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File Number: T 385/89 - 3.3.3

Application No.: 81 302 723.2

Publication No.: 0 044 141

Title of invention: Filled compositions of thermoplastic polyamide and polyester

Classification: C08L 77/00

D E C I S I O N  
of 7 April 1992

Proprietor of the patent: IMPERIAL CHEMICAL INDUSTRIES

Opponent: BASF Aktiengesellschaft

Headword:

EPC Article 56

Keyword: "Inventive step (denied) - promising prior art teaching pointing to the solution claimed"

Headnote



Case Number : T 385/89 - 3.3.3

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.3  
of 7 April 1992

**Appellant :**  
(Proprietor of the patent)

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**Respondent :**  
(Opponent)

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**Decision under appeal :**

Decision of Opposition Division of the European  
Patent Office dated 9 December 1988, issued on  
18 April 1989 revoking European patent  
No. 0 044 141 pursuant to Article 102(1) EPC.

**Composition of the Board :**

**Chairman :** C. Gérardin  
**Members :** S. Schoedel  
M. Aúz Castro

**Summary of Facts and Submissions**

I. The mention of the grant of the patent No. 44 141 in respect of European patent application No. 81 302 723.2 filed on 17 June 1981 and claiming priority of 14 July 1980 of an earlier application in the United Kingdom, was published on 17 April 1985 on the basis of 9 claims.

Claim 1 reads as follows:

"An extruded profile section characterised in that it has been extruded from a composition comprising a blend of a linear polyamide, a linear polyester containing at least 80% by weight of ethylene terephthalate units and an inorganic fibrous filler wherein the composition contains 5 to 60% by weight of the composition of the filler, the weight ratio of polyamide to polyester is between 1:1 and 49:1 and the polyamide contains an excess of amine end groups over carboxyl end groups".

Claim 6 is another independent claim which is directed to a method of extruding a profile section characterised in that a composition as defined above is intimately blended and extruded through a die of profiled section.

Claim 8 was a further independent claim which, before the later amendment mentioned hereinbelow, concerned the thermoplastic composition suitable for extrusion into extruded profiled sections comprising a blend as defined above.

As to Claims 2 to 5, 7 and 9, they are dependent claims dealing with preferred embodiments of the subject-matter claimed in the three independent claims.

II. On 11 January 1986 the Opponent filed a Notice of Opposition against the grant of the patent on the grounds of insufficient disclosure within the meaning of Article 100(b) EPC as well as lack of inventive step (Article 100(a) EPC). These objections were based on a single document.

In support of its observations submitted on 31 March 1987 the Patentee referred to inter alia US-A-3 676 400 (document (2)).

In a communication dated 6 October 1987 the Opposition Division additionally introduced FR-A-2 074 500 (document (6)), already taken into consideration during the examining procedure, which it regarded as closer state of the art than the citation relied upon by the Opponent.

During oral proceedings before the Opposition Division on 9 December 1988 the Patentee amended claim 8 by specifying that "the melt flow index of the composition is not greater than 2 measured according to ASTM-D1238 using a standard die of bor 2.096 mm, length 8 mm and a load of 2.16 kg at a temperature of 285°C".

III. By a decision delivered orally on 9 December 1988, with written reasons posted on 18 April 1989, the Opposition Division revoked the patent on the ground that the thermoplastic composition according to Claim 8 as amended did not involve an inventive step. More specifically, it was stated in that decision that the only difference between that subject-matter and the composition known from document (6) was the excess of amine end groups over the carboxyl end groups in the polyamide component. The other difference, namely the requirement regarding the upper limit of melt flow index, was in fact the result to be

achieved, i.e. the technical problem underlying the patent in suit; as such, it was not an independent feature, but the direct consequence of cross-linking reactions between polyamide and polyester. The use of reactive polyamide, i.e. polyamide having the appropriate functionality, in order to lower the melt flow index or increase the molecular weight was self-evident for the skilled man, since document (2) solved an identical problem in an analogous manner.

IV. The Appellant (Patentee), thereafter, lodged a Notice of Appeal on 12 June 1989 and paid the prescribed fee at the same time. Together with the Statement of Grounds of Appeal received on 28 August 1989 the Appellant filed, as main request, the set of 9 claims already submitted on 9 December 1988 and, as auxiliary request, a set of 7 claims corresponding to Claims 1 to 7 as granted. During oral proceedings, which were held on 7 April 1992, the set of claims to be considered as main request was modified, Claim 8 being amended as to read "... and wherein the polyamide contains a sufficient excess of terminal amino groups over the carboxyl groups to provide a composition with a melt flow index not greater ...".

The arguments presented by the Appellant concerned in the first place the problem underlying the patent in suit. That problem was to provide a composition enabling extrudates to be produced with a reduced risk of profile sagging. The teaching of document (2) could not provide an incentive for the skilled man to incorporate amine end groups into the polyamide disclosed in document (6) for two reasons. First, the other polymer component according to document (2) was an addition copolymer containing carboxyl groups extensively distributed along the main chain; the extent of these carboxyl groups was so great that under melt conditions the likelihood of reaction with

an amino group of the polyamide was high, and thereby the compatibility between the two polymers correspondingly increased. By contrast, in the present case, the polyamide and the polyester being basically incompatible, the opportunity for reaction was correspondingly low. The skilled man had thus no reason to consider cross-linking reactions as a means of lowering the melt flow index. Secondly, as demonstrated during the opposition procedure, an adequate melt flow index could only be obtained when relatively large amounts of glass fibers were used, i.e. in the framework of ternary compositions which were not at all envisaged in document (2). For these reasons, the mere knowledge of the chemistry of carbonamide formation and that the presence of fillers in a molten composition increased the melt viscosity did not provide the information necessary to enable the skilled man to solve the above-defined problem.

- V. In response to these arguments the Respondent (Opponent) relied basically on the arguments presented during the opposition procedure. A mixture of a polyester and a polyamide with amine end groups in excess would self-evidently have a lower melt flow index than the corresponding mixture comprising a polyamide with equivalent end groups. Equally self-evident was the addition of glass fibers in order to increase the viscosity. It followed that the modifications of the compositions disclosed in document (6) in the manner claimed in the patent in suit did not involve an inventive step.
- VI. The Appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of Claims 1 to 9 filed during oral proceedings as main request or, alternatively, on the basis of Claims 1 to 7 filed by letter of 18 August 1989 as auxiliary request.

The Respondent requested that the appeal be dismissed.

**Reasons for the Decision**

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is admissible.

Main request

2. The current wording of the claims does not give rise to any objections under Article 123 EPC.

Claim 1 is identical with Claim 1 as granted, which itself is the combination of Claims 1 and 4 as originally filed.

Claim 8 appears basically as the combination of Claim 8 as granted, which itself corresponds to original Claim 9, with Claim 3 as granted and originally filed; the further amendment is nothing more than a rewording of the connection between the functionality of the polyamide and the melt flow index of the composition, now expressed in terms of a functional definition of that composition, which has no impact on the scope of protection.

As to the dependent Claims 2 to 7 and 9, they correspond to Claims 2 to 7 and 9 as granted, which themselves correspond respectively to Claims 2, 3, 5 to 8 and 10 as originally filed, with their numbers and, where appropriate, appendancies adjusted.

3. The patent in suit concerns filled compositions of thermoplastic polyamide and polyester. Such compositions are disclosed in document (6) which the Board, like the

Opposition Division, regards as the closest state of the art. That citation describes ternary compositions containing a linear polyamide, a linear polyester, such as polyethylene terephthalate, and glass fibers (Claims 1 and 2). In those blends the weight ratio of polyamide to polyester is between 90:10 and 50:50 and the weight ratio of glass fibers to polymer components is between 40:60 and 10:90 (page 2, lines 16 to 27; Examples 2 and 3). Polymers obtained by standard method as well as scrap polymers, especially when economic factors come into consideration, are said to be equally suitable (page 1, lines 1 to 7; page 3, lines 4 to 15); nylon 6, which normally contains an equivalent amount of amine and carboxylic end groups, is advantageously chosen as polyamide (page 2, lines 33 to 39). The objects prepared by transformation of these mixtures exhibit in general outstanding mechanical properties, in particular optimal dimensional stability properties, which is attributed to a marked synergistic effect (page 6, lines 10 to 19).

4. One of the main points raised by the Respondent during oral proceedings is that the experimental data in the patent in suit are not conclusive regarding a particular suitability of the claimed compositions for extrusion into profiles, as alleged by the Appellant. In Example 1 several ternary compositions based on different polyamides - nylon 66, nylon 6, mixture thereof in the weight ratio 90:10, and nylon 66 containing an excess of 30  $\mu$  equivalents/g of amine over carboxyl end groups - have been prepared by compounding in a single extruder at a nominal melt temperature of 285°C (Table 1 in conjunction with footnote b). Whereas the composition B to J can be regarded as corresponding to the blends disclosed in document (6), the composition K contains a polyamide having an excess of amine end groups as defined above, thus within the terms of the patent in suit. All the



compositions B to K are said to be suitable for producing profiled extrusions of good dimensional tolerance (page 4, lines 29/30). Further, the profiles from the compositions D, F and K, which are suitable as thermal barriers in double-glazed aluminium framed windows, have particularly good dimensional tolerance in that the dimensions match the profile of the die more closely (page 4, lines 35 to 38). This means that the presence of an excess of amine over carboxyl end groups in the polyamide component does not result in unexpected advantages for the application envisaged by the Appellant.

What the experimental results in Tables 1 and 2 bring to light, however, is that the melt flow index of the composition is greatly influenced by the presence of amine end groups in the polyamide. More specifically, the comparison of the values of the melt flow index of the compositions D and K in Table 1, which only differ by the presence of an excess of amine end groups in the polyamide of the latter composition, and, further, the analysis of the data in Table 2, which illustrate the variation of the melt flow index according to the excess of amine over carboxyl end groups in the polyamide, show that this parameter decreases when the excess of amine end groups increases.

In the light of this technical effect, the problem underlying the patent in suit can thus be seen in providing further ternary compositions suitable for extrusion into profiles, which have reduced melt flow index values.

According to Claim 8 of the main request, this problem is solved by using a polyamide component which contains an excess of amine end groups over carboxyl end groups.

In view of the experimental data referred to above, the Board is satisfied that the above-defined technical problem is effectively solved. These results have not been disputed by the Respondent.

5. After examination of the cited documents, the Board has reached the conclusion that the solution as claimed by the Appellant is not disclosed in any of them and that the subject-matter of the independent composition Claim 8 is, therefore, novel. Since the Respondent has not raised the issue of novelty, it is not necessary to consider this matter in detail.
  
6. It still remains to be decided whether that subject-matter involves an inventive step with regard to the cited documents.
  
- 6.1 The introductory section of document (6) mentions various mechanical properties which are positively influenced by the incorporation of fillers into polymer compositions, in particular glass fillers into linear polyamides and linear polyesters. The high viscosity of the polymers is said to be a further parameter which has a beneficial effect on these properties, especially on dimensional stability (page 1, lines 1 to 23). Although this might suggest increasing the molecular weight of the polyamide component in order to improve the dimensional stability of the composition, in practice this would not be a very attractive solution, for it would involve an additional post-polycondensation step in the solid phase.

Document (2) discloses a much less cumbersome method for increasing the melt viscosity of such compositions. That citation describes thermoplastic blends of 50 to 90% by weight of a polyamide having from 20 to 140 gm equivalents of amine end groups in excess of the number of gm

equivalents of carboxyl end groups per  $10^6$  gm of polyamide, and, complementally, from 10 to 50% by weight of an acid-containing olefin copolymer containing from 0.1 to 10 mole percent of acid groups derived from an  $\alpha, \beta$ -unsaturated carboxylic acid (column 1, lines 10 to 17). These blends, wherein the olefin copolymer is generally present as a dispersed phase in a continuous phase of polyamide, exhibit properties which are neither additive, nor predictable from the properties of the individual polymer components; this applies in particular to the melt viscosity which is generally greater than either of the components (column 1, lines 21 to 25). Further, these blends exhibit improved mechanical properties, especially surprisingly high flex modulus, elongation and toughness, which make them suitable for the fabrication by extrusion of a wide range of articles (column 1, lines 29 to 34; column 2, line 67 to column 3, line 6). These advantageous properties are explicitly attributed to the presence of terminal amine groups in the polyamide component, whereby reactivity with the carboxyl groups and cross-linking with the acid-containing olefin copolymer are ensured (column 1, line 65 to column 2, line 3; column 2, lines 59 to 66; Tables I, II and IV to VIII). This is particularly interesting in the case of polyamides, whose molecular weight would be insufficient to develop optimal properties for moulding applications, but which are hereby converted to extremely valuable materials (column 3, lines 14 to 20).

- 6.2 It may be true, as argued by the Appellant in the Statement of Grounds of Appeal (page 1, paragraphs 5 and 6), that the olefin copolymers envisaged in document (2), which are addition copolymers of olefins and monoethylenically  $\alpha, \beta$ -unsaturated carboxylic acids containing carboxyl groups extensively distributed along the copolymer chain, are quite different from the linear

polyesters considered in document (6), wherein the carboxyl groups may only be present at the end(s) of the polymer chain. As noted above, however, the teaching of document (2) does not boil down to the sole definition of thermoplastic blends, including thus the structure of the copolymer containing carboxyl groups, but extends to the influence of the amine end groups in the polyamide component on the properties of these reactive blends, especially on their melt viscosity and mechanical properties. In the Board's view, such a teaching looks so promising that it would represent an incentive for the skilled man to adopt a solution along the same line in order to obtain similar advantages.

The determination of the appropriate amount of amine end groups in the polyamide component would not involve more than routine experiments based on trial and error. For that purpose, two factors which have the opposite effect would have to be taken into account. The first one is the incompatibility of the polyamide and the polyester which causes the minor constituent (the polyester) to be present as relatively large droplets within a continuous melt of the major constituent (the polyamide), whereby the opportunity for reaction between corresponding groups is reduced (see Statement of Grounds of Appeal, passage bridging pages 1 and 2). The second one is the synergistic effect due to the additional presence of glass fibers in the ternary compositions to be improved, whereby the increase of melt viscosity as the result of the reaction between the two polymer components will be enhanced (see document (6), page 6, lines 10 to 19).

According to document (2), the preferred number of amine end groups is from 25 to 60 gm equivalents in excess of the number of carboxyl end groups per  $10^6$  gm of polyamide (column 1, line 75 to column , line 2). It is significant

that this range, which is given without any reference to the quantitative and qualitative features of the composition, corresponds practically to the amount mentioned in the patent in suit; it is specified there that, in order to obtain a composition having a melt flow index of at most 2, the polyamide should contain at least 20 g equivalents of terminal amine groups per  $10^6$  g of polyamide in excess of the carboxyl groups (see Claim 4 as well as page 2, line 65 to page 3, line 2).

It follows that the determination of the excess of amine end groups in the polyamide component corresponding to a melt flow index of the composition not greater than 2 is not inventive. This means as well that this upper limit does not represent an inventive feature.

- 6.3 For these reasons, the subject-matter of Claim 8 of the main request does not involve an inventive step.
7. In the absence of a separate request directed to the specific features mentioned in the other claims, the latter must fall with Claim 8, since a request can only be considered as a whole. Besides, no argument in favour of the inventiveness of any of these features has been provided by the Appellant.

Auxiliary request

8. No objection arises having regard to Article 123 EPC, since the seven claims are indetical with Claims 1 to 7 of the main request and are thus formally admissible for the reasons given above in point 2.
9. As stated above, document (2) teaches that reactive compositions based on a polyamide having an excess of amine end groups over carboxyl end groups, as major

component, and on an addition copolymer containing carboxyl groups distributed along the main chain, as minor component, are suitable for the fabrication of numerous articles by extrusion. The same suitability can be expected from the compositions according to the patent in suit, whose general properties, in particular melt viscosity and mechanical properties, are based on the same concept, i.e. the reactivity between the two polymer components. Therefore, in the absence of any argument or evidence showing that the extruded profile section according to Claim 1 of the auxiliary request has improved properties, no inventive step can be acknowledged for that subject-matter.

10. In the absence of a separate request directed to the specific features mentioned in the other claims, the latter must fall with Claim 1, since a request can only be considered as a whole.
  
11. Even if, for the sake of argument, an improvement of one or several mechanical properties had been demonstrated by the Appellant as the result of lower melt flow index values, this effect would not be regarded as involving an inventive step.

As noted above, the prior art teaches the beneficial effect of using polyamides of higher molecular weight, thus of higher viscosity or lower melt flow index, on the mechanical properties of the composition, whether this is achieved by an additional post-polycondensation step, as suggested in document (6) in general terms, or by interaction with the other polymer component, as illustrated in document (2). Improved mechanical properties would thus only meet the expectations of the skilled man. This means that even a more ambitious definition of the problem underlying the patent in suit,

based on an objective demonstration of improved mechanical properties, would not have led to a different conclusion regarding the issue of inventive step.

Order

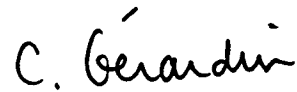
For these reasons, it is decided that:

The appeal is dismissed.

The Registrar:

  
E. Gergmaier

The Chairman:

  
C. Gérardin