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BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

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File Number: T 206/91 - 3.3.2

Application No.: 83 106 245.0

Publication No.: 0 097 944

Title of invention: Method for directly bonding ceramic and metal members and laminated body of the same

Classification: C04B 37/02

D E C I S I O N
of 8 April 1992

Applicant: KABUSHIKI KAISHA TOSHIBA

Opponent: 01) ASEA BROWN BOVERI AKTIENGESELLSCHAFT
02) DODUCO KG DR EUGEN DÜRRWÄCHTER

Headword:

EPC Art. 56

Keyword: "Inventive step (no) - obvious solution"

Headnote

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number : T 206/91 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 8 April 1992

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Decision under appeal :
Decision of Opposition Division of the European Patent Office dated 10.10.190 posted on 06.02.1991 revoking European patent No. 0 097 944 pursuant to Article 102(1) EPC.

Composition of the Board :

Chairman : P.A.M. Lançon
Members : A.J. Nuss
F. Benussi

Summary of Facts and Submissions

I. European patent No. 0 097 944 was granted with eight claims on European Patent application No. 83 106 245.0.

II. The two Respondents (Opponents I and II) filed notices of opposition against the European patent. Fourteen prior art documents were cited of which the following remain relevant in the present appeal:

(1) US-A-3108887

(2) US-A-3994430

(11) "New Hybrid Power Technique Utilizing a Direct Copper to Ceramic Bond", Sun and Driscoll, IEEE Transactions on Electronic Devices, Vol. ED23, no. 8, August 1976, pages 961 to 967.

III. The Opposition Division revoked the patent on the grounds of lack of inventive step. Having regard to document (2) disclosing in general direct bonding of a ceramic member to a metal member with the aid of a eutectic melt, it was the Opposition Division's view that although the examples according to this prior art did relate to oxide ceramics, a person skilled in the art would expect that the method known from (2) could also be applied to nonoxide type-ceramic materials which in their normal state are covered with a thin oxide layer. It was then obvious to form a sufficiently thick oxid layer on the surface of a nonoxide type-ceramic member prior to bonding, with the aid of a method described in document (1). The Opposition Division also took the view that document (11) disclosed a process according to the pre-characterising portion of Claim 1 of the patent-in-suit and it was suggested in this document that SiC may be bonded to Cu with the aid of a Cu-Cu_2O eutectic melt. Finally, an auxiliary petition claim filed

by the Proprietor during oral proceedings was regarded as being obvious in the light of the cited prior art.

IV. The Appellant (Proprietor) lodged an appeal against this decision which was supported by experimental data in the form of a diagram showing a relationship between the peeling strength and the thickness of aluminium oxide formed on an AlN ceramic member.

The arguments of the Appellant, both in the written procedure and at the oral proceedings on 8 April 1992 may be summarised as follows.

None of the cited documents (1), (2) and (11) dealt with the actual object of the invention, namely directly bonding a nonoxide-type ceramic member to a metal member.

Even when accepting that the closest prior art, i.e. document (11) disclosed a laminated body comprised of a SiC member and a Cu member, this document could not, however, be considered to teach how to employ a Cu/Cu_2O-eutectic to successfully bond copper to SiC ceramic. Starting from the general teaching of (11), in particular when following the scheme according to Figure 4 and the proposed temperatur of 1065°C, it would not be possible to produce a laminated body by directly bonding a nonoxide-type ceramic member to a metal member as requested by the present invention. In addition to those measures it was actually necessary to take into account the different characteristics of oxide ceramics and nonoxide-type ceramics and to recognize that it was essential to treat the surface of the nonoxide-type ceramic member with oxygen as bonding agent before carrying out the direct bonding between the ceramic member and the metal member. The experimental data attached to the grounds of appeal constituted sufficient evidence to show the importance of

the presence of an oxide layer on the nonoxide ceramic member for improving the peeling strength.

A combination of the teachings of (1) with (2) would also not lead in an obvious manner to the method of Claim 1 of the patent-in-suit.

Document (1) disclosed nothing more than a method for the formation of a thin oxide layer on aluminium nitride to prevent hydrolysation of the material. Since document (2) proposed three different ways of directly bonding a ceramic member to a metal member whereby only one of them related to the possibility of depositing the bonding agent, or a compound of the bonding agent and the metal, in a particulate form on either the substrate or the metal, there was no reason for a person skilled in the art to stabilize the ceramic substrate with a thin oxide layer by a heat-treatment step according to document (1).

In addition, it was to be noted that the prior art as a whole and in particular (11) would rather propose as a general rule to oxidize the surface of the metal member.

At the oral proceedings, the Appellant deleted all product claims, i.e. Claims 5 to 8 as granted, in order to avoid a novelty objection in regard of the disclosure of (11).

V. At the oral proceedings before the Board Respondent I stressed that those skilled in the art were familiar with the so-called DCB-(direct-copper-bonding-) process and that it was well-known in that art that both the wetting conditions and the thickness of the oxid layer on the surface of the ceramic member played an important role in the binding process. It was therefore not necessary to have an explicit reference to nonoxide-type ceramics in document (2) or (11). Since it was furthermore well-known

that nonoxide-type ceramic bodies normally had thin oxide layers and document (1) provided a clear teaching as to how oxide layers on ceramic members could be formed, it was merely a matter of optimization, when starting from the disclosure in (11), to form a sufficient thick oxide layer on the nonoxide-type ceramic material as presently claimed. The obviousness of the claimed solution was further reinforced, as it was also well-known that even at room temperature in the presence of atmospheric oxygen an oxide layer having a thickness within the claimed range would be formed on pure Si. In addition, the Respondent expressed the opinion that oxidizing conditions at room temperature were not excluded by the wording of Claim 1.

VI. Respondent II took the view that there was no need to comment on the grounds of appeal. As announced in a letter dated 17 March 1992, he did not attend the oral proceedings.

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 4 as granted, and, as an auxiliary request that the patent maintained on the basis of the single claim filed during oral proceedings.

The Respondents requested that the appeal be dismissed.

VIII. (i) Claim 1 according to the present main request reads as follows:

"1. A method of directly bonding a nonoxide-type ceramic member to a metal member with the aid of a bonding agent, which comprises heating said nonoxide-type ceramic member and said metal member while said nonoxide-type ceramic member is in contact with said metal member through said bonding agent at a temperature below the melting

temperature of the metal of said metal member and above the eutectic temperature of said metal and said bonding agent, characterized in that said heating step is preceded by a step of oxidizing a surface of said nonoxide-type ceramic member for the formation of an oxide layer containing the required oxygen as said bonding agent."

Dependent Claims 2 to 4 concern preferred embodiments of the method claimed in accordance with Claim 1.

(ii) The single claim according to the auxiliary request differs from Claim 1 of the main request in that the characterising portion reads as follows:

"... characterized in that said heating step is preceded by a step of oxidizing a surface of said nonoxide-type ceramic member in an air atmosphere at a temperature of 1000 to 1400°C or in a wet gas atmosphere at a temperature of 1200 to 1500°C for the formation of an oxide layer containing the required oxygen as said bonding agent, said oxidizing step being performed to such an extent as to form an oxide layer of about 20 micrometers or 10 micrometers in thickness."

Reasons for the Decision

1. The appeal is admissible.

2. Since Claims 1 to 4 of the main request correspond to unamended Claims 1 to 4 as granted and the single claim according to the auxiliary request is supported by Claims 1 to 4 as granted (correspond to Claims 1 to 4 originally filed) and column 3, lines 21 to 23 of the patent-in-suit (corresponds to page 4, lines 27 to 30 of the original description), the requirements of Article 123(2) and 123(3) EPC are satisfied.

3. The patent-in-suit relates to a method of directly bonding a nonoxide-type ceramic member to a metal member.

3.1 It was undisputed during the oral proceedings that the closest prior art is document (11). This document discloses a method of bonding copper directly to ceramic employing the copper-copper oxide eutectic melt to wet both the copper and the ceramic to form a strong bond after cool-down (cf. page 962, left column, second and third paragraph and Fig. 4 on page 963). According to the process sequence in Fig. 4, a Cu foil is laid on top of the ceramic and the assembly is heated up to a temperature around 1070°C in an atmosphere which contains primary N_2 with a small addition of O_2 (cf. page 963, left column last paragraph up to right column first paragraph). This prior art document also provides some theoretical background information about the direct copper bonding process, namely a discussion of the phase diagram of the Cu-O system, the Cu-Cu_2O eutectic liquid as a function of temperature and interfacial surface energy between copper and alumina as a function of oxygen content in copper (cf. page 962, left column, last paragraph up to page 963, second paragraph and Fig. 1 to 3). It is then summarized on page 967 without any reference to a concrete working example that the said Cu_2X eutectic (where X can be O, S or Te) has been successfully employed to bond copper to SiC, a nonoxide-type ceramic member.

It has been not contested by the parties during oral proceedings that it is common practice to use oxygen for the formation of the eutectic when bonding to ceramics and that the resulting Cu_2O eutectic (see above) has a poor wettability with a nonoxide-type ceramic such as SiC and thus forming a strong bond between said ceramic member and

Cu will be difficult. Confirmation of this can also be found in the patent-in-suit, see in particular column 1, lines 46 to 64.

3.2 In the light of said prior art, the technical problem underlying the patent-in-suit can be seen in providing a strong bond between a nonoxide-type ceramic member and a metal member by directly bonding the metal to said ceramic.

3.3 In order to solve this problem, Claim 1 of the main request as well as the single claim of the auxiliary request proposes to modify the direct bonding procedure in such a way that, in essence, it is preceded by a step of oxidizing a surface of the nonoxide-type ceramic member for the formation of an oxide layer containing the required oxygen as bonding agent.
Having regard to the comparative experiments appearing in the patent-in-suit, the Board is satisfied that the problem has been solved.

4. After examination of the prior art, the Board has come to the conclusion that the claimed subject-matter is novel. Since novelty is no longer disputed, there is no need to further deal with this matter.

5. It remains to consider whether or not said solution satisfies the requirements of Article 56 EPC in respect of inventive step.

5.1 According to present Claim 1 the bonding process comprises a heating step at a temperature below the melting temperature of the metal to be bonded and above the eutectic temperature of said metal. As stated above, document (11) describes such a process by utilizing an oxygen copper eutectic melt which is regarded to wet and

bond to the ceramic. Having regard to Figure 1 to Figure 3 and corresponding explanations, the skilled person would not only get detailed information about the phase conditions of the oxygen copper eutectic melt formed at 1065°C and the important role which oxygen plays as one of the components forming the eutectic composition, but also the further information that the absolute amount of oxygen is relevant in the overall bonding process. The Board agrees with Respondent I that in particular Figure 3, showing that the interfacial energy between molten Cu and alumina drops drastically with the introduction of a small amount of oxygen, suggests to the skilled person that the bonding process is influenced by an interaction between oxygen and said ceramic material. Since in this context it is also stated that a mechanism for this decrease of energy has been suggested as the formation of a chemical bonding between the Cu_2O and the ceramic, those skilled in the art would infer from all this that the bonding agent, i.e. oxygen, must be compatible with both the metal and the ceramic substrate. Moreover, since in document (11) SiC has been directly bonded to Cu by employing an oxygen copper eutectic melt, there is no reason why the skilled reader of this document - although aware of the fact that the theoretical background discussed therein is based on alumina, an oxide-type ceramic - should not expect that this technique could be applied to ceramics in general, thus also to nonoxide-type ceramics, which in their normal state, are covered with a naturally formed oxide layer. As regards the thickness of such a thin oxide layer, the Respondent has argued at the oral proceedings that even at room temperature for example silicon has an oxide layer of about 100Å, a thickness which clearly falls within the range of 0.001 μm or more as required in accordance with the teaching of the patent-in-suit (see col. 3, lines 24 to 26). This was not contested by the Appellant.

5.2 In view of the above considerations, the Board endorses the view of the Opposition Division that there is a strong incentive for the skilled person to apply the direct copper bonding technique to nonoxide-type ceramics. The first instance came to this appreciation on the basis of the disclosure contained in document (2). The latter document indeed contains theoretical background information about the oxygen copper eutectic melt and the wetting conditions of the members to be bonded and further describes a practical way to carry out the direct copper bonding technique in the case of a ceramic substrate. Emphasis should be put on the fact that it is expressis verbis stated as a theory "that there must be a potentially stable compound of the substrate and eutectic and that if such a compound is not formed, there will be no tenacious bond." The whole of the following theoretical discussion is based on the assumption "that the bonding agent selected is compatible with the metal and the substrate" (cf. col. 2, line 32 up to col. 4, line 9 and Fig. 2 to Fig. 4). Although document (2) does not expressly mention to apply the direct bonding technique to nonoxide-type ceramics, it is clearly stated there that "the term ceramics is to be construed broadly" (see col. 4, lines 67/68). Therefore, the man skilled in the art had no reason to interpret the teaching of this document as being limited to oxide-type ceramics.

5.3 Furthermore, even if one assumes that by following the teaching in document (11) in order to try solve the present problem the skilled worker would fail to produce a laminated body having the desired strong bond between a particular non-oxide ceramic and a copper member as a result of bad wetting of the oxygen copper eutectic, the preceding findings show that he would realize that strong bonding can only be achieved if there is sufficient natural oxide available on the nonoxide-type ceramic. It

is readily comprehensible that there is a strong affinity of the Cu_2O in the eutectic mixture to the oxidized surface of the ceramic and thus, for energetic reasons, the thickness of the oxide layer on the surface of the ceramic influences the strength of the bond which the man skilled in the art estimates for example by measuring the peel strength of the laminated body. In other words, in the light of the prior art, the skilled worker would come to the conclusion that it is exclusively said oxide layer which makes the ceramic material compatible with the eutectic liquid. Therefore, before carrying out the direct bonding procedure he would make sure that the oxide layer on the surface of a nonoxide-type ceramic member is of such a thickness that a strong bond is formed. If need be, the skilled person could find in document (1) the necessary information, so that there are no technical difficulties to form a sufficient thick oxide layer on a nonoxide-type ceramic (see in particular col. 3, lines 46 to 55 and Table I).

The Appellant has argued that document (1) only discloses a method for the formation of a thin oxide layer against hydrolysation and should therefore not combined with the teaching of documents (11) or (2). Although it is true that document (1) relates to a process for stabilizing aluminium nitride by a specific heat treatment, the Board, however, cannot accept the Appellant's submission because it is clear from the preceding paragraphs that it is not even necessary to refer to document (1) to establish obviousness of Claim 1. For the rest, this prior art confirms that the step of oxidizing a surface of a nonoxide-type ceramic material is a trivial measure.

5.4 In view of the fact that Claim 1 according to Appellant's main request does not define any specific oxidizing conditions, the Respondent has argued that the process

feature "that said heating step is preceded by a step of oxidizing a surface of said nonoxide-type ceramic member" could also be regarded as being achieved by a normal oxidizing process at room temperature which naturally occurs without any specific measures to be taken by a skilled person. The Appellant has not contested this interpretation of the wording of Claim 1.

5.5 It is true that the single claim according to the auxiliary request contains such process parameters (see point VIII (ii) above). However, in the opinion of the Board no unexpected effect has been disclosed or put forward by the Appellant in connection with a preoxidizing step carried out in an air atmosphere at a temperature of 1000 to 1400°C or in a wet gas atmosphere at a temperature of 1200 to 1500°C and performed to such an extent as to form an oxide layer of about 20 micrometers or 10 micrometers in thickness. Indeed, as only evidence in favour of a surprising effect, the Appellant has submitted with the grounds of appeal experimental data summarized in Figure 2 of an "explanation sheet" showing a relationship between the peeling strength and the thickness of the aluminium oxide layer formed on an AlN ceramic member bonded to a copper member. It is however to be noted that for the said discrete values of 20 μm and 10 μm the peeling strength is extremely low; they clearly fall outside the most suitable thickness range apt to provide a strong bond as required in the patent-in-suit (see col. 2, lines 42 to 49). Consequently, the oxidizing conditions mentioned in combination with these two particular thickness values are nothing else than a series of arbitrary features because it is not credible that they contribute to the solution of the underlying technical problem. The said features are therefore of no relevance for the question of inventive step (see decision T 37/82, OJ EPO 1984, 71). Under these circumstances, the

considerations made in connection with Claim 1 of the main request fully apply to the single claim of the auxiliary request.

5.6 For the above reasons the subject-matter of Claim 1 of the main request as well as that of the single claim according to the auxiliary request lacks inventive step.

5.7 Dependent Claims 2 to 4 of the main request must fall with Claim 1, since the main request can only be considered as a whole.

Consequently, both requests must be rejected.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar

P. Martorana

The Chairman

P.A.M. Lançon