BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

Internal distribution code:
(A) [ ] Publication in OJ
(B) [ ] To Chairmen and Members
(C) [X] To Chairmen

D E C I S I O N
of 27 September 1994

Case Number: T 0656/91 - 3.3.2

Application Number: 85300084.2

Publication Number: 0153000

IPC: C04B 35/56

Language of the proceedings: EN

Title of invention:
Refractories of silicon carbide and related materials having a modified silicon nitride bonding phase

Patentee:
The Carborundum Company

Opponent:
Annawerk Keramische Betriebe GmbH

Headword:
Refractories/CARBORUNDUM

Relevant legal provisions:

EPC Art. 83, 56

Keyword:
"Sufficiency of disclosure (yes)"
"Inventive step (yes) - non-obvious modification"

Decisions cited:
T 0150/82, T 0014/83, T 0219/83, T 0226/85, T 0390/88, T 0435/91

Catchword:
-

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0656/91 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 27 September 1994

Appellant: (Opponent)
Annawerk
Keramische Betriebe GmbH
Oeslaler Strasse 35
D-96472 Rödental (DE)

Representative:
Bockhorni, Josef, Dipl.-Ing.
Patentanwälte
Herrmann-Trentepohl, Kirschner, Grosse, Bockhorni
Forstenrieder Allee 59
D-81476 München (DE)

Respondent: (Proprietor of the patent)
The Carborundum Company
200 Public Square
Cleveland
Ohio 44114-2375 (US)

Representative:
Lederer, Franz
Prinzregenten Straße 16
D-80538 München (DE)

Decision under appeal: **Decision of the Opposition Division of the European Patent Office dated 8 March 1991, posted on 21 June 1991 rejecting the opposition filed against European patent No. 0 153 000 pursuant to Article 102(2) EPC.**

Composition of the Board:

Chairman: A. J. Nuss
Members: I. A. Holliday
R. L. J. Schulte

Summary of Facts and Submissions

I. European patent No. 0 153 000, relating to refractories of silicon carbide in a ceramic bonding phase, was granted on the basis of 17 claims contained in European patent application No. 85 300 084.2

Claim 1 reads as follows:

"1. A bonded silicon carbide article consisting of silicon carbide and a sialon bond phase, characterised in that the article is made by a process consisting of:
(a) forming a uniform mixture consisting essentially of from 4 to 8 percent aluminium powder, from 10 to 16 percent silicon powder and the remainder being granular silicon carbide and optionally, a temporary binder,
(b) shaping the mixture into a green compact in the form of the article,
(c) firing the green compact in a non-oxidative nitrogenous atmosphere until substantially all of the silicon and aluminium is combined with nitrogen to complete formation of the article; wherein the bond phase comprises from 50 to 80 weight percent Si-Al-O-N, and contains from 1½ to 6½ weight percent oxygen based on the total weight of bond phase, and from 6 to 20 weight percent aluminium based on the total weight of bond phase."

Independent Claim 6 related to a raw material batch for the manufacture of a bonded silicon carbide article according to Claim 1 and independent Claim 10 to a method of manufacturing such a bonded silicon carbide article.

II. The Appellant filed an opposition against the granted patent, raising objections under Articles 100(a) and 100(b) EPC. Of the documents cited only the following remains relevant to the present decisions:

(12) Bauer et al, Int. Feuerfest Kolloquium, Aachen, 14 and 15 October 1982, pages 22 to 27.

The Opposition Division held that the requirements of Article 83 EPC were satisfied. Although the claims specified only elemental Al and Si as starting materials, the oxygen required to form the sialon could come from impurities in the starting materials. The expression "non-oxidative atmosphere" would not necessarily exclude the presence of oxygen but imply a gas of low oxygen content.

Novelty was recognised since the bond phase of known materials had a greater oxygen content than the products of the patent in suit.

The Opposition Division considered document (12) to be the closest prior art. Neither (12) nor any other of the cited documents suggested the production of SiC bonded materials with such a low oxygen content. There was no incentive from the prior art to avoid the use of oxides as starting materials for the preparation of the sialon binding phase. An inventive step was thus recognised.

III. The Appellant lodged an appeal against the decision of the Opposition Division. Oral proceedings took place on 27 September 1994. At the oral proceedings, the arguments of the Appellant were confined to sufficiency and inventive step.

IV. The arguments of the Appellant, both in the written procedure and at the oral proceedings may be summarised as follows:

Under Article 100(b), the Appellant objected to the formulation of the claim and also questioned whether the process was reproducible. The Appellant considered that a combination of a "product-by-process" claim and a "product-by-parameter" claim was neither permissible nor sufficient for defining the product to be protected. The claim defined the starting materials as 4 to 8% by weight of powdered aluminium, 10 to 14% by weight of silicon powder and the remainder granular silicon carbide; the "non-oxidative nitrogenous atmosphere also implied nitrogen as starting material. The product was defined in terms of the oxygen and aluminium content of the bond phase. Not only was it not clear how the specified amount of aluminium could be achieved but also the claim gave no indication of where the oxygen came from. The description referred to impure starting materials and bentonite clay (which contained 68% by weight of SiO_2) was mentioned as a temporary binder in Claim 6. The Appellant argued that this was insufficient to yield a sialon with the requisite oxygen content. The Appellant also argued that free carbon in the SiC would also take up oxygen in competition with the sialon forming reaction. Attention was drawn to a Review in Trans. J. Br. Ceram. Soc., **81**, 141-144 (1982) (document (1) in the opposition procedure) indicating that ß-sialon formed a very small region of a quaternary phase diagram. The Appellant also saw an inconsistency between Claim 1 which specified that "substantially all of the Si and Al is combined with nitrogen" and a passage in the description of page 6 of the patent in suit which referred to Si_3N_4 containing Al in solid solution. The essential point of the Appellant's argumentation was that considerable experimentation would be necessary in

order to achieve a product having the desired properties. Decision T 226/85 (OJ EPO 1988, 336) was referred to.

In respect of inventive step, the Appellant criticised the Opposition Division's analysis of the prior art, arguing that documents other than (12) related to sialons having low oxygen content. The Appellant considered the disclosure of (12) to be rather vague and referred to US-A-4 242 621 (13) which had been mentioned at the examination stage; it was considered that Examples 23 in particular was very close to the claimed subject-matter. The problem underlying (13), i.e. providing a refractory material suitable for lining a blast furnace, appeared to be essentially the same as that of the patent in suit. The Appellant questioned whether the products of the patent in suit had superior properties to those known from (13). At the oral proceedings there was considerable discussion of Table IV on page 8 of the disputed patent, the Appellant arguing that Example 7, which was outside the scope of the claimed subject-matter appeared to have superior physical properties to Example 6 which was in accordance with Claim 1.

V. The Respondent's arguments in the written procedure and during the oral proceedings may be summarised essentially as follows:

The Respondent argued that Claim 1 when read in conjunction with the description gave clear instructions relating to the starting materials and process. Only products having the parameters specified relating to the bond phase, oxygen and aluminium content would fall within the claims; i.e., any material which was prepared by such a process which failed to exhibit the said parameters would be excluded. Several passages in the

description indicated oxygen sources. The Respondent had never maintained that the bond phase was a homogeneous sialon, especially having regard to the passage on page 6 referring to a solid solution of Al in Si_3N_4. The Respondent had provided two examples in accordance with Claim 1 which together with the general description provided adequate information to enable one skilled in the art to operate the process and obtain the desired product. In any event neither the nature of the bonding phase nor the mechanism of its formation had been mentioned in the original statement of opposition.

In arguing for the presence of inventive step, the Respondent denied that the finished product and the raw material batch were known in the prior art; each of the documents (1) to (12) cited in the opposition were analysed. The Respondent argued that any comparison with document (13) included a wide variety of refractory materials of which sialon bonded SiC was merely an example. There was no reason why the skilled person would select this particular material from the broad disclosure of the patent. However, in respect of alkali resistance the products of the patent in suit were superior to those known from (13). In any event, the comparison experiments in the patent in suit related to closer art than that of (13). When considering Examples 6 and 7 of the patent in suit, the Respondent argued that the overall properties were of importance. From Table VI, it was apparent that the steam oxidation resistance of the product of Example 6 was superior to that of Example 7. The Respondent concluded that nowhere is the combination of starting materials, the manufacturing process or the final product disclosed, made obvious or suggested by the prior art.

VI. The summons to oral proceedings was accompanied by a communication pursuant to Article 11(2) EPC of the rules of procedure of the Boards of Appeal. The communication indicated that (13) was considered to be the closest state of the art and that Example 7 thereof appeared to be especially relevant.

VII. The Appellant requested that the decision of the Opposition Division be set aside and that the patent be revoked.

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

2. This decision is based on the claims as granted. The Board has no reason to doubt that the requirements of Article 123 are satisfied.

3. *Sufficiency of disclosure*

3.1 The refractory article of the patent in suit consists of silicon carbide and a bond phase. According to Claim 1, the bond phase comprises 50 to 80% by weight of sialon. It is generally known that sialons have a complex structure. Since the bond phase is not simply a sialon but contains other material believed to be Si_3N_4 containing aluminium in solid solution (page 6, lines 15 to 16), it cannot be adequately defined in absolute terms. Accordingly the claim satisfies the criteria for a product-by-process claim set out in decision T 150/82 (OJ EPO 1984, 309) since it sets out the necessary step to be taken in order to manufacture the desired product,

which the Appellant has not shown to be either inadequate nor ineffective for obtaining a bonded product consisting of silicon carbide and a sialon bond phase. In such a situation, however, the sufficiency of the disclosure cannot be successfully challenged by objecting that no complete **product** characterisation is available to one skilled in the art.

3.2 As far as the indicated parameters of the product are concerned, the Board can accept the Respondent's arguments that only products which have a bond phase as specified in part (c) of Claim 1 fall within the claimed subject-matter. Products not fulfilling these criteria would be excluded.

3.3 However, the question as to whether an invention has been disclosed sufficiently clearly and completely for it to be carried out by a person skilled in the art is not to be decided solely on the basis of the content of the claims (see T 14/83, OJ EPO 1984, 105). Although Claim 1 fails to mention a source of oxygen required to produce the sialon, the description states on page 2, lines 52 to 53 that such oxygen is derived from surface oxygen present on the silicon and silicon carbide raw materials. This is amplified in respect of powdered silicon on page 4, lines 42 to 43 and in respect of silicon carbide on page 5, lines 21 to 22. It is also apparent from the table at the foot on page 5 that bentonite clay, used as a temporary binder consists essentially of oxides, especially SiO_2. Having regard to the starting materials specified in Claim 1, it is also clear that the binder forms a minor proportion of the total refractory body, a preferred range of 15 to 25% by weight being mentioned on page 5, lines 39 to 41. Thus 1% by weight of oxygen present in the total weight of starting material could represent at least 4% by weight

based on the weight of the binder. Such would be sufficient to give a bond phase with the desired oxygen content.

3.4 The Appellant argued at the oral proceedings that free carbon present in the silicon carbide would also react with oxygen in competition with the sialon forming reaction. The argument was not, however, backed with experimental evidence and can thus be regarded merely as an unsubstantiated allegation not sufficient to challenge the sufficiency of the disclosure (see T 219/83, OJ EPO 1986, 211).

3.5 According to Claim 1 "substantially all" of the silicon and aluminium is combined with nitrogen in the formation of the refractory article; such would include the formation of Si_3N_4 as well as the sialon. Although page 6 noted above refers to a solid solution of Al in Si_3N_4, this need not necessarily be inconsistent with "substantially all" of the aluminium reacting since no figures are given as to how much aluminium might be present in the solid solution.

3.6 The Appellant referred to decision T 226/85 according to which substantially any embodiment of the invention, as defined by the broadest claim, must be capable of being realised by the disclosure. However, what the Appellant has failed to show in the present case is that by following the instructions contained in the patent in suit, i.e. by construing the claims in conjunction with the description, the skilled person might systematically obtain an unsatisfactory product not having the desired refractory properties. Some trial and error might indeed be involved in obtaining the required product but this is in conformity with decision T 226/85 which follows the already mentioned T 14/83, referred to therein. Such trial and error experiments would not amount to carrying

out a "research programme" in order to obtain the desired product (cf. decision T 435/91 of 9 March 1994, to be published in OJ EPO, Reasons, point 2.2.1 last paragraph).

3.7 Accordingly, the Board is satisfied that requirements of Article 83 EPC are satisfied.

4. *Novelty*

The Board is convinced that none of the documents cited during the procedure discloses the article of Claim 1, the raw batch mixture of Claim 6 or the method of Claim 10. In any event novelty is no longer in dispute.

5. *Problem and solution*

5.1 The Board considers document (13) to be the closest prior art. Examples 7 of (13) relates to a silicon carbide powder bonded with a sialon prepared from a mixture of aluminium, silicon and silica, all in powder form. The SiC and the binder components are mixed and the whole is sintered together to form a sialon bonded SiC in which the bond phase comprises 35% by weight of the whole. The product of Example 7 is stated to have good refractory properties (column 14, lines 42 to 45 and Table 3). The products prepared in accordance with document (13) are suitable for the lining of blast furnaces (column 12, line 1).

5.2 Starting from (13), the problem to be solved can be seen in providing an alternative sialon bonded silicon carbide refractory which is suitable as a lining for blast furnaces. Having regard to the examples and comparative examples which appear in the patent in suit,

the Board is satisfied that the problem has indeed been solved by the bonded silicon carbide article presently claimed.

6. *Inventive step*

6.1 According to the patent in suit a refractory body is manufactured by first forming a mixture (by weight) of 4 to 8% aluminium powder, 10 to 16% silicon powder and a balance of granular silicon carbide. The mixture is shaped into a green compact body which is preferably raised gradually to 1350 to 1600°C (Claim 12) and then sintered at such a temperature for at least 6 hours. A preferred temperature is 1420°C (Claim 13 and Examples). The bonded phase preferably comprises 15 to 20% by weight of the entire body (page 5, lines 39 to 41).

6.2 Example 7 of document (13) relates to a sialon bonded refractory body manufactured from a mixture (by weight) of silica fines (14%), atomised aluminium powder (9%), silicon powder (12%) and powdered silicon carbide of a specified particle size distribution (65%). The mixture is compressed and heated gradually to 1450°C, nitriding sintered at this temperature for 10 hours. The bonding phase is stated to be a ß'-sialon. In Example 23 of (13), referred to by the Appellant, a preformed sialon from a mixture (by weight) of silica fines (60 parts), atomised aluminium (40 part) and silicon powder (50 part) is crushed to a mean particle size of 1.2 µm. The said sialon powder (30% by weight) is mixed with powdered silicon carbide (70%), moulded and finally sintered at 1750°C. Other examples related to sialon bonded alumina, sialon bonded silicon nitride and sialon bonded silicon nitride/boron nitride mixtures. Out of 24 worked Examples only Examples 7 and 23 relate to sialon bonded SiC. In each case the sialon is prepared from a mixture of silica, silicon and aluminium. There is thus

no hint in document (13) that a sialon bonded SiC could be prepared from a mixture which **avoided** the use of silica.

6.3 Document (12), regarded by the Opposition Division as the closest prior art, describes in general terms that sialon bonded SiC may be prepared from a mixture of SiC, silicon metal and ultrafine alumina. In this case, an oxide, alumina, is again required in order to prepare the sialon.

6.4 At the oral proceedings, the Appellant admitted that not one of the documents cited during the opposition or examination procedure gave any hint that it might be possible to prepare a sialon bonded SiC from a mixture of SiC, Si and Al, that is without the use of an oxide (silica or alumina). In the judgement of the Board, the process of Claim 10 of the patent in suit must thus be regarded as a non-obvious alternative process for the manufacture of a sialon bonded SiC refractory; an inventive step can accordingly be recognised. Analogous arguments apply to the product-by-process Claim 1 and the raw material mixture of Claim 6. The dependent claims derive their patentability from Claims 1, 6 and 10 respectively.

7. At the oral proceedings, there was considerable discussion concerning the examples and comparative examples of the patent in suit and of the comparative experiments in the Respondent's letter dated 13 September 1994. Particular doubt was cast on the latter since the test method was not clearly defined. Since the Board has decided that the process of the patent in suit is a non-obvious alternative which is in no way foreshadowed by the prior art, it is not necessary to demonstrate that the products thereof have

advantageous properties over those known from the said prior art (cf. decision T 390/88 of 20 February 1990, not published in OJ EPO, esp. Reasons point 14).

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

P. Martorana A. J. Nuss