BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

A		B		C	X

File Number: T 541/90 - 3.3.2

Application No.: 84 307 058.2

Publication No.: 0 140 638

Title of invention: High-strength zirconia type sintered body and process for preparation thereof

Classification: C04B 35/48

D E C I S I O N
of 10 September 1992

Proprietor of the patent: Tosoh Corporation

Opponent: Stora Feldmühle AG

Headword: Sintered body/TOSOH

EPC Article 56

Keyword: "Inventive step (denied) - obvious modification"

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number : T 541/90 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 10 September 1992

Appellant : (Proprietor of the patent)
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Representative :

Decision under appeal :
Decision of Opposition Division of the European Patent Office dated 17 May 1990 revoking European patent No. 0 140 638 pursuant to Article 102(1) EPC.

Composition of the Board :

Chairman : P.A.M. Lançon
Members : I.A. Holliday
R.L.J. Schulte

Summary of Facts and Submissions

I. European patent No. 0 140 638 was granted on the basis of sixteen claims contained in European patent application No. 84 307 058.2.

II. The Respondent filed a notice of opposition against the European patent requesting revocation of the patent on the grounds that its subject-matter lacked inventive step. The following documents were cited:

(1) DE-A-3 300 211
(2) DE-A-3 027 401
(3) DE-A-2 744 700
(4) JP-A-79-25908 (English translation)
(5) Thermal and mechanical properties of Y_2O_3- stabilized tetragonal zirconia polycrystals, K. Tsukuma et al. Paper presented at conference "Zirconia '83" at Stuttgart in June 1983.

III. The patent was revoked by the Opposition Division on the grounds that both the main and auxiliary requests failed to meet the requirements of Article 56 EPC.

In the view of the Opposition Division, the closest prior art was document (5), according to which sintered yttria stabilised tetragonal zirconia bodies (Y-TZP) were prepared by a preliminary sintering followed by hot isostatic pressing (HIP) in analogous manner to the process of the patent in suit. In a typical example, a three-point bending strength of 1700 MPa was obtained for a specimen containing 3 vol% yttria.

The zirconia bodies of the patent in suit differ only in the presence of minor amounts of alumina, spinel or mullite. It was, however, known from document (1) that the

bending strength of Y-TZP could be further improved by addition of small amounts of alumina, although not employing HIP. It was thus the opinion of the Opposition Division that the subject-matter of the patent in suit could be derived in an obvious manner from the teachings of document (5) and document (1).

IV. The Appellant lodged an appeal against the decision of the Opposition Division. The arguments of the Appellant, both in the written procedure and at the oral proceedings on 10 September 1992, may be summarised as follows.

Two tables of comparative data were filed. Table A was filed to illustrate the exceptionally high bending strength values obtained when employing the process of the patent in suit in comparison with those recorded in document (1). Table B showed that, with normal sintering, the bending strength of Y-TZP fell when the content of alumina was increased.

The Appellant sought to introduce a further reference: Lange, Journal American Ceramic Soc., Vol. 66, 1983, pp. 396-398 (6). Attention was drawn to a passage which taught that the flexural strength of a $ZrO_2/Y_2O_3/Al_2O_3$ ceramic body using presintering followed by HIP was less than that achieved by hot pressing. It was argued that the skilled man would be led away from incorporating alumina into a Y-TZP body when seeking to enhance the bonding strength.

With the statement of appeal the Appellant requested, as a main request, maintenance of the patent with the claims as granted, i.e. claims to a product and process for its preparation. As a first auxiliary request (claims of the second preference), maintenance was requested on the basis of process claims only, which were based essentially on a

combination of granted Claims 1 and 4. As a second auxiliary request, the Appellant requested maintenance based on a claim for the preparation of bodies having a bending strength of at least 2000 MPa in which the ranges of zirconia, yttria and alumina were all narrower than those specified in the first auxiliary request.

With a fax communication dated 9 September 1992, the Appellant filed claims of the fourth preference which related to a process with even narrower ranges of zirconia, yttria and alumina. At the oral proceedings, the Appellant apologised for the late filing of these claims but argued that they were an attempt to meet the arguments of the Respondent (see point V below). The claims were accompanied by Fig. A which was intended to demonstrate that bodies prepared within the ranges specified in the newly filed claims had, in general, a three-point bending strength of at least 2000 MPa.

The Board having signalised agreement to the filing of the said claims of the fourth preference, the Appellant withdrew the first three requests mentioned above.

V. At the oral proceedings, the Respondent argued that document (6) should be considered closest state of the art since it was concerned with the preparation of ceramic bodies based on zirconia, yttria and alumina in which HIP was employed. In particular reference was made to Figure 1 which plotted the flexural strength in relation to the zirconia content.

The Respondent also referred to documents document (2) to document (5), each of which indicated that HIP was a well established process at the priority date of the patent in suit; document (4) in particular expressed a preference

for HIP since the process was not restricted to the pressing of simple forms.

The Respondent's technical expert, Dr. Burger, supplied a formula for converting the volume per cent of document (6) into weight percentages used in the claims of the patent in suit. The said conversion formula was not disputed by the Appellant.

In the written procedure, the Respondent supported the decision of the Opposition Division in that the data for the hot pressed bodies of document (1) were a more realistic comparison than those prepared according to Table B by sintering without pressure. The Respondent also questioned the Appellant's interpretation of document (6), arguing that the test results were not directly comparable.

The Respondent also supplied a copy of the Tables occurring on pages 7 to 23 of the granted patent which indicated how many of the original test samples, a number of which had bending strengths over 2000 MPa, fell outside the scope of the claims of the third preference.

A corresponding objection was also made at the oral proceedings when the Respondent drew attention to the fact that a considerable number of the samples recorded in Table A, which fell outside the scope of the request currently on file, also appeared to have satisfactory values of bending strength.

VI. Claim 1 filed on 9 September 1992 reads as follows:

"1. A process for the preparation of a zirconia type sintered body, said sintered body having a three-point bending strength of at least 2000 MPa, the average grain

size of the crystal of the sintered body being not larger than 2μm, the bulk density of the sintered body being at least 99.5% of the theoretical density, and the sintered body not containing therein pores having a size of 30μm or more, the process comprising moulding a powdery composition consisting essentially of 70 to 85% by weight of (a1) a powder of zirconia containing 2.5 to 3 mole % of yttria or (a2) a mixture comprising 2.5 to 3 mole % of a yttria powder and 97 to 97.5 mole % of a zirconia powder and 15 to 30% by weight of (b) a powder of an alumina type oxide selected from alumina, an alumina-magnesia type oxide, alumina-silica type oxide, a mixture of alumina and magnesia and a mixture of alumina and silica, subjecting the moulded powdery composition to preliminary sintering at a first temperature and isostatically hot-pressing the sintered composition under a pressure of at least 50 MPa at a second temperature of 1300°C to 1500°C, the first temperature being up to 1500°C."

VII. The Appellant requested that the decision of the Opposition Division be set aside and the patent maintained on the basis of Claims 1 to 13 filed on 9 September 1990.

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

2. The Appellant explained at the oral proceedings and the Board accepted that the new claims filed on 9 September 1992 were intended to meet the objections raised by the Respondent to the claims of the third preference filed on 27 September 1990. Notwithstanding the late filing of the said new claims, the Board decided to allow them into the

proceedings. Having regard to its relevance as shown under point 5 below, the Board also decided to admit document (6).

3. The reference in the new Claim 1 to a three-point bending strength of at least 2000 MPa has basis in the printed patent specification on p. 6, line 22 (p. 14, lines 4-6 as originally filed). In relation to the higher bending strength now claimed, the Board is satisfied that the new ranges have adequate support. The scope of the new claims is narrower than that of the claims as granted. The requirements of Articles 123(2) and (3) are accordingly satisfied.

4. The Board can accept the argument of the Respondent that document (6) represents the closest state of the art in relation to the product claims which were still on file as the main request when the Respondent prepared arguments to be presented at the oral proceedings. Document (6) is concerned with compositions in general containing a major proportion of alumina together with minor amounts of zirconia/yttria and the preparation of ceramic bodies therefrom employing inter alia sintering and HIP. However, document (5) relates to a process very similar to that of the patent in suit and, with regard to the present request is to be considered, in the view of the Board, as the closest state of the art.

4.1 The authors of the paper document (5) are the inventors of the patent in suit which can thus be considered as part of the same research programme. Document (5) is a study of yttria stabilised tetragonal zirconia (Y-TZP) in which it is shown that three-point bending strengths of up to 1700 MPa may be obtained by using 3 mole % yttria. The products are prepared by pressureless sintering at temperatures of 1400-1600°C followed by HIP also at 1400-1600°C, i.e. by

essentially the same process steps as those employed in the patent in suit. It is shown that the final bending strength depends on the initial sintering temperature as well as the yttria content and the isostatic pressing temperature. It was apparent that HIP was instrumental in obtaining the high bending strength values of up to 1700 MPa.

4.2 In relation to document (5), the problem to be solved lies in the preparation of ceramic bodies having an even higher three-point bending strength.

4.3 The solution rests in the addition of minor amounts of alumina to the Y-TZP compositions disclosed in document (5) prior to sintering and HIP. Having regard to the worked examples of the patent in suit, Table A presented during the opposition procedure and Figure A filed on 9 September 1992, the Board is satisfied that addition of alumina has indeed solved the problem.

5. The Board is satisfied that the subject-matter currently claimed is novel. The composition known from document (6) which comes closest to that obtained according to the process of the patent in suit contains the lowest amount of alumina, i.e. 50 vol% together with 50 vol% zirconia/yttria. Using the conversion equation presented by the Respondent at the oral proceedings, 50 vol% alumina corresponds to approximately 40 weight %. Thus the said composition known from (6) would apparently have anticipated the product claims of the original main request and the process for their preparation. However, this does not apply to the process presently claimed wherein the addition of alumina is restricted to a maximum of 30% by weight.

5.1 The compositions of (5) do not contain alumina and thus, as far as novelty is concerned, the corresponding processes are further from the subject-matter of the patent in suit.

5.2 It is to be noted that novelty has not been disputed by the Respondent.

6. It remains to consider whether or not the subject-matter of Claim 1 satisfies the requirements of Article 56 in respect of inventive step.

6.1 The process of the patent in suit is very close to that described in document (5) in that a mixture of zirconia and yttria is subjected to a preliminary sintering at up to 1500°C, followed by HIP at a pressure of at least 50 MPa and a temperature of 1300-1500°C. In accordance with the claims now on file, the process differs in that the initial mixture also contains alumina. The said mixture consists essentially of 70-85% by weight of zirconia containing 2.5 to 3 mole % yttria and correspondingly 30-15% by weight of alumina, alumina/magnesia (spinel) or alumina/silica (mullite); the majority of the worked examples use alumina without silica or magnesia. It is apparent from the worked examples and from Table A filed by the Appellant on 9 September 1992 that, when working within the prescribed ranges of yttria and alumina, ceramic bodies can be obtained having three-point bending strength of 2000 MPa or more. This compares with bending strengths of up to 1700 MPa recorded in (5) in the absence of alumina.

6.2 According to Table 4 on p. 13 of the patent in suit, the results of which are also recorded in Table A filed by the Appellant, samples of zirconia containing 2 mole % yttria with increasing contents of alumina are subjected to

sintering at 1400°C without any further treatment. The results show that with increasing content of alumina the three-point bending strength declines from 1070 MPa in the absence of alumina to 810 MPa with a 40% by weight alumina content. The Appellant has argued that this teaches away from the process of the patent in suit.

6.3 On the other hand, employing the hot pressing process of document (1) in which samples are sintered at 1500-1600°C in a carbon mould under a pressure of 200 kg/cm^2 (19.6 MPa), increasing the content of alumina leads to an increase in the bending strength. Thus, a sample of zirconia containing 2 mole % yttria shows a bending strength of 40.1 kg/mm^2 (393.2 MPa) with an alumina content of 0.1% by weight which rises to 133.0 kg/mm^2 (1304 MPa) with an alumina content of 10% by weight and 162.3 kg/mm^2 (1592 MPa) with 40% by weight alumina (see Table 2 on p. 13 of (1)). The above conversions are made by the Board on the assumption that kilogram force (kgf) is intended. In other words when sintering takes place under pressure (hot pressing) the inverse effect is observed to that obtained in the pressureless sintering experiments carried out by the Appellant.

6.3.1 According to document (3) sintered bodies are prepared inter alia from alumina containing 1-50 vol% of zirconia. On p. 7, lines 20-21 both hot pressing and HIP are mentioned as alternative processes for their preparation. Document (4) relates to the preparation of ceramic bodies of alumina with minor amounts of zirconia. The paragraph bridging pages 6 and 7 expresses a preference for presintering followed by HIP; such a process forms the subject-matter of Claim 3.

6.3.2 The process of document (2) relates to the preparation of ceramic bodies from alumina and titanium nitride mixtures.

In the first complete paragraph on page 14, hot pressing and presintering followed by HIP are described as alternative processes for the manufacture of the products. Some preference for HIP is expressed in the final paragraph on p. 15 and Table I on p. 20 shows the low porosity obtained when using HIP.

6.3.3 Documents (3) and (4) were published in 1979 and (2) in 1980. It is accordingly clear that at the first priority date of the patent in suit (17 October 1983) both hot pressing (sintering under pressure) and pressureless sintering followed by HIP (the pressure generated under an inert gas) were well known alternative methods for the manufacture of ceramic bodies. This was not disputed by the Appellant at the oral proceedings. The documents cited tend to show a preference for the use of HIP.

6.4 The Appellant has relied on the passage under III in document (6) where it is stated that isostatically hot pressed (HIP) material was still weaker than the hot pressed material. A footnote is attached to indicate that the zirconia in the hot pressed material contained 2 mole % yttria whilst the zirconia in the HIP material contained 3 mole % yttria. It was made clear at the oral proceedings that the resultant bending strength depended not only on whether hot pressing or HIP was used but also on the yttria content and on the initial particle size. It is accordingly the Board's view that the samples in document (6) are not directly comparable and furthermore that the above statement cannot be taken as a generality that all samples prepared by hot pressing would have higher bending strength values than corresponding samples prepared by presintering and HIP.

6.5 In summary, the prior art, whilst expressing some preference for HIP, treats hot pressing and HIP as

equivalents. Accordingly, the Board is inclined to follow the view of the Opposition Division that the hot pressed products of document (1) are a more reliable guide to the influence of alumina in yttria stabilised zirconia bodies than the pressureless sintered experiments recorded in Table A. Thus, the skilled person wishing to solve the problem underlying the patent in suit and knowing that alumina had a positive influence on the bending strength of the hot pressed bodies prepared according to document (1), would have the incentive to try if the addition of alumina would also increase the bending strength of bodies prepared by the HIP process of document (5).

6.6 The Board cannot see any basis for an inventive selection based on the ranges now claimed. Had the samples within the new range of alumina content shown exceptional bending strengths and those outside the range notably inferior bending strengths, there might have been a basis for such a selection. However, as pointed out by the Respondent at the oral proceedings this was not the case. It is to be noted that very few of the worked examples which appear in the granted patent specification fall within the scope of the present Claim 1 and also that many examples which are outside the scope show bending strengths considerably higher than the prescribed minimum of 2000 MPa.

6.7 It follows from the preceding paragraphs that the subject-matter of Claim 1 lacks inventive step. The same applies to Claims 2 to 6 which relate to the preferred embodiments of the process of Claim 1.

6.8 Although not the subject of any separate request, the Board cannot recognise an inventive step in the coprecipitation process claimed to prepare the oxide mixtures in the independent Claim 7. It is apparent from the prior art that such coprecipitation is a recognised

technique, mentioned, for example, in the Experimental Procedure of document (5) and also in Example 2 of document (1). In analogous manner, Claims 8-13 relating to preferred embodiments of the process of Claim 7 also lack inventive step.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

P. Martorana P. Lançon