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D E C I S I O N
of 11 October 1996

Case Number: T 0209/94 - 3.3.3

Application Number: 91100412.5

Publication Number: 0438117

IPC: D01F 9/10

Language of the proceedings: EN

Title of invention:
Preparation of substantially crystalline silicon carbide fibers from polycarbosilane

Applicant:
DOW CORNING CORPORATION

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54

Keyword:
"Novelty (yes) - distinguishing functional feature"

Decisions cited:
-

Catchword:
-

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Case Number: T 0209/94 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 11 October 1996

Appellant: DOW CORNING CORPORATION
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 7 December 1993 refusing European patent application No. 91 100 412.5 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: C. Gérardin
Members: P. Kitzmantel
J. A. Stephens-Ofner

Summary of Facts and Submissions

I. This appeal, which was filed on 7 February 1994, lies against the decision of the Examining Division dated 7 December 1993, refusing European patent application No. 91 100 412.5 in the name of Dow Corning Corporation filed on 15 January 1991 and published under No. 438 117. Together with the Notice of Appeal the appeal fee was paid and a Statement of Grounds of Appeal was filed.

II. The decision under appeal referred to Claims 1 and 2 as originally filed and to Claim 3 as filed on 5 June 1992, which claims read as follows:

"1. A process for the preparation of thermally stable, substantially polycrystalline silicon carbide fibers comprising: forming fibers from a preceramic polymer comprising a polycarbosilane resin having at least about 0.2% by weight boron incorporated therein; infusibilizing said fibers; and pyrolyzing said fibers at a temperature of greater than about 1600°C. in a nonoxidizing environment."

"2. A process for the preparation of thermally stable, substantially crystalline silicon carbide fibers comprising: forming fibers from a preceramic polymer comprising a polycarbosilane resin; infusibilizing said fibers; and pyrolyzing said fibers at a temperature of greater than about 1600°C. in a non-oxidizing environment, wherein at least about 0.2% by weight boron is incorporated during said infusibilization or said pyrolysis."

"3. Substantially polycrystalline [silicon] carbide fibers containing at least 0,2 weight percent boron and having a density of at least 2,9 g/cm^3, an average grain size of less than 0,5 micrometers and an average tensile strength of at least 164 ± 47 ksi, virtually all of the oxygen and/or nitrogen originally present in, or introduced into, the fiber having been removed by a high temperature pyrolysis step."

The word "silicon", inserted into the above quotation of Claim 3 in square brackets, was inadvertently omitted in its version as filed. Further, in the same claim an editorial omission in the word "temperature" has been corrected.

According to that decision the statement in Claim 3 "at least 164 ± 47 ksi" amounted to an open-ended range of the tensile strength which was not backed by the individual tensile stength values exemplified in the original application. Claim 3 did not, therefore, comply with the requirement of Article 123(2) EPC.

Furthermore, according to that decision, the subject-matter of Claim 1 was deprived of novelty by the process for the preparation of silicon carbide fibers by infusibilizing and pyrolyzing precursor fibers from boron containing polycarbosilane polymers known from document

D1: DE-A-3 447 411,

since this document encompassed pyrolyzing temperatures of up to 1800°C which fell within the temperature range of >1600°C as required according to said Claim 1. In view of the wording of this claim, which allowed for the presence of nitrogen in the fibers, as was the case according to D1, this feature was not considered distinguishing. The fact that in the worked examples of

D1 pyrolyzing temperatures of only 1300°C were used could also not establish the novelty of the process according to Claim 1 of the application in suit, since this did not mean that the temperature range of between 1600° and 1800°C, which was within the general disclosure of D1, would be ineffective.

III. With his Statement of Grounds of Appeal the Applicant (Appellant) requested rectification of the decision on the basis of an amended set of two claims. With a letter dated 5 April 1994 (i.e. well after expiry of the time limit of one month set for interlocutory revision under Article 109(2) EPC) he replaced these claims by a set of new claims, which, following a suggestion of the Board, have been further amended and, according to the Appellant's submission dated 2 October 1996, read as follows:

"1. A process for the preparation of thermally stable, substantially polycrystalline silicon carbide fibers comprising:

forming fibers from a preceramic polymer comprising a polycarbosilane resin having at least about 0.2% by weight boron incorporated therein;

infusibilizing said fibers; and

pyrolyzing said fibers at a temperature of greater than about 1600°C. in a nonoxidizing environment for a period of time sufficient to reduce oxygen and/or nitrogen content of the fibers to below about 0,5% by weight."

"2. A process for the preparation of thermally stable, substantially polycrystalline silicon carbide fibers comprising:

forming fibers from a preceramic polymer comprising a polycarbosilane resin; infusibilizing said fibers; and

pyrolyzing said fibers at a temperature of greater than about 1600°C. in a non-oxidizing environment for a period of time sufficient to reduce oxygen and/or nitrogen content of the fibers to below about 0,5% by weight, wherein at least about 0.2% by weight boron is incorporated during said infusibilization or said pyrolysis."

The Appellant contended that by restricting the claims to the preparation of boron containing silicon carbide fibers having an oxygen and/or nitrogen content of below 0.5% by weight not only the novelty of the subject-matter of Claims 1 and 2 was established, but also their foundation on an inventive step was made conspicuous. This, because D1 required the presence of higher amounts of nitrogen in the fibers, which, in consequence, had lower tensile strengths.

IV. The Appellant requested that the appealed decision be set aside and that a patent be granted on the basis of the following documents:

Claims 1 and 2, as submitted on 2 October 1996;
Description pages 1 to 3 and 6 to 10 as originally filed;
Description page 4 as filed on 2 October 1996;
Description pages 5 and 11 to 13 as filed on 4 March 1993.

Reasons for the Decision

1. The appeal is admissible.

2. *Article 109 EPC*

According to Article 109(1) EPC the department whose decision is contested shall rectify its decision if it considers the appeal to be admissible and well founded.

In the present case the Examining Division did not rectify its decision within the time limit stipulated in Article 109(2) EPC. Since thereafter the Appellant himself abandoned the claims filed together with his Notice of Appeal and on the basis of which he had requested rectification of the appealed decision, it must be assumed that the request for rectification on the basis of these claims was not considered to be "admissible and well founded" by the Appellant himself. The Board sees no reason for a different opinion on this matter.

3. *Article 123 (2) EPC*

3.1 Neither of the two claims now operative contains the open-ended range for the "average tensile strength" of "at least 164 ± 47 ksi", which range in the appealed decision was, in contravention of the requirement of Article 123(2) EPC, considered to extend beyond the content of the application as filed.

3.2 The identical statement in Claims 1 and 2, namely:

"for a period of time sufficient to reduce oxygen and/or nitrogen content of the fibers to below about 0,5% by weight" is based on page 9, second paragraph, first sentence of the original application. Otherwise

these claims correspond fully to those which have been originally filed. Thus, they comply with the requirement of Article 123(2) EPC.

4. *Novelty (Article 54 EPC)*

4.1 Document D1:

This document discloses a method for preparing inorganic fibers comprising silicon, carbon, boron and nitrogen, whereby preceramic fibers of a boron-containing polycarbosilane are infusibilized and thereafter pyrolyzed in vacuum or an inert gas at a temperature within the range of from 900 to 1800°C, preferably of from 1000 to 1600°C (see Claims 1 and 9; page 6, line 5 to page 7, line 5; page 11, line 23 to page 12, line 12). From Table 1, page 16 it appears that these fibers, like further prior art fibers of the same composition, contain additional amounts of oxygen.

According to the worked examples of D1 the pyrolyzing step is carried out at a temperature of 1200°C (Example 1, page 14, line 16; Example 4, page 19, line 21) or 1300°C (Example 2, page 15, line 18; Example 3, page 18, line 26). The results of elementary analysis and of tensile strength measurements (tested at room temperature and after treatment for 1 hour at temperatures of from 500° to 1500°C: see page 15, lines 21 to 31; page 16) of the pyrolyzed fibers prepared according to Example 2 are disclosed in Table 1 on page 16. Dependent upon the insolubilizing process employed (by oxidative treatment or by electron beam irradiation, respectively), the fibres according to Example 2 of D1 are reported in this Table 1 to have a nitrogen content of 1.15% or of 1.18% by weight and an oxygen content of 7.56% or of 2.71% by weight (establishing a combined content of oxygen and nitrogen of, respectively, 8.71% and 3.89% by weight). As can

best be seen in Figure 1, the fibers prepared in accordance with the teaching of D1 (see curves "I" and "II") maintain their high tensile strength at high temperatures much better than a conventional Si-C fiber (see curve "III"). However, even the tensile strength of the fibers according to D1 starts to drop quite rapidly at temperatures above about 1100°C. It is speculated on page 12, lines 20 to 33 of D1 that the BC- and BN-portions which are located between very small crystallites from β-SiC, prohibit the latters' growth at temperatures of 1000°C and above and prevent thereby too high a drop of the fiber strength at high temperatures. From this it can be concluded that the presence of nitrogen (and boron) in the fibers in certain amounts is essential for their temperature stability.

4.2 Since this improved thermal stability of the fibers is the very object of the invention disclosed in D1 (see e.g. page 4, first paragraph), fibers which would not meet this requirement cannot be considered to be within the teaching of this document. This means, by implication, that fibers having a nitrogen (and boron) content which is too low to produce the desired thermal stability are not within the scope of the invention disclosed in D1. Taking into account that the lowest combined content of nitrogen and oxygen disclosed in D1 is 3.89% by weight (corresponding to 1.18% by weight of nitrogen: Table 1, fibers infusibilized by electron beam irradiation) and that both these amounts are rather remote from the upper limit of the combined content of nitrogen and/or oxygen of about 0.5% by weight set according to Claims 1 and 2 of the application in suit, it must be concluded that the

preparation of fibers having contents of nitrogen and/or oxygen which are as low as this maximum amount permitted according to the application in suit are not within the disclosure of document D1.

In consequence, the fiber preparation process carried out according to Claim 1 of D1 and including a pyrolyzing step in a temperature range of from 900° to 1800°C cannot be considered to yield fibers having a combined nitrogen and/or oxygen content of below about 0.5% by weight. Rather this process must be carried out in such a fashion as to ensure the higher contents of nitrogen and/or oxygen exemplified in D1 which are required to achieve the desired thermal stability of the ultimate fibers.

4.3 The functional limitation of the pyrolyzing step established by the statement introduced into Claims 1 and 2 of the application in suit "for a period of time sufficient to reduce oxygen and/or nitrogen content of the fibers to below about 0,5% by weight" is therefore able to distinguish the process defined in Claims 1 and 2 of the application in suit from the process according to D1 in spite of the fact that the temperature ranges of the pyrolyzing steps are overlapping ("greater than about 1600°C" according to the present invention and "from 900° to 1800°C" according to D1).

Consequently, the subject-matter of Claims 1 and 2 of the application in suit is novel over the disclosure of document D1.

5. *Inventive step (Article 56 EPC)*

From the arguments presented by the Appellant it appears that the problem underlying the application in suit was the provision of a process for the preparation silicon carbide fibers having improved mechanical properties (average tensile strength and elastic modulus).

This problem was allegedly solved by the claimed pyrolyzing step at a temperature of greater than 1600°C for a period of time sufficient to reduce oxygen and/or nitrogen content of the fibers to below about 0.5% by weight.

The assessment of inventive step therefore turns on the question whether, for the skilled person starting from the disclosure of document D1 and wishing to solve the above-mentioned technical problem, it would require inventive skill to arrive at this solution.

In order to be able to answer the above question, it is necessary, as a first step, to ascertain whether the claimed pyrolyzing step is indeed capable of providing silicon carbide fibers having the desired properties. It appears, however, that the evidence present in the application in suit is not sufficiently convincing. On the one hand, data concerning the nitrogen and/or oxygen content of the pyrolyzed fibers are missing, and, on the other hand, the results of the measurements of tensile strength and elastic modulus of the reported for Examples 1, 2 and 3 are inconsistent: only Example 1 is able to demonstrate that by a second pyrolyzing step at 1800°C (which step is considered to be necessary for reducing the oxygen and/or nitrogen to below about 0.5% by weight) the average tensile strength of the fibers, which previously had already been pyrolyzed at 1200°C, can be further improved (here

from 215 ± 49 Ksi to 236 ± 72 Ksi); contrary thereto according to Examples 2 and 3 the average tensile strength of the fibers pyrolyzed at 1200°C of 247 ± 47 Ksi and 271 ± 63 Ksi, respectively, drops to 164 ± 47 Ksi and 243 ± 19 Ksi, respectively, after pyrolyzing at 1800°C; similarly, according to Examples 1 and 3 the elastic modulus of the fibers is enhanced by pyrolyzing at 1800°C from 25,0 ± 3,1 Msi and 25,9 ± 1,9 Msi, respectively, to 32,0 ± 2,4 Msi and 39,1 ± 1,6 Msi, respectively, whereas according to Example 2 the pyrolyzing treatment at 1800°C causes the elastic modulus to drop from 27,7 ± 1,3 Msi to 25,7 ± 1,3 Msi. These data appear to be unable to prove that the desired property improvement can be achieved in a consistent fashion, thus casting doubt on the reliability of the claimed solution of the alleged problem.

Furthermore a comparison with the tensile strength data disclosed in D1 is not straightforward and the conversion of the value of 220 x 10^{-1} N/mm^2 in Table 1 of D1 (= highest 1500°C value) to 1,1575 GPa offered on page 4, lines 18 to 21 of Applicant's letter of 4 March 1993 is incomprehensible.

In view of this situation and taking into account the fact that the matter of inventive step was not dealt with in the appealed decision it appears appropriate for the Board to exercise its powers under Article 111 EPC and to remit the case to the Examining Division for further examination.

Order

for these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance for further prosecution.

The Registrar:

E. Görgmaier

The Chairman:

C. Gérardin