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Bezeichnung der Erfindung: Production of polyamide-based objects
Title of invention:
Titre de l'invention :

Klassifikation / Classification / Classement : C08G 69/26

ENTSCHEIDUNG / DECISION

vom / of / du 28 February 1990

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent / Titulaire du brevet : Stamicarbon B.V.

Einsprechender / Opponent / Opposant : BASF Aktiengesellschaft

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Articles 56, 114

Schlagwort / Keyword / Mot clé : "Inventive step (confirmed) - long-felt need"

Leitsatz / Headnote / Sommaire

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Case Number : T 226/89 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 28 February 1990

Appellant : (Opponent)
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Representative :

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Decision under appeal :
Decision of the Opposition Division of the European Patent Office dated 30 January 1989 rejecting the opposition filed against European patent No. 0 038 582 pursuant to Article 102(2) EPC.

Composition of the Board :

Chairman : K. Jahn
Members : C. Gérardin
J. Stephens-Ofner

Summary of Facts and Submissions

I. The mention of the grant of the patent no. 38 582 in respect of European patent application no. 81 200 325.9 filed on 25 March 1981 and claiming priority of 26 March 1980 of an earlier application in The Netherlands, was published on 28 August 1985 on the basis of 6 claims.

Claim 1 reads as follows:

"Use of a melt of a polyamide having a molecular chain of units of

$$[NH - (CH_2)_4 - NH - CO - (CH_2)_4 - CO]$$

optionally containing up to 20 wt-% related to the total of units derived from other polyamide forming compounds, which polyamide has a relative viscosity of at least 2.0 measured at 20°C, on a 1% by weight solution in 96% sulfuric acid and which polyamide has been produced by preparing a prepolymer in the presence of an excess of 1,4 diaminobutane of 0.5-15 mole %, and after condensing the solid prepolymer in the presence of an atmosphere containing water vapour, for preparing objects by extrusion or injection moulding".

Whereas Claims 2 to 4 are directed to preferred embodiments of this use claim, the independent Claims 5 and 6 are concerned with respectively a process for producing an object by extrusion or injection moulding and the resuling injection moulded or extruded object, both claims reciting the features of the polyamide 4,6 and of the two-step preparation thereof, as specified in Claim 1.

II. On 27 May 1986 the Appellant (Opponent) filed a notice of opposition requesting the revocation of the whole patent on the ground that the subject-matter of the patent in suit did

not involve an inventive step with regard to the teaching of documents (1) to (4), especially

(1) Journal of Polymer Science, Polymer Chemistry Edition, volume 15, pages 537 to 545 (1977).

Together with the subsequent statement of 16 June 1987 the Appellant filed three additional documents (5) to (7) in support of the same objection.

III. By decision of 30 January 1989 the Opposition Division rejected the opposition on the ground that the requirement of inventive step was met. In this decision it was first stated that documents (5) to (7) had been considered not relevant for the decision and in view of their late submission they had been disregarded in accordance with the terms of Article 114(2) EPC.

As to documents (1) to (4) it was specified that neither in isolation nor in combination they could have led the skilled man to use a melt of a polyamide 4,6 prepared by a two step condensation process in order to produce an object by extrusion or injection moulding with improved notched impact resistance and higher heat distortion temperature.

IV. The Appellant thereafter lodged a notice of appeal on 28 March 1989 and paid the prescribed fee at the same time.

In the Statement of grounds of appeal filed simultaneously as well as in the later submission filed on 4 October 1989 the Appellant did not diretly challenge the reasoning set out in the decision of the Opposition Division rejecting the opposition, but relied exclusively upon one of the late filed documents, namely

(5) ACS Symposium Series 104 (1979), pages 137 to 148.

The main objection was that the teaching of this document had been misinterpreted by the Opposition Division and that by disregarding it the first instance had abused its power

under Article 114(2) EPC. More specifically, the Appellant put forward that document (5), through its reference to document (1) among the literature cited at the end of the article, actually described the same two step preparation of polyamide 4,6 and mentioned the same good color properties thereof as the patent in suit; he then concluded that the mere use of such polymer in extrusion or injection moulding processes could not be regarded as inventive as such, since this specific application was common in the field of polyamides.

V. In his counter-statement filed on 18 August 1989 the Respondent basically argued that the subject-matter of document (5) was the preparation of polyamide 4,6 whereas the patent in suit was directed to a specific use thereof and that consequently the essential criterion of patentability should be the inventiveness of the use of polyamide 4,6 for injection moulding or extrusion. Since a polymer with these recurrent units having good mechanical properties could not be obtained by prior art methods, the improved impact resistance of the moulding objects which the specific polyamide 4,6 affords must be regarded as inventive.

VI. The Appellant requested that the decision under appeal be set aside and that the patent be revoked. 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, therefore, admissible.

2. The whole argumentation presented by the Appellant is based on the assumption that document (5) describes the same two-step process of preparation of polyamide 4,6 and

consequently the same polymer as the polymer used according to the patent in suit. In fact, there is a major difference in the first step, and the operative features of the second step chosen by the Respondent do not follow the prior art teaching.

2.1 Document (5) deals with the polyamidation in the solid phase process of polyamide 4,6; in this article various factors which influence the polymerization rate are reviewed and some structural features of the final polymer, especially the degradation with color formation, are examined (page 137, paragraphs 2 and 3; page 139, paragraph 2).

As starting materials for the polyamidation or polycondensation reactions are mentioned low molecular weight polymers prepared by reacting 1,4-diaminobutane and adipic acid for two hours at 220°C in a capsule in an autoclave (page 139, paragraph 3). This passage is followed by a reference to a citation identified at the end of the article as the present document (1).

In document (1) which likewise is concerned with the two-step preparation of polyamide 4,6, it is specified that the first step consists in the preparation of a colorless low molecular weight salt with an inherent viscosity of 0.18 dl/g measured in 0.5% solution in 98% formic acid, the reaction being carried out with an excess of 1,4-diaminobutane (page 537, Synopis, lines 1 to 5; page 538, Polymerization, lines 1 to 5 and Characterization, lines 1 to 4; page 539, Polymerization, lines 1 to 3). This excess is necessary not only because this compound is more volatile than adipic acid and can cyclize to pyrrolidine, but as well in order to obtain a prepolymer of higher pH which insures a higher molecular weight of polyamide 4,6 in the second step (page 538, paragraph 1; page 539, last line to page 540, whole paragraph in combination with Tables I and II and Figure 1).

The data relative to the inherent viscosity available in document (5) confirm that the molecular weight of the prepolymers A and B disclosed therein corresponds to the molecular weight of the product after the first step according to document (1). Although the values of the inherent viscosity have been determined according to document (5) in 0.5% solutions in 90% formic acid and according to document (1) in 0.5% solutions in 98% formic acid, it is reasonable to assume that this difference in concentration alone does not affect the value of this parameter significantly and that consequently these prepolymers are at least very similar as far as their molecular weight is concerned.

The endgroup analysis of these prior art prepolymers brings to light that the latter have been prepared by using relative amounts of 1,4 - diaminobutane to adipic acid much higher than according to the requirements of Claim 1 of the patent in suit. Table I of document (5) specifies that prepolymer A exhibits 43.5 carboxylic groups and 58.9 amine groups and prepolymer B 48.2 carboxylic groups and 57.8 amine groups, all these figures being expressed in eq/10^5g, which represents an excess of amine endgroups of respectively 35.4 and 19.9 mole %; since in the reaction conditions 1,4 - diaminobutane is liable to volatilize as well as give rise to pyrrolidine rings by cyclization, it is self evident that the initial excess of diamine must have been even higher than these figures, thus clearly outside the range of 0.5 to 15 mole % claimed by the Respondent.

In conclusion, the results of endgroup analysis provide evidence that the prepolymers described in documents (1) and (5) do not correspond to those envisaged in the patent in suit.

2.2 The option made by the Respondent in the second step, i.e. the polycondensation or polyamidation reaction, does not follow the prior art teaching either.

According to document (5) the prepolymer is subjected to polycondensation in a fluidization bed reactor, the carrier gas being dry nitrogen or super-heated steam, both at a pressure of 1 bar, or in a rotary vacuum dyer (page 139, Experimental, paragraph 2; page 146, Type of reactor gas and Table IV). The alternative super-heated steam, which corresponds to the physical conditions required in Claim 1 of the patent in suit, has the advantage of suppressing the side reactions which lead to colored products which occur with dry nitrogen or in the rotating flask under vacuum, but at the same time reduces the overall polycondensation rates. Absence of coloration and high molecular weight of polyamide 4,6 appear thus as antagonistic features, which means that one property can only be achieved at the expense of the other. In practice, as it appears from the Table IV in combination with Table I, the molecular weight of 15700 which is obtained when polyamidation is carried out with super-heated steam during 4 hours at 250°C corresponds to an inherent viscosity which hardly exceeds 1 dl/g; such low value clearly does not correspond to the high molecular weights required in the patent in suit and would hardly be compatible with the requirements of extrusion and injection moulding. The mere reference to non colored products when polycondensation is carried out in presence of steam cannot thus be interpreted as the availability of high molecular weight polyamide 4,6, as the Appellant put forward in statement of grounds of appeal (point 1, last two paragraphs).

Even the general data provided by viscosimetry in Table I, wherein the features of the second step as well as the coloration of the polymer have been left out of account, do not afford another conclusion. They show that the prior art range for inherent viscosity does not extend much beyond 2.7 dl/g. Although this may not be exactly the value obtainable with another method of determination of this parameter, such as in the patent in suit where the measurement occurs on a 1% solution in 96% sulfuric acid, it nevertheless reflects an order of magnitude which is modest in comparison with the values actually envisaged by the Respondent for objects which have to satisfy high requirements (compare patent in suit, column 2, lines 9 to 18).

Furthermore, the fact that water vapor results in opposite effects in document (5), where it reduces the molecular weight, and in the patent in suit, where it affords very high molecular weights, confirms the conclusion of paragraph 2. 1 that the prepolymer starting material itself, i.e. the reaction product of the first step, is different.

2.3 For these various reasons, the prior art polymers cannot be equated with the specific polyamide 4,6 used in the patent in suit.

3. Extrusion and injection moulding application not being mentioned in the prior art cited, the subject-matter of the patent in suit is, therefore, novel. Since the issue of novelty has not been raised by the Appellant, it is not necessary to consider this matter in more detail.

4. It still remains to be examined whether the subject-matter of the patent in suit involves an inventive step.

The objection under Article 56 EPC raised by the Appellant is based on the fact that the application specifically claimed in Claim 1 was self-evident in the field of polyamides at the priority date of the patent in suit and that the advantageous properties put forward in the patent in suit (column 3, table) were in fact meaningless.

4.1 Although polyamide 4,6 was reported as early as 1938, as indicated in the introduction section of document (1) (page 537, Introduction, paragraph 2), it appears that the relatively late development of this polymer is due to the difficulty of synthetizing a product of high molecular weight. Although documents (1) and (5) were published about 40 years later, there is still no mention of polyamide 4,6 with both good color properties and an inherent viscosity higher than 2 dl/g in document (5), as noted above, or in document (1), irrespective of the method chosen to carry out the polycondensation step, i.e. in the solid state or in the melt (pages 539 and 540, Polymerization in combination with Table I and Figure 1). In fact, document (5) is nothing more than an experimental study of the various features and parameters which determine the rate of solid phase polymerization (page 137, paragraph 3; page 139, paragraph 2; pages 146/147, Conclusions) and does not contain the least reference to any application. Even if some properties of polyamide 4,6 are mentioned in document (1) (pages 543 to 545, Properties), the only application actually disclosed is the preparation of cast films (page 539, paragraph 4; page 544, line 3; page 545, paragraphs 3 and 4) which are fundamentally different from shaped objects. It is therefore not surprising to find no incentive in the prior art to modify the process of preparation of polyamide 4,6 in the way claimed by the Respondent in order to obtain molecular weights higher than in documents (1) and (5). That thereby an improved notched impact resistance may be achieved in accordance with the

problem underlying the patent in suit is, in the Board's judgement, plausible.

4.2 Moreover, in view of the numerous prior art which reflects the intensive works devoted to polyamide 4,6, as apparent from the lists of reference citations at the end of both documents (1) and (5), and the fact that extrusion and injection moulding operations had been standard applications for similar polyamides for many years before the priority date of the patent in suit, it can be fairly said that there was a long-felt need to extend such processing method to polyamide 4,6 which those involved in the field could not satisfy. As already appreciated in the non published decision T 41/84 of 12 March 1985, the fulfilment of such need is generally evidence of an inventive step.

4.3 As far as the properties of moulded objects are concerned, it is noted that the Appellant no longer disputes the validity of the comparative data provided in the patent in suit (column 3, Table) in order to demonstrate the superior Izod notched impact resistance of test bars made in polyamide 4,6 with regard to the figures obtainable with polyamide 6 and polyamide 6,6. Since the results are unexpectedly favorable for both dry and conditioned test bars in polyamide 4,6, an inventive step cannot reasonably be denied.

4.4 In conclusion, although the use of polyamides in extrusion and injection moulding may generally be regarded as usual, as the Appellant put forward repeatedly, the results obtained with polyamide 4,6 together with the history of this specific polymer confer an inventive step to this novel application.

5. The above conclusions show that the teaching of document (5), in isolation or in combination, cannot be

regarded as relevant and obviously does not prejudice the maintenance of the patent in suit. For this reason and in view of the late filing of document (5), the Board considers that the Opposition Division was entitled to exercise a discretionary power in relation to the admissibility of this document in the proceedings and eventually decide to disregard it under the terms of Article 114(2) EPC. In the Board's judgement thus, there can be no question of procedural violation.

6. Claim 1 being allowable, the same applies to Claims 2 to 4 which correspond to preferred embodiments of the main claim as well as to Claims 5 to 6 which, although drafted as independent process and product claims, recite all the features of the preparation of polyamide 4,6 as specified in Claim 1, since these subject-matters are supported by the same inventive idea.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

M. Beer K. Jahn