DECISION of 17 March 2006

Case Number: T 0721/03 - 3.3.05
Application Number: 94909546.7
Publication Number: 0746532
IPC: C04B 35/52
Language of the proceedings: EN

Title of invention:
Dense, self-sintered silicon carbide/carbon-graphite composite and process for producing same

Patentee:
THE MORGAN CRUCIBLE COMPANY PLC

Opponent:
Wacker Chemie AG

Headword:
Silicon carbide composites/MORGAN CRUCIBLE

Relevant legal provisions:
EPC Art. 114(2), 56

Keyword:
"Inventive step (no)"

Decisions cited:
-

Catchword:
-
Case Number: T 0721/03 - 3.3.05

DECISION
of the Technical Board of Appeal 3.3.05
of 17 March 2006

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Decision under appeal: Interlocutory decision of the Opposition
Division of the European Patent Office posted
15 May 2003 concerning maintenance of European
patent No. 0746532 in amended form.

Composition of the Board:
Chairman: M. Eberhard
Members: J.-M. Schwaller
J. Willems
Summary of Facts and Submissions

I. The appeal was lodged by the proprietor of the patent against the interlocutory decision of the opposition division posted on 15 May 2003, to the extent that the main request (claims as granted) and the first, second and third auxiliary requests submitted during the oral proceedings of 18 March 2003 were rejected.

II. The following prior art documents were inter alia relied upon during the opposition proceedings:

E1: English translation of JP 63-260861
E4: DE 3329225 C2

III. In the decision, the opposition division argued principally as follows:

The subject-matter of claim 1 of the main request differed from E1 in the average grain size of silicon carbide and graphite. Paragraph [0005] of the opposed patent indicated that the objective of the invention was to incorporate large amounts of graphite into a ceramic matrix without causing cracks in the microstructure or without increasing the material porosity. The porosity appeared to be a very important physical property of the composite of the invention when used as a seal. It was not made plausible that the
specified selection of average grain sizes of silicon carbide and graphite could lead to any particular effect. In particular, it was not demonstrated that this particular selection could lead to a reduction of the porosity. The subject-matter of claim 1 of the main request was therefore not inventive.

In claim 1 of the 1st auxiliary request, the qualification "impervious" merely represented a vague result to be achieved and did not limit the scope of protection thereof. As a consequence, the subject-matter of this claim was not considered as inventive.

Figure 2 of E1 disclosed that a density of 90% of the theoretical density could be achieved by using an amount of graphite lower than 10% by volume. This was also shown by Example 59 of Table 11 of E4 which indicated a density of 91.8%. As a consequence, the subject-matter of claim 1 of the 2nd auxiliary request was not considered inventive over the cited prior art.

The 3rd auxiliary request being the combination of the 1st and 2nd auxiliary requests, claim 1 of this request lacked an inventive step for the reasons indicated before for the 1st and 2nd auxiliary requests.

IV. In the grounds of appeal, the appellant maintained the four sets of claims rejected in the decision and filed two graphs, respectively called "Graph 1" and "Graph 2", along with observations.

V. In a communication, the board informed the parties of its preliminary opinion that the subject-matter of claims 1 and 12 of the main request and claims 1 and 11
of the 2nd auxiliary request lacked an inventive step. It also objected to the term "impervious" in the 1st and 3rd auxiliary requests under lack of clarity.


VII. At the oral proceedings which took place on 17 March 2006, the appellant filed five new sets of claims, respectively as main and 1st to 4th auxiliary requests to replace all the previous requests.

Claim 1 of the main request is identical to granted claim 1 and reads as follows:

"1. A dense, self-sintered silicon carbide/carbon-graphite composite material comprising:
(a) silicon carbide,
(b) between 2 and 30 percent by total material weight of carbon-graphite, and
(c) between 0.1 and 15 percent by total material weight of sintering aid,
the silicon carbide having an average grain size between 2 and 15 μm and the graphite having an average grain size between 10 and 75 μm, the average grain size of the graphite being greater than the average grain size of the silicon carbide, the composite material having a density of at least 80 percent of theoretical
as determined by the rule of mixtures for a composite material."

Claim 1 of the 1st auxiliary request is identical to claim 1 of the main request.

Claim 1 of the 2nd auxiliary request differs from claim 1 of the main request by the density being amended to "at least 90 percent of theoretical" instead of "at least 80 percent of theoretical" in the main request.

Claim 1 of the 3rd auxiliary request is identical to claim 1 of the 2nd auxiliary request.

Claim 1 of the 4th auxiliary request differs from claim 1 of the 3rd auxiliary request in that the feature "and a grain size less than 75 \( \mu m \)" is inserted between "10 and 75 \( \mu m \)" and "the average grain size".

VIII. The appellant essentially argued as follows:

Ell, which was filed in response to the board's communication, shows in an analogous technical field that the most porous region in a composite material comprising particles dispersed in a matrix is at the interface between the particles and the matrix and that coarse particles permit more easily the formation of pathways in the material than smaller particles. Ell should therefore be admitted in the appeal proceedings.

The term "carbon-graphite" means graphite particles bonded by carbon formed from a carbon-containing precursor. Although the US document mentioned in
paragraph [0007] of the patent in suit is considered as the closest prior art, E1 can be taken as the starting point for inventive step considerations. The object of the invention is to improve the sealing properties by providing an impervious material. The claimed silicon carbide/carbon-graphite composite exhibits the imperviousness and surface properties necessary for sealing applications as a consequence of the selection of grain sizes for silicon carbide and graphite. In contrast to the patent in suit, E1 aims at producing composites suitable as bearings and having high strength and lubrication capacity. In E1, best lubrication capacity is obtained with 10 to 30% by volume of a granular graphite powder having a particle size in the range of from about 63 to 250 μm (60 to 250 mesh); on the other hand, inadequate self-lubrication is observed with graphite loadings below 10% by volume or with graphite particle sizes below 63 μm. Figure 2 of E1 reveals that the effect of graphite grain size on density is negligible at low graphite loadings of 5% and 10 vol. %, however at high loadings the larger the amount of graphite present, the greater the effect of particle size on density. Where there is an effect of particle size on density, it is that as the graphite particle size increases, so does the density. Thus starting from E1 and assuming porosity to be correlated with density, the skilled person seeking to lower the porosity of the materials of E1 would move to larger particle sizes. There is nothing in E1 teaching how to provide an impervious material suitable for sealing applications. The grain size of the silicon carbide of E1 after sintering is above the upper value of the range claimed because on the one hand the starting material is a β-SiC and on the
other hand the silicon carbide is granulated to a powder having particles below 120 mesh (< 125 μm) before sintering.

E4, which does not teach the use of carbon-graphite, cannot be combined with E1.

Any conclusion as to the grain size of silicon carbide in a self-sintered silicon carbide/carbon-graphite of the type disclosed in the patent in suit using the teaching of either E2, E5 or E9 would be speculative, because these documents disclose the influence of different parameters on grain growth of silicon carbide during sintering without however making use of any graphite inclusions.

Graph 1 reveals that as the graphite grain size increases in a composite made according to the patent in suit, so does the porosity. Figure 2 of E1 indicates the opposite effect of grain size on density. Thus the patent shows an effect which would not have been predicted from E1. Graph 2 shows that low seal wear is achieved with the materials of the invention down to very low graphite loadings with a rapid increase in wear towards zero loading.

The limitation to a density of at least 90% in auxiliary request 2 does not change the argumentation, but represents a tightening of the density range in which a water absorption of zero can be observed as evidenced by Figure 2 of the patent in suit. E1 gives no indication that the rejected materials of Figure 2 of E1 have any use whatsoever and would give an impervious structure.
Auxiliary request 4 was filed to make clear that the composite of test bake 15 does not belong to the invention since it has a carbon-graphite grain size above 75 μm. The question whether or not test bake 15 was outside the claimed range of graphite particle size having been raised for the first time during the oral proceedings, the 4th auxiliary request was filed in reply to this situation.

The appellant indicated that he no longer relied on the photograph attached to the letter of 23 February 2006 in reply to the respondent's objection concerning the lack of information about this photograph.

IX. The respondent (opponent) principally argued as follows:

E11 and the photograph have been filed late. E11 belongs to a different technical field and is not relevant. Regarding the photograph, no data was given concerning the test and the material used, so that the respondent did not have the possibility to verify the conclusions drawn therefrom. Thus they should not be admitted in the proceedings.

The novelty objection raised in writing is not maintained.

Neither the claims nor the patent in suit are limited to the use of the silicon carbide/carbon-graphite composites as seals. In the patent in suit, such a use is disclosed among others, such as bearings, pump seals, etc. The water absorption values of the composites prepared in Test bakes 12 and 15
(respectively 3.87% and 2.56%) show that the problem of obtaining an impervious product is not solved over the whole ambit of claim 1.

Graph 1 establishes that a composite comprising 15% of carbon-graphite having a particle size of 60 to 75 μm has substantial porosity. Graph 2 shows that with 2% wt. of graphite, substantial seal wear - which means high friction - occurs.

It is clear from E1, Figure 3 that all the silicon carbide/ carbon-graphite composites have certain self-lubricating properties. Starting from E1, the problem solved is the provision of an alternative self-sintered composite having lubricating capability and comparable porosity.

E4 discloses self-sintered silicon carbide/graphite composites which can be used as sealing material. Example 9 and Table 11 of E4 furthermore disclose composites containing 0.5 to 10% volume of graphite and having a density greater than 92% of theoretical density. In such high-density materials, the residual porosity is inevitably a closed porosity. The average size of the graphite particle is 1-50 μm, in particular 50 μm. The claimed SiC average particle size is usual in the prior art (see E2 and E5). The subject-matter claimed is obvious from E1 in combination with E4.

The 4th auxiliary request should not be admitted in the proceedings because it has been filed late and as a consequence of an ambiguity concerning Test bake 15 raised by the appellant itself. The request is also prima facie not allowable, since it is doubtful whether
the amendments satisfy the requirements of Article 123(2). The subject-matter of claim 1 of this request also does not meet the requirements of 84 EPC, because there is no method indicated in the patent in suit as regards the measurement of the absolute grain size of carbon-graphite. Furthermore, there is a discrepancy in claim 1, because it is impossible to have simultaneously an average grain size of 75 \( \mu m \) and a particle size less than 75 \( \mu m \).

X. The appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of the main request filed on 17 March 2006 or alternatively, on the basis of the auxiliary requests 1 to 4, also filed on 17 March 2006.

The respondent requested that neither document E11 nor auxiliary request 4 be admitted into the proceedings and that the appeal be dismissed.

Reasons for the Decision

1. **Admissibility of late filed document E11**

The appellant argued at the oral proceedings that E11 was filed in answer to the board's communication to show, from the teaching of an analogous technical field, that the most porous region in a composite material comprising particles dispersed in a matrix is at the interface between the particles and the matrix and that coarse particles more easily permit the formation of pathways in such a material than small particles.
The board observes that E11 was cited for the first time three weeks before the oral proceedings without indication of any of the passages relevant to support the appellant's arguments. This document, which was published after the priority date, examines the impact of particle size range and cumulative distribution on the permeability and drying behaviour of high-alumina, ultra-low-cement refractory castable compositions (see page 9401, 4th and 5th paragraphs). For the study of permeability behaviour that was considered as relevant by the appellant at the oral proceedings, the castable compositions were formulated with 99 wt.% of different aluminas and 1 wt.% of calcium aluminate cement (see page 9402 "Castable Design"). Therefore E11 concerns a completely different kind of composite which neither contains silicon carbide, nor graphite, let alone carbon-graphite. Furthermore, the permeability tests were conducted on specimens of green bodies which had been cured for 24 h at 25 °C and about 100 % humidity and then dried at 110 °C for 24 h (see page 9403, left-hand column). As pointed out by the respondent, these permeability results cannot be compared with those obtained with sintered silicon carbide/carbon-graphite composites that have been sintered at temperatures above 2000 °C, because of the matrix shrinkage at such temperatures. For the preceding reasons, the board considers that this late filed document is not relevant and would not change the outcome of the present decision. Therefore E11 is not introduced into the appeal proceedings (Article 114(2) EPC).
2. **Main and 1st to 3rd auxiliary requests: allowability under Article 123(2) EPC - Novelty**

No objection under Article 123(2)(3) EPC has been raised against any of the amended claims of the main and 1st to 3rd auxiliary requests and novelty was no longer contested at the oral proceedings. The board also considering that the requirements of Article 123(2)(3) EPC and of novelty are met, no further comments on these matters are needed.

3. **Main and 1st auxiliary requests - Inventive step**

Claim 1 of the main request and claim 1 of the 1st auxiliary request being identical, the arguments below apply to both requests.

3.1 **E1**, also considered as the closest prior art by the respondent and the opposition division, discloses silicon carbide-graphite self-lubricating ceramic composites, defined in claim 1 of E1 as having a matrix of silicon carbide and containing from 10 to 30 % by volume of a granular graphite powder with particle diameters in the range of from 60 to 250 mesh, i.e. about 250 to 63 μm. In the Examples (see pages 8 and 9), on the one hand a granular silicon carbide is prepared by wet mixing 100 parts by weight of β-silicon carbide powder having a particle size of 0.5 μm with 0.4 parts by weight of boron carbide as a sintering aid and 4 parts by weight of a phenolic resin as a binder in the presence of ethanol, drying said mixture and powdering the dried material, thus obtaining a granular silicon carbide powder with particle sizes below 120 mesh (< 125 μm). On the other hand, 100 parts by
weight of graphite having a primary particle size of 5 to 8 μm are compounded with 0.4 parts by weight of boron carbide as a sintering aid and 15 parts by weight of a phenolic resin as a binder, said components being then wet mixed in a ball mill in the presence of ethanol. This mixture is then dried, powdered and the powder is graded into 5 sieve fractions: 16-32 mesh (500 μm-1 mm), 32-60 mesh (250-500 μm), 60-120 mesh (125-250 μm), 120-250 mesh (63-125 μm) and > 250 mesh (< 63 μm). Silicon carbide-graphite composites are then manufactured by dry-mixing different amounts (0%, 5%, 10%, 20%, 40% by volume) of each of said graded fraction of granular graphite powder with said granular silicon carbide powder and pressure-moulding the mixture. The moulded materials are next sintered at 2180°C for 45 minutes to give self-sintered lubricating silicon carbide/graphite composites. It has not been contested that this process leads to a self-sintered silicon carbide/carbon/graphite composite material comprising particles of graphite coated with carbon, i.e. "carbon-graphite" particles.

Figure 2 of E1 shows that the self-sintered composites containing up to 30% vol. (i.e. ~ 23.1% wt.) of carbon-graphite have a relative density at least 80% of the theoretical density and those comprising 5% vol. (~ 3.5% wt.) or less of carbon-graphite have a relative density above 90% of the theoretical density. The average grain sizes of silicon carbide and carbon-graphite in said self-sintered composites are not directly and unambiguously derivable from E1.
3.2 The subject-matter of claim 1 of the main and 1st auxiliary requests is thus distinguished from E1 in that:

i) the average grain size of silicon carbide in the sintered composite is between 2 and 15 μm;

ii) the average grain size of the carbon-graphite in the sintered composite is between 10 and 75 μm;

iii) the average grain size of graphite is greater than the average grain size of the silicon carbide.

3.3 The patent in suit, starting from US-A-4525461, established the problem to be solved as the provision of a dense, impervious self-sintered silicon carbide body incorporating larger amounts of graphite to increase lubricating capability while maintaining the integrity of the microstructure, i.e. relative absence of cracks and porosity (paragraphs [0008], [0009] and [0013]). During the proceedings in writing, E1 was nevertheless considered as the closest prior art; furthermore there is no mention of the said US patent in the grounds of appeal and during the oral proceedings, the appellant took E1 as the starting point to define the problem to be solved. The question thus arises which technical problem is actually solved by the subject-matter of claim 1 of the main and 1st auxiliary requests starting from E1.

As indicated in item 3.1 supra, E1 discloses dense self-sintered silicon carbide/carbon-graphite composites incorporating large amounts of graphite. Although E1 is silent about porosity, it can be assumed that composites which have been sintered at 2180 °C and have a relative density higher than 90% of the
theoretical density, have low or even no open porosity, as pointed out by the respondent at the oral proceedings and not contested by the appellant. Concerning the "relative absence of cracks or porosity" presented as an advantage in the patent in suit (column 3, lines 33-35), nothing attests either in the file, or in the patent specification, that the composites according to claim 1 of the present requests give rise to any improvement over those of E1. Graph 1 of the grounds of appeal shows that for a particular graphite average particle size, silicon carbide/carbon-graphite composites according to the patent in suit and containing 15 wt.% of carbon-graphite have less porosity than the same composite containing 15 wt.% of free graphite instead of carbon-graphite; Graph 1 further shows that in silicon carbide/carbon-graphite composites according to the patent in suit, porosity decreases with the average particle size of graphite and is very low with a carbon-graphite average particle size of less than 40 μm. Graph 1 however neither indicates any porosity value for composites with graphite loadings < 15% by weight, nor does it show any comparative data with the composites of E1. Accordingly, the composites of claim 1 of the main and 1st auxiliary requests cannot be considered less porous than those of E1, in particular for low carbon-graphite loadings.

Figure 3 of E1 shows that all the exemplified sintered silicon carbide/carbon-graphite composites, i.e. up to an amount of 40 vol.% graphite in the composite, have certain lubricating properties. Since no improvement was shown as regards the composites of the patent in suit over those of E1, the alleged "increased
lubricating capability" stated in the patent in suit (column 2, lines 24-29 or column 3, lines 32-33) cannot be taken into consideration. The appellant's argument that better self-lubrication at low graphite loadings was provided is also not convincing, because Graph 2 of the grounds of appeal shows that seal wear (which according to the appellant is a better measure of self-lubrication properties than the coefficient of friction) rapidly increases with low graphite loadings. In particular with graphite loadings of 2 wt.% or slightly above 2 wt.% - which fall within the scope of claim 1 of the present requests - seal wear is much higher than with a graphite loading of 4 wt.%.

Since Graph 2 furthermore does not show any comparative data with the composites of E1, it also does not constitute evidence for an improvement as regards the lubricating capability of the composites of the patent in suit over those of E1.

At the oral proceedings, the appellant starting from E1 defined the problem as improving the sealing properties of silicon carbide/graphite composites by providing an impervious material. The board can however not start from this problem for the assessment of inventive step since it has not been shown that the sealing properties (such as seal wear or imperviousness) are improved over those of the composites disclosed in E1. Regarding the seal wear, Graph 2 does not show that it is improved with respect to the composites of E1, in particular at low loadings of carbon-graphite (about 2 wt.%) as already indicated above. Concerning imperviousness, Figure 2 of the patent in suit shows that in the Test bakes 9-18, which are not presented as comparative (contrary to Test bakes 1-8), some of the composites
have a water absorption of zero or close to zero: see Test bakes 9, 11, 16, 17 and 18, whereas in other Examples, the composites have a substantial water absorption (see Test bakes 12 and 15) or a non-negligible water absorption of about 1% (see Test bakes 10 and 14). In view of the "graphite size" given in Figure 2 for Test bake 14 (-200, +325 mesh; i.e. ~ 45-75 μm), this example clearly falls within the definition of claim 1 of present main and 1st auxiliary requests. Concerning Test bake 15, it cannot be clearly derived from the size indicated in Figure 2 (-100, +325 mesh; i.e. ~ 45-150 μm) or from the data given in paragraph [0027] of the patent in suit concerning the carbon-graphite particle size whether this example lies outside the claimed range of average grain size (10-75 μm) or not. The board observes however that in the amended documents considered by the opposition division as meeting the requirements of the EPC, Test bake 15 was expressly regarded by the appellant as an example according to the invention (i.e. having an average grain size of between 10 to 75 μm), however with a water absorption of 2.36%, it cannot be considered as impervious to water. In any case, it cannot be deduced from the data in Figure 2 that the imperviousness of the claimed composite is improved with respect to those of E1, in particular those containing about 5 vol.% (~ 3.5 wt.%) carbon-graphite and having a relative density of at least 90%.

3.4 Accordingly, starting from E1, the technical problem underlying the subject-matter of claim 1 of the present requests can be seen in the provision of another dense self-sintered silicon carbide/carbon-graphite composite having lubricating capability and low porosity. In view
of the data of the patent in suit, in particular those of Figure 2, it is credible that this problem has actually been solved by the composites as defined in claim 1.

3.5 The solution as proposed in claim 1 of said requests does not involve an inventive step for the following reasons:

As mentioned above, E1 discloses dense substantially non-porous silicon carbide/carbon-graphite composites having certain lubricating capability and high density. From Figures 2 and 3 of E1, it can be inferred that for those composites comprising e.g. 5 vol.% (~ 3.5 wt.%) or less of carbon-graphite, the relative density is not substantially influenced by the graphite grain size and is of at least 90%, whatever the chosen grain size.

The appellant argued that although the effect of the graphite grain size on density was negligible at a graphite loading of in particular 5 vol.% (~ 3.5 wt.%), the skilled person would nevertheless be discouraged from selecting the graphite grain size in the range < 63 μm among the five ranges disclosed in Figures 1-3 of E1, because - as shown by Figure 3 of E1 - the lowest coefficient of friction is observed at a carbon-graphite loading of from 10 to 30 vol.% and with a carbon-graphite grain size in the range 60 to 250 mesh (63-250 μm).

The board observes that claim 1 of the main and 1st auxiliary requests is not limited to composites having a low coefficient of friction. In other words, the lubricating properties may be low and even lower
than those illustrated for carbon-graphite grain sizes of 63 to 250 μm in Figure 3 of E1. This is reflected in the formulation of the technical problem stated in point 3.4 supra, where only "lubricating capability" is required.

It is true that according to Figure 3 of E1, the coefficient of friction is the lowest with a carbon-graphite grain size of from 63 to 250 μm and with a carbon-graphite loading of from 10 to 30 vol.%, while a carbon-graphite grain size below 63 μm leads to a higher coefficient of friction. However, it cannot be inferred from Figure 3 that the resulting composite would have no lubricating capability. For a graphite loading of e.g. 5 vol.%, the coefficient of friction does not differ substantially with graphite particle sizes varying within the ranges 125-250 μm, 125-63 μm, < 63 μm (corresponding to 60-120 mesh, 120-250 mesh, > 250 mesh respectively). Accordingly, the skilled person faced with the problem of providing another dense self-sintered SiC/carbon-graphite composite having lubricating capability and low porosity would not disregard E1 since he can infer from both Figure 2 and Figure 3 of E1 that a composite with a high relative density of above 90%, and thus a low open porosity, and still having lubricating capability can be obtained with a graphite loading of about 5 vol.%. It is noted furthermore that not only E1 was part of the prior art before the priority date, but also E4.

3.6 E4 (claims; page 2, line 54-page 3, line 9) discloses self-sintered composites having low porosity and a density preferably not less than 90% of the theoretical density, said composites containing silicon carbide and
from 1 to 20% vol. (with respect to the volume of SiC) of a lubricating substance having an average grain size preferably up to 50 μm (most preferably 1-50 μm), said lubricating substance being selected from boron nitride, graphite, soot or mixtures thereof. At page 3, lines 18-20 of E4 it is indicated that said composites have inter alia sealing and sliding properties. In Example 9 of E4, a powder of β-silicon carbide (average particle size 0.3 μm) is mixed with 0.5 wt.% boron carbide, 6 wt.% of phenolic resin and different amounts of graphite having an average grain size of 50 μm (see Examples 9 and 4). The mixture thus obtained is wet mixed with water, dried, sieved and formed into rings, these being then calcined and sintered at 2050 °C under normal pressure. Among the materials thus prepared, a density above 90% of theoretical density is measured in the composites having a graphite content of up to 10 vol.% and the lowest friction coefficient and wear values are observed for those having a graphite loading of 5 vol.% and 10 vol.% (with respect to the volume of SiC). Accordingly, among the silicon carbide-graphite composites prepared in E4, very good results regarding lubricating ability and relative density are obtained with a graphite average grain size of 50 μm and a graphite loading of either 5 vol.% or 10 vol.% (with respect to the volume of SiC). In view of this teaching, the skilled person starting from E1 and faced with the problem stated above would contemplate trying either graphite or carbon-graphite inclusions having average particle sizes of about 50 μm in amounts of about 5 vol.% up to 10 vol.% in the composite of E1 in order to obtain another composite having lubricating capability and low porosity and would thus also investigate the range < 63 μm (> 250 mesh) disclosed in
E1. Doing so, he would arrive by routine experimentation at a composite with an average grain size of the carbon-graphite particles falling within the claimed range. In this context it is observed that, as confirmed by the respondent at the oral proceedings and not contested by the appellant, the graphite particle size is not substantially changed during the sintering step.

Although, as pointed out by the appellant, E4 makes use of graphite grains instead of carbon-graphite, both documents E1 and E4 concern the same type of composite and thus obviously belong to the same technical field. Furthermore, these documents contain no information which might deter the skilled person from combining their teachings.

3.7 E1 being silent as to the grain size of the silicon carbide in the sintered composites, it remains to be examined whether features i) and iii) identified in point 3.2 supra may render the claimed composite inventive in combination with the remaining features thereof.

As to feature i), the appellant argued that in the self-sintered composites of E1, the average grain size of the silicon carbide would be above the upper value of the range defined in claim 1 of the present requests (i.e. above 15 μm), because before sintering, the silicon carbide is granulated to a powder having particles below 120 mesh (< 125 μm) to prevent mixing of finely powdered graphite with individual silicon carbide particles. Concerning said granulating operation, the board observes that E1 discloses in the
paragraph bridging pages 6 and 7 that, when the granules of silicon carbide powder are mixed with the graphite as the solid lubricant, mixing of finely powdered graphite with individual silicon carbide granules is prevented, so that the sintering of the silicon carbide will continue unchecked. This means that sintering of the individual silicon carbide particles in the silicon carbide granules takes place without the sintering operation being inhibited by the graphite particles. In view of this teaching, the skilled person would thus expect that at least at low carbon-graphite loadings, the silicon carbide particles used as starting material in E1 will after sintering have the particle sizes normally obtained by self-sintering silicon carbide under comparable sintering conditions.

As to the normal particle sizes of a sintered silicon carbide, the appellant stated that any conclusion drawn from E2, E5 or E9 would be speculative, because the sintering studies carried out in these documents are made on silicon carbide samples having no graphite inclusions, i.e. a situation different from that in E1, wherein carbon-graphite is admixed with the silicon carbide. The board is not convinced by this argument because as mentioned in the previous paragraph, the granulating operation in E1 prevents the mixing of finely powdered graphite with individual silicon carbide granules and thus avoids any inhibition of the sintering of the silicon carbide by graphite particles. Furthermore, at low carbon-graphite loadings, for instance at a loading of about 5 vol.% (~ 3.5 wt.%) or lower, the skilled person would not expect the carbon-
graphite having a substantial influence on the grain growth of silicon carbide during sintering.

3.8 E2 (see page 32, item headed "2. Experimental"; Figure 2; page 33, middle of the right column starting with "Fig. 2 …) discloses in particular that at 2040 °C for 30 minutes in the presence of 0.5% B₄C and 4% C (derived from a phenolformaldehyde resin), α-SiC with a particle size of 0.64 μm sinters to grains having an average intercept length of 2.45 μm. At sintering temperatures of 2100 °C and 2200 °C, values of 4.28 μm and 14.78 μm are respectively measured.

In E5, silicon carbide samples having average grain sizes of 12 μm and 7 μm have been obtained after sintering at 2080 °C for 30 minutes of an α-SiC starting material having an average particle size of respectively 1.7 μm and 0.77 μm in the presence of 4% C and 0.5% B as B₄C (see page 217, table I and page 218, "Role of Powder Surface Area").

E9 (pages 1022, "II. Experimental procedure"; page 1023 "Experimental procedure"; page 1026, lines 1-5) shows that the average grain size (measured by mean intercept length method) of a β-SiC (starting grain size undisclosed) sintered at 2150 °C for 1 hour in the presence of 0.26 wt.% boron remains constant at 2 μm during creep experiments carried out on the sintered body.

Thus these documents show that an average grain size of from 2 to 15 μm is not unusual for silicon carbide self-sintered at temperatures between 2040 and 2200 °C and
in the presence of the known sintering additives, such as boron carbide and carbon, used either in E4 or E1.

For the preceding reasons, feature i) is considered to be obvious to the skilled person faced with the technical problem stated above.

Furthermore, it follows from the preceding considerations (points 3.6 and 3.7) that a self-sintered composite resulting from the combination of the teachings of E1 and E4 as contemplated in point 3.6 comprises carbon-graphite grains having an average grain size of about 50 μm and silicon carbide grains with an average grain size which the skilled person would expect to be well below 50 μm under the sintering conditions used in E1. Therefore feature iii), i.e. that the average grain size of the graphite be greater than the average grain size of the silicon carbide, would be fulfilled in such a composite. Therefore feature iii) can also not render the claimed subject-matter inventive in view of the cited prior art.

3.9 In conclusion, for all the reasons indicated above, the subject-matter of claim 1 of the present requests is obvious in the light of the cited documents for the skilled person faced with the problem identified in item 3.4 supra and thus lacks an inventive step. The main request and the 1st auxiliary request are therefore rejected.

4. 2nd and 3rd auxiliary requests - Inventive step

Claim 1 of the 2nd and 3rd auxiliary requests being identical, these requests will be treated together.
As already indicated above, E1 also discloses composite material having a relative density of at least 90%. The appellant stated that the limitation to a density of at least 90 % of the theoretical density does not change the argumentation used for the previous requests, but represents a tightening of the density range in which - as evidenced by Figure 2 of the patent in suit - a water absorption of zero can be observed. Concerning the embodiments which in Figure 2 have a density of at least 90% of the theoretical density (namely Test bakes 9 to 11 and 15 to 18), the board observes that two composites do not have a zero water absorption, namely those of Test bakes 10 and 15, which have a water absorption of respectively 1.18% and 2.36%. As mentioned in item 3.3 supra, it cannot be clearly derived from the patent in suit whether Test bake 15 lies outside the claimed range of average grain sizes (10-75 μm) or not. However, in any case, Test bake 10 does not have a water absorption of zero. Furthermore there is no evidence that the imperviousness of the composites of claim 1 of these requests is improved over those having a relative density of at least 90 % in E1. Under these circumstances, the technical problem solved by the subject-matter of claim 1 of the present requests is the same as for claim 1 of the main and 1st auxiliary requests (see point 3.4 above) and the considerations in items 3.5 to 3.9 supra also apply mutatis mutandis to claim 1 of the present requests. These requests are therefore rejected for lack of inventive step of the subject-matter of claim 1.
5. **Fourth auxiliary request - Admissibility**

The appellant explained that this request was filed to make clear that the composite of Test bake 15 would not belong to the invention, since it includes graphite particles having a size above 75 $\mu$m. It also argued that claim 1 thus amended would be _prima facie_ patentable since lamination did not occur when carbon-graphite had such a size; therefore the request should be admissible.

The board notes that the present request was not filed in response to an objection raised either by the board or by the respondent, but was the consequence of an ambiguity engendered by the appellant's representative himself who stated during the oral proceedings that the embodiment identified Test bake 15 in Figure 2 would have an average graphite grain size falling outside the claimed range of 10 to 75 $\mu$m.

As regards the feature introduced into claim 1 of this request, namely that the grain size of the graphite be less than 75 $\mu$m in the sintered composite, it appears to be disclosed in the description of the patent in suit (column 7, lines 15-18). However this feature has no counterpart in the granted claims directed to the sintered composite and its insertion into independent claim 1 could not be expected by the respondent who was taken by surprise, all the more so as the question whether or not Test bake 15 is an embodiment according to the invention was raised for the first time by the appellant during the oral proceedings. Under these circumstances, the acceptance of such an amendment at such a late stage of the proceedings would have
deprived the respondent of an opportunity of dealing properly with this request which raised new issues. In particular, as pointed out by the respondent at the oral proceedings, the amended claim contains both an upper limit of 75 μm for the average grain size of the graphite and a graphite grain size < 75 μm, which features would appear not to be compatible with each other. Therefore, amended claim 1 of this request raised the new issue of whether it meets the requirements of clarity set out in Article 84 EPC. Under these particular circumstances and considering that such a request could have been filed earlier, the board in the exercise of its discretion pursuant to Article 10b(1) of the Rules of Procedure of the Boards of Appeal (EPO, OJ 2003, 93, RPBA apply to all appeals filed after 1 May 2003) has decided not to admit the 4th request filed shortly before the end of the oral proceedings into the appeal procedure.
Order

For these reasons it is decided that:

- Document E11 is not admitted in the proceedings.

- Auxiliary request 4 is not admitted in the proceedings.

- The appeal is dismissed.

The Registrar:      The Chairman:

G. Rauh        M. Eberhard