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Aktenzeichen / Case Number / N° du recours : T 171/87 - 3.3.1
Anmeldenummer / Filing No / N° de la demande : 80 302 077.5
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Bezeichnung der Erfindung: Honeycomb structure for use as a catalyst support
Title of invention: 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 :

EPO / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé :

"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 171/87 - 3.3.1

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



Appellant :
(Proprietor of the patent)

NGK INSULATORS, LTD.
2-56, Suda-cho, Mizuho-ku
Nagoya-shi, Aichi 467
Japan

Representative :

Paget, Hugh Charles Edward
MEWBURN ELLIS & CO.
2/3 Cursitor Street
London EC4A 1BQ
GB

Respondent :
(Opponent 01)

Hoechst CeramTec AG
Postfach 1508
D-8672 Selb/Bayern

Representative :

Reuter, Johann-Heinrich, Dr.
HOECHST AKTIENGESELLSCHAFT
Zentrale Patentabteilung
Postfach 80 03 20
D-6230 Frankfurt/Main 80

Respondent :
(Opponent 02)

Hutschenreuther AG
D-8672 Selb

Representative :

Czowalla, Ernst
Patentanwälte E. Czowalla P. Matschkur
Dr.-Kurt-Schumacher-Strasse 23
Postfach 9109
D-8500 Nürnberg 11

Decision under appeal :

Decision of the Opposition Division of the European
Patent Office of 22 October 1986, posted on
20 February 1987, revoking European patent
No. 0 036 052 pursuant to Article 102(1) EPC.

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. 36 052 in respect of European patent application No. 80 302 077.5 filed on 19 June 1980 and claiming priority of 19 March 1980 of an earlier application in Japan was announced on 27 July 1983 (Bulletin 83/30). The patent specification contained nine claims. Claim 1 read as follows:

"A low-expansion ceramic material having a chemical composition consisting essentially of from 2 to 20% by weight of magnesia, from 10 to 68% by weight of alumina, and 30 to 80% by weight of titanium oxide (calculated as titanium dioxide); the major component of the crystalline phase of the material being a magnesium-aluminum-titanate phase, and the material having a coefficient of thermal expansion of not more than 20×10^{-7} ($1/^{\circ}\text{C}$) in the temperature range of 25° to 800°C and a melting point of not less than $1,500^{\circ}\text{C}$."

Two notices of opposition, supported by new documents, were filed against this patent on 15 February 1984 and 18 April 1984, requesting the revocation of the above patent in its entirety on grounds of lack of novelty and inventive step.

Finally the patent was only defended in restricted form by limiting the statement of claim to a low expansion ceramic material containing from 0.5 to 20% by weight of one or more of rutile, spinel and corundum crystals as secondary phases, auxiliarily by further limiting the subject-matter to that material in the form of a honeycomb structure.

- II. By a decision of 22 October 1986 posted 20 February 1987 the Opposition Division revoked the patent, stating

substantially that the subject-matter of both proposed amended claims did not involve an inventive step regarding documents

- (1) DD-A-29 794
- (3) H. Walter: Silikattechnik 21 (1970), Volume 9, pages 304-306.
- (8) Berezhnoi e.a. Ukrain. Chem. Zhur. 162 (1955), page 162.
- (9) Keramische Massen auf der Basis von Aluminiumtitanat, Tonindustrie Zeitung, Volume 98 (1974), No. 12, pages 315-318.
- (11) WADC Technical Report 53-165 by N.R. Thielke (1953).

The Opposition Division considered (1) as the closest prior art, especially Examples 1 and 3. These compositions were said to have a melting point, a coefficient of thermal expansion (CTE) and an elementary composition in the ranges specified in the patent in suit, the only difference being that the claimed compositions contain secondary phases which are not explicitly mentioned in (1).

The technical problem vis-à-vis (1) was seen in improving heat resistance and mechanical strength of the known materials without adversely affecting the low expanding characteristics. The solution of this problem by the intentional presence of small amounts of the above secondary phases was considered obvious since it was already known from (11) that secondary phases such as alumina and rutile in aluminium titanate improve the strength of these ceramics. Also, the firing conditions

were said to be common in the art regarding e.g. (3), page 305.

The further limited subject-matter was likewise considered unallowable because the honeycomb form of ceramic materials was known in the art as admitted by the patentee.

III. On 29 April 1987 the Appellant (the Patentee) filed a notice of appeal by a telex confirmed on 2 May 1987 at the same time paying the appeal fee. A statement of grounds was filed on 20 June 1987.

IV. In reply to this statement of grounds Respondent I filed a new document

(12) Trans. J. Brit. Ceram. Soc. 5(1972), 215-220

demonstrating that under equilibrium conditions secondary phases of the kind and in the amounts specified in the above Claim 1 are present in the system $\text{MgO-Al}_2\text{O}_3\text{-TiO}_2$ at concentrations of these three constituents within the ranges specified in Claim 1.

V. Oral proceedings took place on 13 December 1988.

VI. The Appellant requested that the decision under appeal be set aside and the patent maintained with the documents filed at the oral proceedings. The only independent claim reads as follows:

"A honeycomb structure for use as a catalyst support in catalytic purifying apparatus for automobile exhaust, formed of a low-expansion ceramic material having a chemical composition consisting essentially of from 2 to 20% by weight of magnesia, from 10 to 68% by weight of

alumina and from 30 to 80% by weight of titanium oxide (calculated as titanium dioxide), the major component of the crystalline phase of the ceramic material being a magnesium-aluminium-titanate phase and the ceramic material further containing from 0.5 to 20% by weight of one or more of rutile, spinel and corundum crystals, and the ceramic material having a coefficient of thermal expansion of not more than 20×10^{-7} (1/°C) in the temperature range of 25°C to 800°C and a melting point of not less than 1,500°C."

He argued that (8) and (12) were scientific publications not relating to any industrial application of the ceramic materials disclosed therein and that also none of the documents (1), (3), (9) and (11) discloses that ceramics on the basis of aluminutitanate (tialite) would be suitable for making honeycomb structures for use as catalyst support in automobile exhausts. Furthermore in his opinion a general prejudice had existed against the presence of secondary phases in low expansion ceramics since such phases were known to increase the CTE and hence would decrease the thermal shock resistance.

VII. The Respondents essentially argued that according to what is already acknowledged in the patent in suit it was known to use cordierite as a ceramic material for honeycomb structures for use as catalyst supports in automobile exhausts. From (1), column 1, line 38 to column 2, line 1 it was further known that the materials disclosed in this document have higher melting points than cordierite and are therefore useful in jet engine and rocket technology, see (1), col. 3, line 27 to 31. Such applications were in the Respondents' opinion comparable with the use envisaged for the claimed honeycomb structures.

The Respondents further stated that at the filing date of (1) in 1962 exhaust gas purification was not a relevant technical problem. Catalytic purification of automobile exhaust gases did not become necessary before about 1975 when in the USA and in Japan severe legal requirements had to be met. Being then faced with the problem a skilled person would have searched the literature for suitable materials and thus would inevitably have considered the materials disclosed in (1). They also submitted that the alleged prejudice against the presence of secondary phases did not exist for materials containing aluminiumtitanate since the beneficial effect of added corundum is shown by (1), Example 3 and that such phases were in any case present in the materials disclosed in (1) if the chemical composition of these materials is not very close to the "Anasovit-line" shown in (8) where solid solutions are formed. The Respondents consequently found the claimed subject-matter obvious and requested that the appeal be dismissed.

VIII. At the oral proceedings the decision was announced to maintain the patent in amended form as requested by the Appellant.

Reasons for the Decision

1. The appeal complies with the requirements of Art. 106 to 108 EPC and Rule 64 and is therefore admissible.

2. The amended claims are properly based on the application documents as filed (for Claim 1 see Claims 1, 5 and 6 and lines 8 and 9 of the last paragraph of page 13, for Claims 2 to 4 see Claims 2 to 4 as filed) and the patent specification as granted (for Claim 1 see Claims 1, 5 and 6 and p. 6, lines 52 and 53, for Claims 2 to 4 see Claims 2 to 4 as granted). Their subject-matter does not extend the scope of protection conferred by the claims as granted. Therefore the requirements of Art. 123(2) and (3) are met.
3. The claimed 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 Claim 1 of the patent in suit. It is therefore novel. As novelty has no longer been disputed, no more detailed explanation is required.
4. The closest prior art with respect to the honeycomb structures now claimed is that acknowledged in the patent in suit, page 2, lines 15 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 supports.

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.

5. In the Board's judgement the expression "for use as a catalyst support in catalytic purifying apparatus for 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.
6. In the light of the preceding construction of Claim 1 the Board is satisfied that the above-defined problem has been plausibly solved.
7. The Board finds that, contrary to the Appellant's submission, document (1) describes true ceramic materials because the oxide mixtures are fired or sintered, see (1), column 3, lines 53 to 56 and that these materials have the same overall composition as the materials used according to the patent in suit. Especially they contain secondary phases as an inevitable result of the application of conventional firing conditions to e.g. the oxide mixture indicated in Example 3 of (1).

This finding is based on the consideration that solid solutions in the system $MgO-Al_2O_3-TiO_2$ are only formed between the constituents magnesium-dititanate ($MgTi_2O_5$) and aluminiumtitanate (Al_2TiO_5), i.e. at certain weight

ratios of the three oxides involved which can be calculated from these formulas. At the weight ratios which are comprised by the disclosure of (1) and are exemplified by the basic composition ("Grundmase") mentioned in Example 3, (5% by weight MgO, 55% by weight Al₂O₃, 40% by weight TiO₂ where as for solid solution conditions 5% by weight MgO would require 42% by weight Al₂O₃ and 53% by weight TiO₂), secondary phases must inevitably be present since it is derivable from (12), Fig. 7 and 8 on page 219 and the compositions no. 4, 5, 10, 11, 12 and 13 mentioned in Table I on page 219 that under equilibrium conditions at temperatures between 1300 and 1550°C secondary phases of spinel, rutile or corundum exist in the concentrations indicated in Claim 1 of the patent in suit as well as in (1), col. 3, lines 11 to 15 and Example 3. The Board cannot see any plausible reason why such secondary phases should not also exist if such ceramic materials are formed under firing conditions which do not result in the phase distribution according to the thermodynamic equilibrium.

8. The Board further considers that (1) teaches that the CTE of the materials disclosed therein may be strongly negative. In this case it is generally recommended in (1), column 3 line 60 to column 4, line 3 to bring the CTE closer to zero by adding materials with a positive CTE, e.g. corundum which is exemplified in Example 3. It is true that the amount of added corundum in the example exceeds 20% by weight of the ceramic material, but this amount is only given as an example within the range indicated in Claim 4, thus demonstrating that also the presence in the ceramic material of phases having a higher CTE in amounts not exceeding 20% by weight is desirable. Therefore the Appellant's submission that a skilled person would not have considered a material containing secondary phases with a relatively high CTE when looking for a

material with good thermal shock resistance is not supported by this prior art.

9. In the Board's view however, 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, but must also 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 29 to 34) 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 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 lining of electrical 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 (9) reflecting the common general knowledge about aluminium-titanate ceramics. In the first complete paragraph on p. 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 SiO_2 is required in order to obtain materials with sufficient physical strength. A person skilled in the art therefore would not have considered to use aluminiumtitanate ceramics free of SiO_2 for purposes where high physical strength is important. In document (9), issued in 1974, it is also stated in the abstract that the high anisotropy of the CTE of aluminiumtitanate is responsible for the weakness of the structural system of ceramic materials containing it. This weakness which had not yet been overcome was said to be the reason why such materials have not yet gained technical importance. In this article therefore the addition of ZrSiO_4 is recommended in order to obtain ceramic materials with practically useful properties.

Document (8) is a scientific document reporting the phase diagram and phase equilibria of the system $\text{MgO-Al}_2\text{O}_3\text{-TiO}_2$ and does not relate to any industrial application.

10. Therefore in the Board's view the Respondents' 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 Claim 1 of the patent in suit 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 SiO₂ 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 Claim 1 of the patent in suit must be regarded as based on an ex-post-facto analysis of the prior art and hence must fail.

11. As Claims 2 to 4 relate to preferred embodiments of the subject-matter of Claim 1, they derive their patentability from this claim.

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 in amended form as submitted at the oral proceedings.

The Registrar

The Chairman

F.Klein

K.Jahn