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Anmeldenummer / Filing No / N° de la demande : 80 302 078.3

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Bezeichnung der Erfindung: A honeycomb structure for use as a catalyst  
Title of invention: support for automobile exhaust  
Titre de l'invention :

Klassifikation / Classification / Classement : C04B 35/46

### ENTSCHEIDUNG / DECISION

vom / of / du 13 December 1988

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /  
Titulaire du brevet :

NGK Insulators, Ltd.

Einsprechender / Opponent / Opposant :

01 Hoechst CeramTec AG  
02 Hutschenreuther AG

Stichwort / Headword / Référence :

EPU / EPC / CBE Articles 54(1), 56

Schlagwort / Keyword / Mot clé :

"Novelty of a ceramic material (denied;  
overlapping numerical ranges) known material in  
a structure for a specific use" - "inventive  
step (affirmed)"

Leitsatz / Headnote / Sommaire

Europäisches  
Patentamt

European Patent  
Office

Office européen  
des brevets

Beschwerdekammern

Boards of Appeal

Chambres de recours

Case Number : T 354/87 - 3.3.1



DECISION  
of the Technical Board of Appeal 3.3.1  
of 13 December 1988

Appellant :  
(Opponent 01)

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Decision under appeal :

Interlocutory decision of the Opposition Division of  
the European Patent Office dated 4 August 1987  
concerning maintenance of European patent  
No. 0 036 462 in amended form.

Composition of the Board :

Chairman : K. Jahn

Members : R. Spangenberg  
G.D. Paterson

## Summary of Facts and Submissions

- I. The mention of grant of European patent No. 036 462 in respect of European patent application No. 80 302 078.3 filed on 19 June 1980 and claiming priority of 26 March 1980 of an earlier application in Japan was announced on 20 July 1983. Two notices of opposition supported by new documents were filed on 10 April 1984 and 17 April 1984 requesting the revocation of the patent on grounds of lack of novelty and inventive step.
- II. By an interlocutory decision dated 4 August 1987 the Opposition Division held that the patent could be maintained in amended form with the text communicated to the parties pursuant to Rule 58(4) on 20 February 1987. The patent specification as amended contained four claims, Claim 1 reading as follows:

"1. A low-expansion ceramic material consisting essentially of from 2.5 to 17% by weight of magnesia, from 13.5 to 62% by weight of alumina, from 31.5 to 75% by weight of titanium oxide (calculated as titanium dioxide), and from 2 to 10% by weight of iron oxide (calculated as ferric oxide), the crystalline phase of the material having a major component which is a magnesium oxide/aluminium oxide/titanium oxide/iron oxide solid solution and containing a secondary phase or phases present in said major component, said secondary phase or phases being in total an amount not exceeding 20% by weight of the material of one or more of rutile, spinel and corundum crystals, the material having a coefficient of thermal expansion of not more than  $20 \times 10^{-7}$  ( $1/^{\circ}\text{C}$ ) in the temperature range  $25^{\circ}\text{C}$  to  $800^{\circ}\text{C}$ , and a melting point of not less than  $1500^{\circ}\text{C}$ , said coefficient of thermal expansion being maintained after

exposure of the material to a temperature of 1100°C for 1000 hours.

The Opposition Division considered twelve documents, the most important of them being

- (1) DD-C-29 794
- (2) US-A-2 776 896.
- (3) H. Walter, Silikattechnik 21 (1970), 304-306
- (6) Pohlmann et al, Ber. Dtsch. Keram. Ges. 52 (1975) 179-183.

The Opposition Division stated that it was known that aluminium titanate has a remarkable low coefficient of thermal expansion (CTE) but does not have adequate strength and decomposes into alumina and titania when exposed to relatively high temperatures and that it was also known, for instance from (2) that aluminium titanate can be stabilised against dissociation by replacement of some of the cations in the aluminium titanate crystal by magnesium or iron. However, no example of a combination of magnesia, alumina, titania and iron oxide in the amounts specified in the above Claim 1 was disclosed in this or any other of the cited documents. The only document disclosing a ceramic material containing all four components was (1). However the composition of Example 4 falls outside the ranges of Claim 1 because the amount of iron oxide is too low and that of magnesia too high.

The Opposition Division stated that the technical problem underlying the patent in suit was to improve the stability of magnesium stabilised aluminium titanate, which according to (2) slowly decomposes at prolonged heating (at least 100 hours) at a temperature between 1000 and 1200°C. The claimed compositions, however, were stable for at least 5000 hours at 1100°C as demonstrated by table 2 of the

specification and hence, considering that neither (2) nor any other document cited in the opposition procedure could suggest to a skilled person that aluminium titanate containing specified amounts of iron oxide and magnesia would be stable to this extent held that the claimed compositions involved an inventive step.

III. The Appellants gave notice of appeal against the above decision; Appellant I by telefax of 1 October 1987, confirmed on 9 October 1987 and Appellant II on 3 October 1987. The appeal fees were paid on the same dates. Statements of grounds were submitted by Appellant II on 3 December 1987 and Appellant I by telecopy of 14 December 1987, confirmed on 18 December 1987.

Appellant I argued that it was well known that ceramic materials on the basis of aluminium titanate have only poor mechanical properties, see for instance (3), page 304, right column and (6), page 179, left column. Since it was known from (1) that the pressure strength of such materials may be improved by incorporating into a basic material containing titania, alumina and magnesia additionally oxides of heavy metals, for instance iron oxide, see column 4, line 32 to column 6, line 4, the claimed compositions are obvious. He further stated that the presence of additional crystalline phases is also envisaged in (1), Example 3, in order to obtain a CTE close to zero and that therefore no prejudice existed against the presence of secondary phases in aluminium titanate ceramics and submitted test results showing that secondary phases are automatically formed if mixtures of oxides within the ranges indicated in the disputed patent are fired at 1600°C. The broad range of firing temperatures indicated in the patent in suit was said to be the range normally used and the presence of secondary phases was found to be the inevitable consequence of the oxide composition.

Appellant II additionally disputed that the claims underlying the decision under appeal are in accordance with Article 100(c) of the EPC, because in his opinion the scope of protection was extended beyond that of the patent as granted. He also drew attention to further formal aspects of the present Claim 1. This Appellant further disputed the novelty of the claimed subject-matter in view of (1), Example 4 and Claims 1 and 5, and the presence of an inventive step, referring to (2), especially column 6 and Claim 1.

- IV. In the Respondent's opinion the present claims are not broader in scope than the claims as granted. He maintained the opinion that the claimed subject-matter is novel over the disclosure in (1) because the claimed material is a selection from (1). He disputed that secondary phases are inevitably formed following the disclosure in (1) since firing is not mentioned in Example 4 which may rather relate to a brick-making process and in any case no information is available about mutual solubilities in the four-component-system according to the claimed invention. Concerning inventive step the Respondent submitted that the Opponent's approach to this question is incorrect and stated that there was no incentive at all for the skilled man to make this selection since there is no indication in the citations that such a selection might give rise to stability. Regarding the test results submitted by Appellant I, the Respondent stated that they confirmed the presence of an inventive step because, except for experiment 5, they show a very much better CTE than the known material of Example 4 of (1) and thus an unexpected effect.

- V. At the oral proceedings on 13 December 1988 the Board

expressed doubts regarding the novelty of the claimed subject-matter.

The Respondent then filed as an auxiliary request one single claim limited to a "honeycomb structure for use as a catalyst support in purifying apparatus for automobile exhaust, formed of" a ceramic material as specified in Claim 1 as maintained by the Opposition Division, and a description adapted thereto.

He argued that none of the cited documents contain an incentive to use ceramic materials containing aluminium titanate for forming honeycomb structures for the use mentioned above, the uses envisaged in (1) are all related to materials in the form of bricks or other structures involving thick layers of material where high mechanical strength is not important. Honeycomb structures for use as catalyst support in automobile exhausts however comprise a matrix of cells surrounded by very thin walls which must remain stable under firing conditions, i.e. at temperatures close to the melting point and must resist mechanical shocks at low and high temperatures. Also, it was not obvious to use for the envisaged purpose materials containing secondary phases because these are generally known to cause an increase in the CTE and consequently a worse thermal shock resistance.

- VI. The Appellants submitted that it is already acknowledged in the description page 2, lines 20 to 24 that ceramic catalyst supports conventionally are made from cordierite and are in the form of a honeycomb structure. As document (1) already recommends the ceramic materials also for use in rocket and jet engine technology and these uses are very similar to the use envisaged in the patent in suit, they found the subject-matter of that claim obvious.

Furthermore, catalytic purification of automobile exhaust gases could not have been mentioned in (1) because it was not a relevant technical problem at the filing date of this document in 1962. When it became necessary later on, after 1970, to look for suitable low expansion ceramic materials for this purpose requiring a higher melting temperature than cordierite, a skilled person would search the literature and find document (1) which discloses in column 1, line 28 to column 2, line 1 that the aluminium titanate ceramics mentioned therein are low expansion ceramics having a higher melting point than cordierite.

- VII. The Appellants requested that the decision under appeal be set aside and the patent revoked. The Respondent requested that the patent be maintained either with description, claims and drawing allowed by the Opposition Division (main request) or in the text submitted at the oral proceedings (auxiliary request).

At the end of the oral proceedings, the decision was announced to maintain the patent on the basis of the text submitted at the oral proceedings.

#### Reasons for the Decision

1. The appeal complies with Articles 106 to 108 EPC and Rule 64. It is therefore admissible.
2. No formal objections arise against the statement of claim according to both requests.

These claims are limited with respect to Claim 1 as originally filed and as granted since they contain the preferred ranges of oxide contents of Claim 2 and further specify that in addition to the major component of the

crystalline phase, certain amounts of certain secondary phases are present. In the Board's judgement this latter feature, which is based on the description as granted (cf. page 4, lines 25 to 32) and the patent application as filed (cf. page 5, last paragraph) also constitutes a clear limitation of Claim 1 as granted and therefore does not extend the scope of protection conferred by that claim. Claim 1 according to the auxiliary request is further limited by the feature of Claim 3 as originally filed and granted. The envisaged use of these honeycomb structures is disclosed in the description as granted, page 5, lines 1 to 4 corresponding to page 10, lines 6 to 10 of the patent application as filed.

The minimum percentages of oxides indicated in the present claims do not add up to 100%. This, however, does not mean that the ceramic material described therein may contain substantial amounts of unspecified components and that these claims are open to objection under Article 100(b) EPC as it is alleged by Appellant II. The language of these claims ("consisting essentially of") being construed in the light of the description not disclosing the presence of any filling material clearly excludes the presence of intentional amounts of unspecified material and the ranges of percentages must therefore be understood as being interdependent, i.e. if a smaller amount of one oxide is present this is made up within the indicated ranges by higher amounts of the others.

3. Claim 1 according to the main request relates to a ceramic material as such. This material is characterised by a content of

from 2.5 to 17% by weight of magnesia,  
from 13.5 to 62% by weight of alumina,  
from 31.5 to 75% by weight of titania and

from 2 to 10% of ferric oxide

and further by the presence of up to 20% by weight of the ceramic material of one or more secondary phases of rutile, corundum or spinel crystals. This material shall further possess a specified low CTE, a melting point not below 1,500°C and a specified thermal stability.

It is obtained according to the patent in suit by firing an appropriate mixture of the starting oxides at temperatures between 1300 and 1700°C for 0.5 to 48 hours. These firing conditions are common in the art for making ceramic materials on the basis of aluminium titanate, see e.g. (3), the sentence bridging pages 305 and 306, and (6), item 3.1.

Document (1) according to Claim 1 relates to a process for manufacturing a highly fire-resistant material consisting of 15 to 75% TiO<sub>2</sub>, 70 to 25% Al<sub>2</sub>O<sub>3</sub>, 0 to 40% SiO<sub>2</sub> and 0 to 20% MgO and having a CTE lower than  $4 \cdot 10^{-6}$ , preferably a negative CTE or a CTE close to zero. More specifically, according to column 3, lines 11 to 15 it relates to materials within the ternary system MgO-Al<sub>2</sub>O<sub>3</sub>-TiO<sub>2</sub> within the above limits. As the system would not be "ternary" if no MgO is present, the Board holds that despite the indication that the MgO-content may be 0% MgO is an essential constituent of these materials. This is further confirmed by the fact that the materials obtained according to all worked examples indeed contain MgO.

The above reference to a "ternary system" as well as the fact that it is further stated in column 3, lines 52 to 59 that the oxide mixture is fired or sintered and that the firing or sintering conditions may be used to adjust the CTE of the resulting material show that - contrary to the Respondent's submission - this document relates to ceramic

materials which are made by firing. It is further stated in (1), column 4, lines 21 to 24 that the mechanical stability of these materials can be improved by adding small amounts of an alkaline earth or heavy metal oxide. Such additives are exemplified in Example 4 wherein BaO is used as an alkaline earth oxide or Fe<sub>2</sub>O<sub>3</sub> as a heavy metal oxide. The amounts are 1% and 1.5%, respectively and there are no further examples for such additives. In Claim 5 of (1) it is explained what is meant by "small amounts", namely 0.05 to 15%, preferably 0.2 to 5% of oxides of 8 elements including iron. The Board therefore holds that Fe<sub>2</sub>O<sub>3</sub> is not mentioned just by way of example but as the preferred heavy metal oxide and that therefore in these circumstances the disclosure of (1) cannot be regarded as being limited to the specific amounts of the oxides mentioned in this example but that (1) discloses a composition containing MgO, Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> and Fe<sub>2</sub>O<sub>3</sub> in the amounts indicated above, with other words, (1) discloses ceramic materials containing from 15-75% TiO<sub>2</sub>, from 70 to 25% Al<sub>2</sub>O<sub>3</sub>, up to 20% MgO and at least from 0.2 to 5% Fe<sub>2</sub>O<sub>3</sub>; i.e. materials which fall within the ranges of oxide composition defined in Claim 1 of the patent in suit.

4. The presence of secondary phases of rutile, corundum or spinel crystals in the Board's view is an inevitable result of the use of oxide quantities outside the weight ratios resulting in the formation of crystalline phases of the "Anasovit"- type (see (3), page 304, in the middle of the right column). These weight ratios correspond to a material consisting of Al<sub>2</sub>TiO<sub>5</sub>, MgTi<sub>2</sub>O<sub>5</sub> and Fe<sub>2</sub>TiO<sub>5</sub> which at equilibrium conditions form a solid solution, i.e. a single crystalline phase. Beyond these weight ratios which are encompassed by the disclosure of (1) however, secondary phases of rutile, corundum or spinel crystals are formed if conventional firing conditions are chosen according to (1), column 3, lines 53 to 56 which result in a material with a

low CTE. This has been confirmed by experiments carried out by Appellant I the results of which have been filed together with his Statement of Grounds for Appeal demonstrating that if the oxide composition of Example 4 of (1) is maintained except that the  $\text{Fe}_2\text{O}_3$  content is raised to 4.5% while keeping the total amount of  $\text{MgO} + \text{Fe}_2\text{O}_3$  constant a material containing spinel is obtained.

Similar results were obtained when other oxide compositions within the claimed range were chosen. The X-ray spectra do not allow the exact determination of the amount of the secondary phases present; however, from the known stoichiometric requirements for the formation of secondary phases it can be inferred that the amounts of oxides are not sufficient for the formation of more than 20% by weight of the secondary phases. Thus, this feature does not distinguish the claimed subject-matter from that disclosed in (1).

5. It is not disputed that when following the teaching of (1) as mentioned above, materials are obtained which have a CTE below the limit indicated in Claim 1 of the patent in suit and a melting point above  $1500^\circ\text{C}$  as also required by that claim (see (1), col. 3, lines 27 to 30). Consequently, these features, too, do not distinguish the claimed subject-matter from that disclosed in (1).

Similar considerations apply to the requirement that the low CTE shall be maintained after exposure of the ceramic material to a temperature of  $1100^\circ\text{C}$  for 1000 hours. There is no disclosure in the patent specification allowing the conclusion that this requirement is not automatically fulfilled if the other requirements mentioned above are met. Especially the firing conditions mentioned in the patent in suit are identical with those conventionally applied. Therefore, in the Board's judgement, the known

materials, which are the result of a process involving identical starting materials and reaction conditions must also possess this property.

7. For the reasons set out in the preceding paragraphs, the Board holds that Claim 1 of the patent in suit according to the main request comprises subject-matter already disclosed in (1) and hence lacks novelty. Therefore, the Respondent's main request fails.
8. According to the auxiliary request, the statement of claim is limited to a honeycomb structure for use as catalyst support in catalytic purifying apparatus for automobile exhausts. Such subject-matter is not disclosed in any of the cited documents since none of them relates to honeycomb structures made of ceramic materials of the chemical composition indicated in the claim under consideration. It is therefore novel.
9. The closest prior art with respect to the honeycomb structures now claimed is that acknowledged in the patent in suit, page 2, lines 16 to 30 where it is stated that honeycomb structures for use as catalyst supports in catalytic purifying apparatus for automobile exhausts are conventionally made of cordierite (magnesium-aluminium-silicate).

These known articles, however, are not sufficiently heat resistant due to the relatively low melting points of the ceramic materials from which they are made. Thus at higher temperatures which arise if in order to ensure good efficiency the catalytic purifying apparatus is situated near the engine the honeycomb structure breaks down and the exhaust is plugged.

The technical problem underlying the claimed subject-matter may therefore be seen in providing honeycomb structures for use as catalyst supports in catalytic purifying apparatus in automobile exhausts which have sufficient mechanical stability at higher temperatures than those tolerated by the above conventional catalyst support.

This problem is solved by providing honeycomb structures formed of a low expansion ceramic having a chemical composition and physical properties within the ranges defined in Claim 1 of the patent in suit.

10. In the Board's judgement, the expression "for use as a catalyst support in catalytic purifying apparatus in automobile exhaust" in Claim 1 of the patent in suit is to be construed as a limiting feature defining not only the specific thin walls of this structure but also the other physical and mechanical properties required for the envisaged use.
11. In the light of the preceding construction of Claim 1 under consideration the Board is satisfied that the above-defined problem has been plausibly solved.
12. It is true that a person skilled in the art may have expected that materials disclosed in (1) are sufficiently stable against chemical decomposition since it was known from (2) that aluminium-titanate ceramics containing iron oxide and magnesium oxide have this property to a certain extent, cf. the table on column 8. These properties, however, while being essential for the envisaged use, are not sufficient therefor.
13. In the Board's view, a ceramic material for use as catalyst support in automobile exhausts must have not only an

excellent thermal shock resistance which is normally provided by any material having a CTE close to zero and sufficient chemical stability but must provide additional properties in combination with them, especially good mechanical shock resistance, sufficient physical strength over a range of temperatures from room temperature up to a temperature close to the melting point and a low gradient of the softening - shrinkage curve from the softening temperature to the melting temperature (see the patent in suit, page 3, line 25 to 32), i.e. a high heat resistance at temperatures near the melting point preventing these materials from softening at the high temperatures involved in catalytic purification of automobile exhaust gases. A person skilled in the art looking for a material having better performance than cordierite as a catalyst support for automobile exhausts therefore had to consider more than the CTE and the chemical stability of the alternative materials envisaged.

However, with respect to these additional requirements, document (1) is completely silent. The structures and uses envisaged in this document are lightweight stones for linings of electric furnaces and highly fire-resistant saggars for porcelain manufacture, i.e. structures involving thick layers of ceramic material which are not exposed to mechanical shock, particularly at high temperatures. Insofar as applications of these materials in rockets or jet engines are concerned, which are generally mentioned in (1) a skilled person would therefore at most consider similar structures, e.g. heat protecting shields or linings for combustion chambers.

Thus, no incentive can be derived from (1) to use materials disclosed therein for the purpose envisaged in the patent in suit, i.e. for forming honeycomb structures for use as catalyst support in automobile exhausts. Further

information concerning the mechanical properties of aluminiumtitanate however is available from (3) and (6) reflecting the common general knowledge about aluminium-titanate ceramics. In the first complete paragraph on page 304 of (3), right column reference is made to (1) and it is stated that ceramic materials on the basis of aluminiumtitanate normally show insufficient mechanical properties and that addition of  $\text{SiO}_2$  is required in order to obtain materials with sufficient physical strength. A person skilled in the art therefore would not have considered the use of aluminiumtitanate ceramics free of  $\text{SiO}_2$  for purposes where high physical strength is important.

In document (6), issued in 1975, it is also stated in the abstract that the thermal and mechanical properties of aluminiumtitanate are improved by adding kaolin. In the absence of this additive the mechanical properties of aluminiumtitanate are said to be poor, see (6), paragraph 1 (introduction) on page 179. The addition of  $\text{MgTi}_2\text{O}_5$  and  $\text{Fe}_2\text{TiO}_5$  are said to improve the stability of  $\text{Al}_2\text{TiO}_5$  against chemical decomposition but to further weaken the physical strength, see (6), page 180, left column, last paragraph and right column, first paragraph.

14. Therefore, in the Board's view, the Appellants' allegation that a person skilled in the art searching the literature concerning aluminiumtitanate ceramics in order to find a material to replace cordierite as catalyst support in automobile exhausts would have selected from the materials disclosed in (1) the materials having the chemical composition and the crystalline phases indicated in the claim under consideration is not supported by the cited prior art. If this prior art would have provided any incentive at all to use honeycomb structures made from aluminiumtitanate ceramics as catalyst support in

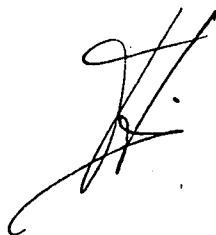
automobile exhausts, it would rather have been directed to the use of ceramics also containing SiO<sub>2</sub> in order to get sufficient physical strength. For this reason, in the Board's judgement, the obviousness objection raised by the Respondents against the subject-matter of the claim of the auxiliary request must be regarded as based on an ex-post-facto analysis of the prior art and hence must fail.

**Order**

For these reasons, it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the Opposition Division with the order to maintain the patent with description, drawing and claim in accordance with the auxiliary request as filed during the oral proceedings.

The Registrar:



The Chairman:

