BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS | BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE | CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

A		B		C	X

File Number: T 563/90 - 3.3.3

Application No.: 82 201 284.5

Publication No.: 0 077 590

Title of invention: Process for the production of polymer filaments having high tensile strength and modulus

Classification: D01F 6/04

D E C I S I O N
of 28 October 1992

Applicant: DSM N.V.

Headword:

EPC Article 56

Keyword: "Inventive step (yes) - no incentive to operate along the line followed in the application in suit"

Case Number : T 563/90 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 28 October 1992

Appellant : DSM N.V.
Het Overloon 1
NL - 6411 TE Heerlen (NL)

Decision under appeal : Decision of Examining Division of the European Patent Office dated 24 November 1989, posted on 22 January 1990 refusing European patent application No. 82 201 284.5 pursuant to Article 97(1) EPC.

Composition of the Board :

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

Summary of Facts and Submissions

I. European patent application No. 82 201 284.5 filed on 15 October 1982, claiming priority of 17 October 1981 from an earlier application in the Netherlands and published under the publication No. 0 077 590, was refused for lack of novelty by a decision of the Examining Division dated 19 June 1985.

II. That decision was set aside by a decision of the Board of Appeal (Decision T 268/85 of 20 May 1988) and the case was referred back to the Examining Division for completion of the examination procedure.

III. By a decision given orally on 24 November 1989, with written reasons posted on 22 January 1990, the Examining Division refused again this application; this decision was based on a set of three claims, of which Claim 1 filed on 19 March 1985 reads as follows:

"Process for the production of polyethylene filaments having high tensile strength, wherein a solution of an ethylene polymer or copolymer containing at most 5% by wt of one or more alkenes with 3 to 8 carbon atoms and having a weight-average molecular weight Mw higher than 4.10^5 kg/kmole with at least 80% by wt of solvent is spun at a temperature above the gel point of that solution, the spun product is cooled to below the gel point and the filament obtained is stretched, in the form of a gel containing or not containing a solvent, to form a filament having a tensile strength of more than 1.5 GPa, measured at room temperature, characterized in that a polyethylene is used having a weight/number-average molecular weight ratio Mw/Mn lower than 5."

Claim 2 is a dependent claim directed to a preferred embodiment of the process according to Claim 1 and independent Claim 3 concerns a solution spun high molecular weight polymer filament, which is characterised by the polymer features mentioned in Claim 1, namely the qualitative and quantitative definition of the alkene, the weight-average molecular weight and the polydispersity.

IV. The ground for this decision was non-compliance with the requirements of Article 56 EPC with regard to the teaching of GB-A-2 042 414, which will be called document (6) hereinunder, and of the following documents:

(1) Ullmanns Encyklopädie der technischen Chemie, 4th ed., volume 19, page 191;

(4) US-A-4 268 470.

The technical problem underlying the application in suit could be defined as the production of polymer filaments having tensile strength and moduli at least comparable with those exhibited by the filaments obtained by the process disclosed in document (6). Since it was known from document (1) that a smaller dispersity generally leads to higher tensile strengths, the solution to that problem was self-evident and, therefore, not inventive. Further, the argument of a prejudice arising from the teaching of document (4) could not be accepted, for in that citation polydispersity was increased to improve creep properties, which could not be equated with tensile strength.

V. The Appellant (Applicant) thereafter lodged a Notice of Appeal against this decision on 21 March 1990 and paid the prescribed fee at the same time. In the Statement of Grounds of Appeal filed on 21 May 1990 the Appellant also referred to two documents which had been discussed during

oral proceedings before the Examining Division, but not mentioned in the decision of refusal, namely

(2) Encyclopedia of Polymer Science and Technology, Volume 7, page 271;

(3) Werkstoffkunde der Kunststoffe by G. Menges, Carl Hanser Verlag München Wien, 1985, pages 44 to 49;

and additionally introduced the following new document:

(5) Journal of Materials Science, P.J. Barham, A. Keller, volume 11, pages 27 to 35 (1976).

It was first underlined that what document (1) taught was not the influence of low polydispersity on tensile strength, but on toughness, which was an entirely different property. Further, that citation indicated that stretchability of polyethylene was enhanced by a broad molecular weight distribution; in view of the importance of that property in the preparation of high strength high modulus polyethylene filaments, the skilled man would not have considered a narrow molecular weight distribution. Furthermore, documents (2) and (3) did not confirm the alleged influence of the latter parameter on tensile strength. As to documents (4) and (5), both taught that a broad molecular weight distribution increased drawability, which showed the existence of a prejudice. In fact, the inventive step did not result from the choice of a narrow molecular weight distribution, but from the combination thereof with specific process features.

VI. The Appellant requested that the impugned decision be set aside and, implicitly, that a patent be granted on the basis of

- Claim 1 as filed on 19 March 1985;

- Claims 2 and 3 corresponding respectively to original Claims 3 and 10 (see letters received on 20 March 1985 and 30 March 1989), as quoted in the decision under appeal.

Reasons for the Decision

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

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

Claim 1 differs basically from Claim 1 as originally filed by the deletion of the expression "by spinning a solution of high molecular weight polyethylene and stretching the filaments" and by the fact that the characterising portion has been limited to the sole feature of the weight/number-average molecular weight ratio. The deletion of the above expression does not extend the subject-matter of the claim beyond the content of the application as filed, since that expression referred to general features of spinning processes, like spinning of a solution and stretching of filaments, which became superfluous after the transfer of the corresponding specific features from the characterising portion into the preamble. Further, the fact that most features have been transferred from the characterising part into the preamble and that the former has been limited to the weight/number-average molecular weight ratio has no impact on the definition of the process. It follows that Claim 1 is in substance equivalent to the originally filed Claim 1.

As to dependent Claim 2 and independent Claim 3, they correspond respectively to Claims 3 and 10 as originally filed.

3. The application in suit concerns a process for the production of polymer filaments having high tensile strength and modulus. Such a process is disclosed in document (6), which the Board, like the Examining Division, regards as the closest state of the art. This citation, identified in the application in suit by its application No. 8 004 157 in the United Kingdom (page 1, lines 4/5), describes a process which corresponds to the preamble of Claim 1; this was acknowledged by the Appellant in the Statement of Grounds of Appeal filed on 18 October 1985 in the appeal case T 268/85 and confirmed by the Board 3.3.2 in its decision of 20 May 1988. It is thus enough to state that in experiments 6 to 11 of Example 1 of document (6) polyethylene, having a molecular weight of 1.5×10^6, can be spun to filaments having tensile strengths between 1.65 and 3.02 GPa. As specified in the application in suit (page 1, lines 15 to 17), this prior art polyethylene has a weight/number-average molecular weight ratio of at least 6.5. However, such tensile strength can only be achieved by applying relatively high stretch ratios, which may cause undesirable intramolecular heat developments.

In the light of this prior art shortcoming, the technical problem underlying the application in suit may thus be seen as the definition of a process which does not require the application of such stretch ratios to obtain the same tensile strength, or, equivalently, whereby higher tensile strengths may be obtained by applying the same stretch ratios.

According to Claim 1 of the application in suit this problem is solved by using a polyethylene characterised by a weight/number-average molecular weight ratio lower than 5.

The comparative data in the application in suit provides evidence that the above-defined technical problem is effectively solved. More specifically, a comparison of tensile strength values in Tables 1 and 2 shows that one obtains better properties when the polydispersity index is 3.5 or 2.9, thus within the terms of the application in suit, than when that parameter is 7.5 or 9, thus according to the prior art document.

4. Since the issue of novelty was dealt with in appeal case T 268/85, it is not necessary in the absence of new documents in the procedure to consider this matter in further detail.

5. It still remains to be decided whether the claimed subject-matter involves an inventive step with regard to the cited documents.

5.1 The first question which arises concerns the actual teaching of document (1) since the Examining Division and the Appellant have given opposite interpretations of that disclosure.

After a discussion of the influence of molecular weight on various properties of polyethylene this citation teaches that products with a narrow molecular weight distribution exhibit a higher toughness, also at low temperature, whereas polymers with a broader molecular weight distribution are easier to process and more stretchable (page 191, left column, first paragraph, last sentence). This statement cannot be interpreted as a correlation of polydispersity with tensile strength, in particular as

meaning that smaller polydispersity leads to higher tensile strength, for toughness cannot be equated with tensile strength. As correctly pointed out by the Appellant (Statement of Grounds of Appeal, page 3, paragraph 4), toughness is defined as the amount of energy absorbed at break and is expressed consequently in J/m^2; by contrast, tensile strength is the force at break expressed in N/m^2.

Further, since a broad molecular weight distribution has a positive influence on stretchability and stretching is an essential step in the preparation of polymer filaments, the skilled man would be more likely to choose a high polydispersity index, which would be contrary to the requirement specified in the characterising part of Claim 1.

It follows that the correlations between polydispersity and toughness as well as between polydispersity and stretchability, as taught in document (1), cannot be an incentive to choose a narrow molecular weight distribution in order to increase the tensile strength of polyethylene filaments.

5.2 Although documents (2) and (3) illustrate the beneficial influence of a narrow molecular weight distribution on some mechanical properties, these teachings cannot be extended to the improvement of tensile strength.

Document (2) is a study of the influence of various parameters related to molecular weight and structure on the properties of polyethylene, in particular of polydispersity to the variations in stress-crack behaviour. As apparent from Table 3, the experiments in which less than 10% by weight of polymer are extractable by solvent correspond to the highest environmental stress-

cracking time; this would suggest that the low end of typical broad molecular weight distributions is heavily involved in the impairment of fracture resistance. However, that property, which gives the resistance to fracture at low stress under influence of a specific environment, is measured in time and is not identical to tensile strength.

Document (3) confirms the finding of document (1). In Table 4.6, page 46, it is indicated that a low weight/number-average weight ratio results in improved impact resistance, which, like toughness, is expressed in J/m^2. Figure 4.16, page 47, shows the influence of polydisperity on several physical polymer parameters, but does not consider tensile strength. This means that document (3) does not provide any pointer for the solution of the above-defined technical problem.

A further point to consider is that these studies have been made by using an unspecified polyethylene (document (2)) and an unspecified polymer (document (3)), i.e. products which must be regarded as isotropic compounds of average molecular weight. It is doubtful whether the results of such studies can be applied at all to high tensile strength filaments, i.e. to anisotropic oriented materials of very high molecular weight.

5.3 Document (4) concerns the drawing of polyethylene materials having a molecular weight higher than 3×10^5 and a weight/number-average molecular weight ratio higher than 5 (Claims 3 and 4). The filaments which may be obtained have high Young's moduli and improved creep properties (column 2, lines 63 to 68; Example 2). An essential feature in that process is the high value of polydispersity of the polymer which is preferably even greater than 9 (column 1, lines 42 to 68); in practice

polyethylenes with polydispersity indices of 16.4 and 9.5 are used (Examples 1 and 2). The tables illustrating the mechanical properties of drawn samples in Example 1 show that a high draw ratio applied to a material characterised by the lower molecular weight and the lower polydispersity leads to satisfactory properties.

Apart from the fact that there is no explicit mention of tensile strength, this finding could at most be an incentive to use a polyethylene having a polydispersity index at the lower end of a range, thus higher than 5 in any case, which is outside the range required in the application in suit.

5.4 Document (5) reports experiments which have been carried out with polyethylenes of different molecular weights. For that purpose, the original molecular weight was first modified by removal of the low molecular weight fraction, then reconstituted (page 28, left-hand column, lines 21 to 25). The modified material (type L) and the reconstituted material (type R) were stretched to obtain high modulus materials. Whereas the moduli of the R-type material corresponded broadly to those of the original material, the removal of the low molecular weight fraction had a detrimental effect on the drawing characteristics of polyethylene; in particular, only draw ratios lower than 20 could be satisfactorily achieved with the L-type sample (Figure 5 in conjunction with page 31, right-hand column, line 18 to page 32, left-hand column, line 7). The authors conclude by stating that the most stringent condition to obtain high moduli is the presence of some very low molecular weight fractions (page 33, right-hand column, first paragraph of Discussion). As pointed out by the Appellant, this clearly suggests a broad molecular weight distribution, thus a teaching against the line followed in the application in suit.

5.5 For these reasons, the Board is satisfied that the subject-matter of the application in suit as defined in Claim 1 involves an inventive step.

6. Claim 1 being allowable, the same applies to dependent Claim 2, which is directed to a preferred embodiment of the subject-matter of Claim 1, as well as to independent Claim 3, which concerns a solution spun high molecular weight polymer filament characterised by the same features as Claim 1, and whose inventiveness is supported by that of the main claim.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Examining Division with the order to grant a patent on the basis of Claims 1 to 3 on which the decision under appeal was based and a description yet to be adapted.

The Registrar: The Chairman:

E. Görgmaier F. Antony