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D E C I S I O N
of 18 May 2000

Case Number: T 0383/96 - 3.3.6

Application Number: 92902245.7

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Language of the proceedings: EN

Title of invention:
Solvated mesophase pitches

Applicant:
CONOCO INC.

Opponent:
-

Headword:
Mesophase Pitch/CONOCO

Relevant legal provisions:
EPC Art. 84, 123(2), 54, 56

Keyword:
"Novelty - yes, after amendment"
"Inventive step - yes, after amendment"

Decisions cited:
-

Catchword:
-



Case Number: T 0383/96 - 3.3.6

D E C I S I O N
of the Technical Board of Appeal 3.3.6
of 18 May 2000

Appellant: CONOCO INC.
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 10 November 1995
refusing European patent application
No. 92 902 245.7 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: P. Krasa
Members: G. Dischinger-Höppler
C. Rennie-Smith

Summary of Facts and Submissions

- I. This appeal is from the decision of the Examining Division to refuse European patent application No. 92 902 245.7 relating to solvated mesophase pitches for lack of novelty, since the carbon artifacts or fibers according to the then pending Claims 16 to 20 could not be distinguished from those carbon fibers which according to the application in suit were known in the art. No particular prior art document was cited by the Examining Division.
- II. During the appeal proceedings, the Appellant submitted several amended sets of claims as new requests, the main amendment being the replacement of the terms "carbon artifacts" and "carbon fibers" by "pitch artifacts" and "pitch fibers" respectively.
- III. Oral proceedings were held before the Appeal Board on 18 May 2000, in the course of which the Appellant submitted one single request (referred to here as the amended main request) on the basis of an amended set of 20 claims. The independent claims read:
- "1. Solvated mesophase pitch comprising a solution of solvent in mesogens, pseudomesogens or a mixture thereof wherein the solvated mesophase pitch is at least 40 volume percent optically anisotropic and wherein the solvated mesophase pitch melts at least 40°C lower than the mesogen component, or where the solvated mesophase pitch contains pseudomesogens, wherein the solvated mesophase pitch melts or fuses and the pseudomesogens do not.

9. A method for forming a solvated mesophase pitch as defined in Claim 1 comprising: (1) combining a carbonaceous aromatic isotropic pitch containing mesogens, pseudomesogens or a mixture thereof and aromatic oils with a solvent; (2) applying sufficient agitation and sufficient heat to cause the insoluble materials in said combination to form suspended liquid mesophase droplets; and (3) recovering the insoluble materials as solid or fluid solvated mesophase pitch.

14. A method for recovering solvated mesophase pitch as defined in Claim 1 from pseudomesogen comprising: (1) combining a carbonaceous aromatic pitch containing said pseudomesogens with a solvent; (2) applying sufficient heat to cause the insolubles to form suspended liquid solvated mesophase droplets or suspended mesophase solids; and thereafter, (3) recovering the separated insolubles, as fluid solvated mesophase pitch, or solid particles which upon further heating form fluid solvated mesophase pitch.

15. A pitch fiber whenever prepared by loss of solvent from solvated mesophase pitch as defined in any of Claims 1 to 8, said fiber having an oriented molecular structure and having the property without having been oxidatively stabilised and without subsequently being oxidatively stabilised, of not fusing when raised to a temperature above 400°C.

16. A pitch fiber whenever prepared from solvated mesophase as defined in any one of Claims 1 to 8, said fiber having an oriented molecular structure and having the property without having been oxidatively stabilised, of either oxidatively stabilising in reduced time as compared to pitch fibers formed from

non-solvated mesophase pitch having the same preparation temperature or of not requiring a fusion preventing stabilization step.

18. A method for preparing pitch fibers comprising: (1) combining and/or forming a mixture of a carbonaceous aromatic pitch containing mesogens or pseudomesogens and aromatic oils with a solvent; (2) applying agitation and sufficient heat and pressure to cause the insoluble materials in said combination to form suspended liquid solvated mesophase pitch droplets under solvent supercritical conditions of temperature and pressure; and (3) effecting phase separation of the solvated mesophase pitch from the solvent solution under solvent supercritical conditions of temperature and pressure; and (4) spinning the solvated mesophase pitch directly into pitch fibers or fibrils."

IV. The following prior art had been cited in the International Search Report:

- (1) EP-A-0 072 242;
- (2) EP-A-0 026 647; and
- (3) US-A-4 277 324.

The Appellant cited inter alia the following document:

- (4) W.C. Stevens, R.J. Diefendorf: "Thermosetting of Mesophase Pitches I: Experimental" in CARBON '86, 4th International Carbon Conference, Baden-Baden 30 June to 4 July 1986; Proceedings; Arbeitskreis Kohlenstoff der deutschen keramischen Gesellschaft e.V..

- V. The Appellant argued in essence that the pitch fibers as now claimed in Claims 15 to 17 were self-stabilizing and could be carbonised without any need for an oxidative stabilisation step. By contrast, known pitch fibers or artifacts, e.g. those mentioned in the prior art cited in the International Search Report, required oxidation bringing about a weight gain resulting from the attachment of oxygen to the product. In this context, reference was made inter alia to document (4).
- VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the amended main request.

Reasons for the Decision

1. *Amendments (Articles 123(2) and 84 EPC)*

The amended claims are supported by the claims as originally filed in combination with the following passages of the original description of the application in suit:

1.1 Concerning all claims:

The application in suit exclusively relates to mesophase pitch obtained from carbonaceous pitch where the mesophase forming mesogens are predominantly aromatic molecules (page 1, lines 8 to 14, page 3, lines 17 to 22 and page 4, lines 6 to 10, page 5, line 20 to page 6, line 3 and page 13, lines 24 to 27). By contrast, the claims as originally filed all use the term "mesophase" alone. This term, however, in its generally accepted meaning, implies any liquid crystal

material independent of its composition (document (2), page 2, lines 3 to 5). In order to find support in the description (Article 84 EPC) all claims have, therefore, been restricted to **mesophase pitch**.

1.2 Concerning Claims 1 and 9:

The inclusion of the term "or a mixture thereof" finds support on page 19, lines 3 to 5 of the application as originally filed.

1.3 Concerning Claim 7:

The added feature "optionally admixed with heptane" is based on the disclosure on page 18, lines 13 to 25 of the application as originally filed.

1.4 Concerning Claim 8:

Being one specific compound within the group of halogenated benzenes mentioned in Claim 7, the particular embodiment of "chlorofluorobenzene" has been excised from original Claim 7 and formulated in the new dependent claim 8 in accordance with Article 84 and Rule 29(4) EPC.

1.5 Concerning Claim 9:

The new wording of step (3) in Claim 9 reading "recovering the insoluble materials as **solid or** fluid solvated ..." is disclosed on page 25, lines 26 to 27 of the application in its original version.

1.6 Concerning Claims 11 and 18:

The application in suit contains no clear definition of the term "near supercritical conditions". Therefore the claims have been restricted to performance "at" or "under supercritical conditions" in order to comply with the requirements of Article 84 EPC.

1.7 Concerning Claims 15 to 19:

According to page 17, line 25 to page 18, line 12 in combination with page 27, line 21 to page 28, line 12 the term "carbon fibers" as used in original Claims 15 to 18 denotes the fibers produced from the solvated mesophase pitch after removal of the solvent but before carbonisation. It is, therefore, equivalent to the term "pitch fibers" as now used in the corresponding new Claims 15 to 19.

1.8 Concerning Claims 15 and 16:

The features "**without having been oxidatively stabilised**" in new Claims 15 and 16 and "**without subsequently being oxidatively stabilized**" in new Claim 15 are based on the term "without oxidative stabilization" in original Claim 15. They define the pitch fibers as spun from the solvated mesophase under loss of solvent which have not been subjected to oxidative stabilisation and, for carbonisation, need no or only reduced oxidative stabilization as compared to non-stabilised prior art pitch fibers. Basis for this definition can be found in the description as originally filed, on page 23, line 19 to page 24, line 6 and page 27, line 21 to page 28, line 23.

1.9 Concerning Claim 20:

Being a particular embodiment of the method of Claim 9, Claim 20 has been made dependent thereon. Further, the inclusion of the term "over a sufficient period" is based upon the disclosure on page 40, lines 12 to 16 of the description as originally filed, where it is stated that a settling period is required for the mixture to equilibrate and settle.

1.10 In summary, the Board concludes that no objections to the amended claims arise under Articles 123(2) and 84 EPC.

2. *Novelty*

2.1 Claims 15 to 17

Refusal of the application was based on the reason that the carbon fibers of the then pending Claims 16 to 20 were not distinguishable from prior art carbon fibers.

The corresponding new Claims 15 to 17 in the amended version define the fibers as spun from solvated mesophase pitch in their particular condition before carbonisation when they are still in pitch form. While documents (1) to (4) all describe pitch compositions suitable for carbon fiber production (see document (1), page 2, lines 10 to 26; document (2), Claim 5 and Example 5; document (3), column 7, lines 9 to 12 and document (4), page 37 "Experimental Procedure"), document (1) is silent as to any particular method of forming and treating the fibers and document (3) merely indicates that the fibers are spun from the neomesophase former fraction of the pitch which was obtained in dried form (column 8, lines 58 to 63 and column 9, lines 9 to 11).

Document (2) however discloses that the fibers are spun from molten or softened mesophase pitch and then thermoset before carbonisation by heating them in air at 2°C per minute to about 375°C (page 2, line 26 to page 3, line 5, page 6, lines 1 to 10 and Example 5).

This kind of process is discussed in more detail in document (4), where it is stated that pitch fibers obtained from mesophase by melt-spinning require thermosetting by heating in air or oxygen to prevent melting upon carbonisation (page 37, first and third paragraph). Experimental studies show that such thermosetting is accompanied by a weight gain as a function of time (page 37, fourth paragraph) due to the attachment of oxygen (page 38, second paragraph). Document (4) further indicates that the fibers generally require a weight gain of about 6% to be sufficiently stabilized for carbonisation (page 38, third paragraph).

The Appellant submitted that - contrary to the process of the state of the art - no significant weight gain by oxidation takes place during the spinning of pitch fibers from the solvated mesophase according to the application in suit. The prior art on file does not contradict this argument and the Board has no other relevant information at its disposal. The Board therefore concludes that the fibers of Claims 15 to 17 actually differ from the prior art melt-spun fibers both before and after thermosetting in that they exhibit the properties either of not fusing and stabilizing in reduced time or of not being oxidised to a considerable extent such as required by a weight gain of 6%.

The subject-matter of Claims 15 to 17 is, therefore, considered to be novel over the cited prior art.

2.2 Claims 1, 9, 14 and 18

In the application in suit it is stated that "solvated mesophase is formed as an intermediate during solvent fractionation of mesogen (or pseudomesogen) containing soaked pitches" (page 24, lines 25 to 27). This statement does not, however, clearly and unambiguously imply the two alternative embodiments of present Claim 1, namely that such intermediate solvated mesophase pitch fractions are at least 40 volume percent anisotropic and exhibit a melting point depression of at least 40°C as compared to the mesogen component contained therein or that said fractions contain infusible pseudomesogens as defined in the application in suit (page 5, lines 14 to 19). Since none of the prior art documents relates to or discloses solvated mesophase pitch, and in the absence of any further information concerning the composition of said intermediate pitch fractions, the solvated mesophase pitch according to Claim 1 is, therefore, not anticipated by the prior art.

The same applies to Claims 9 and 14 which relate to methods of forming and recovering solvated mesophase pitch according to these two alternative embodiments of product Claim 1, because processes for obtaining novel products must themselves also be novel.

Likewise, the process of Claim 18 is not anticipated by the prior art which does not mention any spinning from a solvated mesophase pitch.

Consequently, in agreement with the position of Examining Division which did not object to the then pending corresponding independent claims, the Board acknowledges the novelty of the subject-matter of these remaining independent claims.

- 2.3 Dependent Claims 2 to 8, 10 to 13 and 19 to 20 are directed to specific embodiments of the subject-matter of the respective independent claims and are, therefore, also considered to be novel.

3. *Inventive step*

The Board further agrees with the Examining Division's positive findings concerning inventive step. In particular, it is agreed that the technical problem as disclosed in the application in suit was to overcome the existing drawbacks of prior art mesophase pitch fibers, namely the requirement for an expensive oxidative treatment for stabilising the pitch fibers to render them infusible prior to carbonisation (see page 2, line 26 to page 3, line 14). The solution to this problem consists in providing the solvated mesophase pitch according to Claim 1 and pitch fibers produced therefrom in accordance with Claims 15 and 16 as well as in appropriate methods for preparing these products (Claims 9, 14 and 18).

In the application in suit it is stated that the solvated mesophase can become unmeltable on loss of solvent and that fibers spun from solvated mesophase will not fuse above 400°C even without oxidative stabilisation (page 23, line 27 to page 24, line 4 and page 28, lines 7 to 12). These statements are corroborated in particular by examples 8 and 10

indicating that fibers containing elongated mesophase structures of 85% anisotropy can be prepared and that the mesophase pitch is at least partly self-stabilising. Since no information to the contrary is available, the Board is satisfied that the claimed subject-matter solves the existing technical problem.

None of the prior art documents on file contains anything suggesting that pitch fibers should be directly spun from solvated mesophase pitch or even that such solvated pitch could be useful for improving prior art processes for manufacturing carbon fibers.

Therefore, the Board concludes that the solution of the existing technical problem as claimed in the application in suit was not obvious from the prior art documents on file, either alone or in combination, but rather that it involves an inventive step and, hence, complies with the requirements of Article 56 EPC.

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 an order to grant a patent on the following basis:

Claims: Claims 1 to 20 as contained in the amended main request;

Description: To be adapted thereto;

Drawings: Figures 1 to 19 as published.

The Registrar:

The Chairman:

M. Hoernell

P. Krasa