flow for both

the random copolymer and the elastomer in the prior art document, and the restrictive conditions imposed on that parameter in the patent in suit, and argued accordingly along the line of a selection invention. In the Board's view, however, although it may be true that the inequalities which the melt indexes ( $F_b$ ) and ( $F_c$ ) have to satisfy in Claim 1 of the patent in suit correspond to relatively narrow ranges, it is essential to appreciate that these ranges are in fact in the middle of the ranges defined as appropriate in the prior art; as such, they correspond to the most obvious values which the skilled man would start from in order to embark on routine investigations intended to optimise the balance of properties achieved in that prior art. For this reason, and in the absence of any surprising effect resulting from that selection, the latter cannot be regarded as inventive.

- 5.4 Similar considerations apply to feature (iii), i.e. to the quantitative requirements which the units derived from the unsaturated ester, acid and metal salt in the ionomer component (B) have to fulfil additionally in Claim 1 according to the auxiliary request.

These inequalities mean that with respect to the comonomers other than ethylene in the ionomer resin (B) the number of moles of ethylenically unsaturated carboxylic acid ester units ( $\alpha$ ) varies between 0 and 60% and the number of moles of ethylenically unsaturated carboxylic acid metal salt units ( $\gamma$ ) between 10 and 70%. As noted by the Board during oral proceedings, such ranges can hardly be regarded as selections. Moreover, in view, of the numerous experimental data reporting neutralisations carried out over practically the whole range between 0 and 100%, it is clear that the additional inequalities defining the limits for ( $\gamma$ ) refer to a

feature which has been intensively studied in the prior art and whose effects on the final properties of the polyamide composition are consequently well known by the skilled man. The same applies to the additional inequalities defining the limits for  $(\alpha)$ , since, on the one hand, the several copolymers quoted which do not contain ester units at all, such as the ethylene/maleic anhydride copolymers, are evidence that values as low as 0 are in fact common, and, on the other hand, the copolymer actually used in Example 113 corresponds to a value of 0.5. In this respect, although the Respondent was expressly invited by the Board to discuss the properties of the polyamide compositions in connection with the combination of features (i), (ii) and (iii), he has not put forward any surprising effect in favour of the inventiveness of that combination.

- 5.5 In conclusion, for the reasons given above, the subject-matter of Claim 1 according to the main request and the auxiliary request does not involve an inventive step.
6. Claim 1 not being allowable, the same applies to dependent Claims 2 to 5 according to the main request as well as to dependent Claims 2 to 4 according to the auxiliary request, which represent preferred embodiments of the subject-matter of the main claim and thus fall with it.

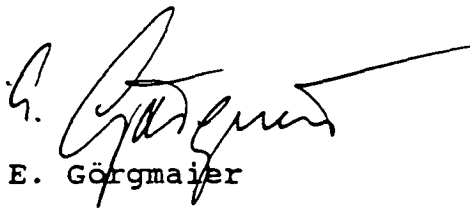
Order

For these reasons, it is decided that:

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

The Registrar:

The Chairman

  
E. Görgmaier

  
F. Antony