

Publication in the Official Journal ~~Yes~~ / No

File Number: T 436/88 - 3.3.2

Application No.: 83 104 855.8

Publication No.: 0 095 130

Title of invention: Coated composite modified silicon aluminium oxynitride
cutting tools

Classification: CO4B 41/87

D E C I S I O N
of 20 February 1992

Proprietor of the patent: GTE Laboratories Incorporated

Opponent: Fried. Krupp GmbH

Headword: Sialons/GTE

EPC Article 56

Keyword: "Inventive step (confirmed)"

Headnote



Case Number : T 436/88 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 20 February 1992

Appellant :
(Opponent)

Fried. Krupp GmbH
Altendorfer Str. 103
Postfach 10 22 52
W-4300 Essen 1 (DE)

Representative :

Vomberg, Friedhelm, Dipl.-Phys.
Schulstrasse 8
W-5650 Solingen 1 (DE)

Respondent :
(Proprietor of the patent)

GTE Laboratories Incorporated
100 W. 10th Street
Wilmington
Delaware (US)

Representative :

Patentanwälte Grünecker, Kinkeldey,
Stockmair & Partner
Maximilianstrasse 58
W-8000 München 22 (DE)

Other Party :
(Opponent)

Sandvik Aktiebolag
Fack
S-811 81 Sandviken 1 (SE)

Decision under appeal :

Decision of Opposition Division of the European
Patent Office dated 8 July 1988 rejecting the
opposition filed against European patent
No. 0 095 130 pursuant to Article 102(2) EPC.

Composition of the Board :

Chairman : P.A.M. Lançon
Members : M.M. Eberhard
F. Benussi

Summary of Facts and Submissions

- I. European patent No. 0 095 130 based on application No. 83 104 855.8 was granted on the basis of eighteen claims. Claim 1 reads as follows:

"1. An abrasion resistant composite ceramic cutting tool comprising a substrate body consisting essentially of a particulate material selected from the group consisting of the refractory metal carbides, nitrides, carbonitrides and mixtures thereof uniformly distributed in a matrix comprising a modified silicon aluminium oxynitride, said particulate material comprising from 5 volume percent to 60 volume percent of said cutting tool and having an average particle size of between 0.5 and 20 microns μm , characterised in that at least one adherent refractory material coating layer is provided and in that said modified silicon aluminum oxynitride comprises a modifier selected from the group consisting of the oxides of silicon, yttrium, magnesium, hafnium, zirconium, beryllium, the lanthanides and combinations thereof, wherein said modified silicon aluminium oxynitride is formed from 2 to 25 volume percent of said modifier, from 20 to 90 volume percent silicon nitride and from 5 to 60 volume percent aluminium oxide.

- II. The Appellant filed a notice of opposition requesting revocation of the patent on the ground of lack of inventive step with respect to document (1), DE-A-2 443 160, and two other documents. During the opposition proceeding the Appellant further referred to

- (4) American Machinist, May 1981, page 167
(5) JP-A-5 632 377

- (6) SU-A-537 986
- (7) JP-A-56 16665
- (8) JP-A-57 17466

III. The Opposition Division rejected the opposition, taking the view that the only relevant document cited in due time with respect to inventive step was (1) and that the invention did not reside in the presence of the coating layer but in the use of a particular substrate body. According to the decision, there was no indication in (1) that impact strength and fracture toughness of a cutting tool could be improved by using a silicon aluminium oxynitride based substrate body instead of the known alumina based bodies.

IV. The Appellant lodged an appeal against this decision. Oral proceedings were held on 20 February 1992. At the appeal stage, the Appellant sought to introduce the following documents into the procedure:

- (9) EP-A-0 035 777
- (10) VI.intern.Pulvermet.Tagung Dresden (1977),
page 45-1.
- (11) Mechanical properties of silicon nitride composite materials, "Metallurgiya", no. 7 (223) (July 1981)
pages 502-505 of the translation.
- (12) Optimisation of properties of a tool material based on silicon nitride, "Metallurgiya", no. 2 (182),
pages 124-127 of the translation, February 1978.
- (13) K.H. Jack, Journal of Materials Science 11 (1976),
pages 1135-1158, "Review Sialons and related nitrogen ceramics".
- (14) US-A-4 252 768

The Appellant's arguments may be summarised as follows:

The cutting tool according to Claim 1 of the patent in suit differed from that of (9) merely by the presence of at least one adherent refractory material coating layer and by the matrix being formed from 5 vol% to 60 vol% aluminium oxide. Taking into account the usual practice since the middle of the 60's and the disclosure of (1) the provision of an adherent refractory material coating layer on a substrate body for a cutting tool had to be regarded as the customary practice followed by a person skilled in the art seeking to improve the wear resistance of a cutting tool. It was also apparent from documents (7) and (8) that especially silicon nitride based sintered bodies coated with e.g. titanium carbide had superior properties.

Furthermore, a sialon was also formed in the matrix according to (9) in the presence of 5 wt% aluminium oxide. Thus, the claimed matrix material overlapped that described in (9) despite the quotation in vol%. In particular, it was not possible to distinguish between alumina acting as densification aid and alumina (more precisely the Al and O atoms thereof) substituting Si and N in the Si_3N_4 -lattice. Therefore, a simple combination of document (9) with (7) or (8) would have led to the subject-matter of Claim 1 at least at the limit of 5% aluminium oxide.

Having regard to documents (5) and (6), in particular to (6) which disclosed substrate bodies with up to 15 wt% aluminium oxide, there was no prejudice against the use of more than 5 vol% aluminium oxide to form the matrix. Furthermore, it was clear that a matrix formed from the starting materials according to (5) and (6) would

inevitably comprise a sialon. As regards the method of preparation of the sialon, Claim 1 would not exclusively relate to a prereacting step leading to a specific sialon matrix. There was also no doubt that the skilled person got a clear teaching from (4) that a sialon containing substrate could be formed from silicon nitride, aluminium oxide and titanium carbide. This prior art indeed did not mention the claimed modifiers; however, it was well-known that they belong to the group of densification aids. Therefore, the skilled person would in any case have included a densification aid in the starting material for the matrix of a cutting tool. In the light of the general teaching in (13) about the continuous substitution of Al and O for Si and N in the β - Si_3N_4 lattice to form a β' -sialon, i.e. a material having an expanded β - Si_3N_4 lattice, the lower limit of 5 wt% alumina appeared not to exist. Moreover, (13) also disclosed the use of densification aids and complex structures based on yttrium-sialon systems. Determination of the proportions of the components forming the composite material was merely a matter of routine experimentation, in particular since document (12) disclosed broad ranges of titanium carbide and alumina in a composite material based on silicon nitride.

- V. The Respondent has strongly contested that the matrix according to (9) might contain a silicon aluminium oxynitride. The amount of less than 5% by weight alumina according to (9) would correspond to a much lower value expressed in volume percent. Accordingly, all of the alumina added to the matrix would appear in a glassy intergranular phase between the silicon nitride grains. According to the patent in suit a homogeneous one phase matrix was formed in contrast to the two phase matrix of (9). For those skilled in the art reading (9) there was furthermore a clear prejudice against the use of more than

5 wt% of alumina since this document indicated to minimize the presence of any additive such as alumina.

The Respondent has further stressed that although (6) did describe compositions formed from starting mixtures comprising much more than 5 vol% alumina, it taught neither that a sialon was obtained nor how to form a specific modified sialon matrix. According to the invention, it was the prereacting step and not a simple mixing procedure which led to a specific matrix having a homogeneous structure. There was no doubt that the wording of Claim 1 would be construed by those skilled in the art as including a prereacting step as an essential feature of the preparation method for the substrate. Following a discussion about the meaning of the terms "formed from" in view of the examples of the patent, an auxiliary request has been submitted during the oral proceedings.

The Respondent has further argued that starting from the silicon nitride based matrix described in (9) there was no reason for a person skilled in the art to take into account documents (4) and (13). These documents relating to sialon materials in general did not disclose any concrete percentage ranges for the components of a ceramic composite material suitable as a cutting tool. It has been emphasised that although documents (1), (7) and (8) indeed described ceramic cutting tools with a coating layer, the cutting tools comprised substrate bodies not comparable to the claimed specific substrate body. The unique combination of the coating with this new composite substrate was not suggested by the cited prior art.

VI. The Appellant requested that the appealed decision be set aside and that the patent be revoked.

The Respondent requested that the appeal be dismissed and that the patent be maintained in the granted form as main request, or with the following amendments as auxiliary request:

- (a) In Claim 1 the phrase "characterised in that" is replaced by "wherein";
- (b) Examples 1, 2 and 3 are cancelled and examples 4, 5 and 6 are renumbered correspondingly.

Reasons for the Decision

1. The appeal is admissible.
2. During oral proceedings the Respondent argued that Claim 1 included the step of prereacting silicon nitride, aluminium oxide and the modifier to form the matrix. This was contested by the Appellant who was of the opinion that the wording of Claim 1 covered both alternatives described in the examples. As inventive step cannot be assessed without having previously clearly set out which combination of features is claimed, this question has to be examined first.

According to Claim 1 the substrate body consists essentially of the particulate refractory material distributed in a matrix comprising a modified silicon aluminium oxynitride which is formed from 2-25 vol% of the modifier, from 20-90 vol% Si_3N_4 and from 5-60 vol% Al_2O_3 . In the Board's opinion the expression "is formed from" is broad and may be construed as meaning that the starting components used to form this oxynitride are alumina, silicon nitride and the modifier, the amounts thereof being as stated above. Accordingly, the particulate refractory material (for example TiC) may be added either

before or after reaction of these starting components. This interpretation is supported by the description which, pursuant to Article 69(1), should be used to interpret the claims. According to examples 1 to 3 of the patent the titanium carbide powder is mixed with the three starting components in form of powders without any prereaction step whereas in examples 4 to 6 the starting components are first prereacted to produce a prereacted modified silicon aluminium oxynitride. All these examples are said to be illustrative of the invention. Under these circumstances, Claim 1 cannot be regarded as being limited to the alternative where the modifier, alumina and silicon nitride are prereacted before addition of the particulate refractory material.

3. After consideration of the late filed documents the Board has decided to disregard (10) and (11) under Article 114(2) EPC. Thus, the short abstract on page 45-1 of (10) does not contain any additional technical information when compared with the disclosure of (6). As regards (11), it neither refers to a silicon aluminium oxynitride matrix nor discloses the presence of aluminium oxide in combination with silicon nitride in the starting material and therefore has no influence upon the outcome of the decision. Document (14), which was cited for the first time during oral proceedings before the Board, is mentioned in the search report and acknowledged in the contested patent, however it is not considered therein as essential or as closest prior art. Therefore, it does not automatically form part of the opposition or appeal proceedings and is also regarded as a late submitted document (cf. T 198/88, OJ EPO, 1991, 254 and T 536/88 to be published). Since (14) describes a sandblasting nozzle, it clearly relates to a far remote technical field which the skilled person in the field of cutting tools would

have no reason to look at. Therefore (14) is also disregarded under Article 114(2).

4. Having regard to the Respondent's submissions during oral proceedings that the matrix of the uncoated composite cutting tool of document (9) does not contain any silicon aluminium oxynitride (termed hereinafter sialon), this document is no longer considered as the closest prior art. Instead, document (8) which discloses coated composite ceramic cutting tools comprising a silicon nitride based substrate body and at least one coating ceramic layer is regarded as a more suitable starting point. The substrate bodies of (8) contain in addition to silicon nitride 1-30 wt% of a hard phase from compounds of C, N or O with IVB, VB and VIB metals, 0-5 wt% of a Fe group metal and 0-5 wt% MgO. The hard refractory material may be for example WC, TiC, TiN, TaC in form of a powder having a particle size of 10 μm or less and the coating layer is preferably a TiC and/or an Al_2O_3 layer. The resulting cutting tools exhibit good cutting performances, in particular a longer life time than the conventional cutting tools, when used for cutting steel at a speed of 200 m/min (cf. Claim 1, page 2, left-hand column, lines 34-38, paragraph bridging the left-hand and the right-hand column; examples 1 to 3).

In the light of this closest prior art, the technical problem underlying the patent can be seen in providing an alternative to this coated composite ceramic cutting tool.

This problem is solved by a cutting tool having a coated substrate body the matrix of which comprises a modified silicon aluminium oxynitride formed from silicon nitride, aluminium oxide and a modifier in the proportions defined in Claim 1, the hard refractory material dispersed in this

matrix being present in an amount of 5-60 vol% (with respect to the substrate body). The Board has no reason to doubt that the coated cutting tools prepared as indicated in the examples of the patent exhibit cutting performances at least comparable to those of the coated cutting tools according to (8). This was also not contested by the Appellant at the oral proceedings. Therefore, the Board is satisfied that the above technical problem has been plausibly solved by the claimed features.

5. After examination of the cited prior art, the Board has reached the conclusion that the claimed subject-matter is novel. Since novelty is not disputed, it is not necessary to consider this matter in details.
6. It remains to examine whether the claimed cutting tool satisfies the requirement of inventive step.
 - 6.1 The starting powder mixture used in (8) contains silicon nitride, the hard refractory material and either MgO or a metal of the iron group or both of them. This document alone does not provide any information which could give the skilled person an incentive to add a certain amount of aluminium oxide to the starting mixture and to treat the blend so as to form a modified silicon aluminium oxynitride.
 - 6.2 Document (9) discloses a composite article containing particles of a hard refractory material such as TiC, uniformly distributed in a matrix which consists essentially of two phases, namely a first phase of silicon nitride and a secondary intergranular phase comprising silicon nitride, a densification aid selected from the group of yttrium oxide, hafnium oxide, zirconium oxide or lanthanides oxides, and optionally additives such as aluminium oxide. The secondary phase may also contain

further additives, for example silicon dioxide. This document teaches that for optimising the high temperature properties of the composite article, it is desirable to minimize the presence of any additive such as aluminium oxide, the aluminium oxide content of the matrix being preferably less than about 5 wt%, more preferably less than about 3 wt% (cf. Claims 1, 2, 11 and 13; page 1, lines 22-30; page 4, lines 12-35). This document further discloses the use of such materials for cutting tools applications. According to Claims 15 and 25 relating to the cutting tool, the secondary phase comprises less than about 4 wt% Al_2O_3 , and less than 5 wt% other additives. None of the exemplified materials contain aluminium oxide in the starting mixture.

The Respondent's arguments that the secondary intergranular phase is a glassy phase was not contested by the Appellant. This seems to be in agreement with the teaching of document (5) (see page 4, lines 3-12 and last paragraph). According to (9) the aluminium oxide is present in the secondary phase of the matrix, i.e. in the glassy phase. Even if this secondary phase were crystalline, this document does not contain any information from which it could be derived that a part of the aluminium reacts with the silicon nitride to form a sialon, for example a β' -sialon. Under these circumstances the Board cannot follow the Appellant's argument that a silicon aluminium oxynitride is formed in the matrix of document (9). As regards the upper limit of 5 wt% for the alumina content of the matrix (4 wt% in the case of the cutting tool), the Appellant's contention that this content would correspond to about 5% by volume and would therefore overlap the claimed range was strongly contested by the Respondent who, for his part, affirmed that the estimated alumina content in volume % would be substantially lower than 5%. As the Appellant's contention

was based upon incorrect calculations, it amounts to an unproven affirmation. In such a situation the Board has to decide in favour of the party not having the burden of proof, i.e. the Respondent, (cf. T 219/83, OJ EPO, 1986, 211), all the more so as the question whether a sialon is formed or not does not only depend upon the amount of Al_2O_3 in the starting mixture but also upon the amount of the other additives, the purity of the silicon nitride, the operating conditions used during sintering or hot-pressing. Therefore, even at the upper limit of 5 wt% alumina a simple combination of the teachings of (9) and (8) would not lead to the claimed subject-matter.

As, on the one hand, (9) does not disclose a sialon containing matrix and, on the other hand, it is recommended to minimize the amount of Al_2O_3 in the matrix in order to avoid a deleterious effect on the high temperature strength and creep resistance of the final article (cf. page 3, line 25 to page 4, line 21), the skilled person faced with the problem defined above would not be encouraged in view of (9) to add from 5 to 60 wt% Al_2O_3 to the starting mixture of (8) and to form a sialon, or to replace the matrix of the substrate body by one comprising a sialon.

- 6.3 Document (6) or its translation in English, document (6a), discloses a composition of ceramic material containing 5-15 wt% MgO or Al_2O_3 (for example 11 wt%), 55-90 wt% silicon nitride and 5-30 wt% titanium carbide. This material can be used for cutting tools (cf. Claim 1, example and page 1, line 2). Therefore, there existed no prejudice against the use of more than 5 wt% aluminium oxide in a silicon nitride based composite materials for cutting tools. This is also confirmed by document (12) which discloses similar silicon nitride based materials. According to (6a) the problem of low hardness and wear

resistance of prior art materials containing silicon nitride, silicon carbide, MgO or Al₂O₃ is solved by using TiC instead of SiC in the composition. In other words the improved properties are not attributed to the presence of aluminium oxide.

Without providing evidence to support his affirmation, the Appellant has alleged that the ceramic material according to (6a) would also comprise a sialon. However, the Respondent has argued that no sialon would be formed under the conditions used for preparing the composite material and that (6a) was silent about the structure of the matrix. In this respect the Board observes that according to the example of (6a) the starting mixture is hot-pressed at a temperature of 1700°C and a pressure of 400 kg/cm² for only 30 minutes, and there is no information as regards the final product apart from its hardness and wear resistance. However, it is well known that the rates of the reactions which lead to the formation of a sialon depend upon a lot of parameters, in particular upon the composition of the starting mixture (i.e. the relative proportions of the components and the presence of impurities, in particular the presence of SiO₂ at the surface of Si₃N₄), upon the different particle sizes of the powders, and upon the temperature and time period of sintering or hot-pressing. According to the patent in suit the starting mixture which contained yttrium oxide and a far higher amount of Al₂O₃ was prereacted at 1725°C for a period of 5 hours or sintered at 1750°C for 1,5 hours to form a modified silicon aluminium oxynitride. Under these circumstances, the board is not convinced, in the absence of any data characterising the structure of the ceramic material of (6a), that a composite material comprising a sialon structure was formed under the conditions reported in this document. Accordingly, the Board comes to the conclusion that document (6a) neither teaches nor suggests

the use of a sialon containing matrix in the substrate body of a cutting tool.

- 6.4 Document (12) relates in a more theoretical and general form to optimisation of the properties of composite ceramic tool materials. The components of the starting mixture are the same as in (6), namely TiC, Al₂O₃ and Si₃N₄. The compositions of table 1, which contain at most about 11 vol% Al₂O₃, were sintered by hot-pressing at a temperature of at most 1720°C for a time of at most 15 minutes, i.e. a temperature slightly higher than in (6) but a period reduced by half. Therefore, the probability to form a sialon matrix is not higher than in (6) and the Board's conclusion in the preceding point 6.3 applies likewise to this document.
- 6.5 The sintered silicon nitride based material disclosed in document (5) and suitable for cutting tools contains up to 10 wt% of one or several sintering accelerators selected from AlN, Al₂O₃, MgO, SiO₂, Fe, Co, Ni and the oxides of rare earth elements, and 5 to 40 wt% of titanium carbide, nitride or carbonitride (cf. page 4, lines 3-11). In example 1 of (5), the only example using alumina in the starting mixture, the alumina content of the matrix is 3 wt%. It derives from the statement at page 4 (last paragraph) about the deposition of a glass phase in the grain boundary regions that the alumina does not form a silicon aluminium oxynitride with the silicon nitride. Therefore in view of the disclosure in (5), taken in combination with (8), (9) and (6) the skilled person would not have been prompted to choose Al₂O₃ from the list of sintering aid accelerators and to add it to the starting mixture including MgO or another sintering aid and then to treat the mixture under such conditions that a sialon structure is formed.

6.6 Document (4) represents a review article concerning the known ceramic and cermet material for cutting tools and comparing their properties. Reference to a hot-pressed sialon material consisting of silicon nitride, aluminium oxide and titanium carbide is made in the small paragraph headlined "Field tests for sialon". It clearly derives from (4) that, in 1981, this material was a new material still in the laboratory-test and preliminary field-test stage. Apart from the statement that this hot-pressed sialon exhibits a hardness of Ra 94 and a transverse rupture strength of 150.000 or 200.000 psi (cf. page 167, third column), there is no further information as to the relative proportions of the individual components in this material. Nor does this document disclose the strength of this material at high cutting temperatures, its creep resistance or its life time when used for cutting steel. Under these circumstances, even if the skilled person could have thought of replacing the substrate body of (8) by a TiC containing sialon, the Board is not convinced that he would have done this in the absence of any information not only about the cutting performances of this new material when cutting steel but also about its composition. Assuming to the Appellant's benefit that the skilled person would nevertheless have been encouraged to make experimentation in this direction, then he would have had to determine the appropriate proportions of the components which lead to the desired cutting performances after hot-pressing and coating with the TiC or Al_2O_3 layer. As, on the one hand, none of the documents (8), (9), (5), (6) and (12) discloses cutting tools having a sialon matrix and, on the other hand, (4) does not mention the presence of additional oxides or the necessity of incorporating additional sintering aids or modifiers in the starting mixture (according to (5) Al_2O_3 belongs itself to the group of sintering aids), the Board cannot follow the Appellant's argument that the composition as

defined in Claim 1 could have been determined by simple routine experimentation in view of these documents.

6.7 Documents (1) and (7) were cited by the Appellant to show that it was known to provide a substrate body for a cutting tool with an adherent refractory material layer in order to improve the performances thereof. The substrate bodies of (1) consist of aluminium oxide with additives such as MgO or hard carbides or nitrides and a binder whereas the substrate bodies of (7) are silicon nitride based sintered bodies which do not include any hard refractory material dispersed in the matrix. As these documents do not relate to cutting tools having a sialon matrix, they cannot point towards the claimed solution.

6.8 As regards document (13), the Appellant filed only pages 1135, 1141 and 1142 thereof with his letter of 4 September 1991. These pages were cited in order to illustrate the general knowledge about the structure of β' -sialons. Although the Appellant was obviously aware of the content of this document before oral proceedings, he did not refer thereto in support of his objections concerning lack of inventive step.

Document (13) is a scientific study about sialons and related nitrogen ceramics disclosing the different phases in the $\text{Si}_3\text{N}_4 - \text{AlN} - \text{Al}_2\text{O}_3 - \text{SiO}_2$ system. The properties of β' -sialons are compared with those of hot-pressed silicon nitride, in particular the thermal shock properties, the oxidation resistance, the creep behaviour and the compatibility with molten metals (cf. pages 1147 and 1148). Although it is pointed out that the sialons are scientifically and technologically of interest and that pressureless sintering to the theoretical density can be achieved more easily with sialons than with silicon nitride, the conclusion as regards their potential uses is

that the metallurgical applications of some of the sialons for holding and conveying molten metals are probably of more immediate importance than their use as engineering ceramics (cf. page 1156, last paragraph and page 1157, 1st and 2nd paragraphs). In this context it is emphasised that their abrasive properties have not been explored. Other various potential uses are mentioned, for example in the field of solid electrolytes, of catalysis or for the preparation of sialon glasses, however (13) is completely silent about a possible application in the field of cutting tools. Furthermore, according to (13) more details and careful investigations are required to establish reliable preparative methods and to characterise the products more precisely (cf. page 1156, right-hand column; page 1157, line 19 and last sentence). Therefore, this document does not give any information as to which starting compositions might in combination with a hard refractory material have been suitable for the manufacture of coated cutting tools having good performances when used for cutting steel and thus, even in combination with the teaching of the other documents, it would have been of no assistance to the skilled person faced with the problem defined above. The fact that (13) further discloses a yttrium silicon oxynitride and refers to yttrium-sialon systems or to the possibility of subsequently incorporating densification aids in the sialon structure would not have been helpful since it does not mention the properties of the resulting products.

- 6.9 Therefore, the Appellant's conclusion that it was obvious to arrive at the claimed cutting tool by a simple combination of only three or four of these documents, for example (8), (4) and (9); or (8), (6) and (9); or (7), (6) and (9), optionally in combination with (13) cannot be followed by the Board. Taking into consideration the teaching of the cited documents analysed above, it seems

that this conclusion has been reached on the basis of an ex post facto analysis.

It results from the preceding that the subject-matter of Claim 1 meets the requirements of inventive step set out in Article 56.

6.10 The same applies to Claims 2 to 18 which relate to particular embodiments of the cutting tool according to Claim 1.

7. Since the main request is allowable, there is no need to investigate the Respondent's auxiliary request.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

E. Görgmaier

P.A.M. Lançon