DECISION
of 12 December 2001

Case Number: T 0160/98 - 3.4.3
Application Number: 89301057.9
Publication Number: 0331292
IPC: H01L 39/12

Language of the proceedings: EN

Title of invention:
Oxyde superconductive material

Patentee:
MATSUSHITA ELECTRIC INDUSTRIAL CO., LTD.

Opponent:
HOECHST AG

Headword:
-

Relevant legal provisions:
EPC Art. 100(b), 54, 56
EPC R. 64(b)

Keyword:
"Admissibility of the appeal (yes);"
"Main request and auxiliary request 1: sufficiency (no);"
"Auxiliary request 2: sufficiency (yes); novelty and inventive step (yes)"

Decisions cited:
T 0409/91, T 0435/91

Catchword:
-
Case Number: T 0160/98 - 3.4.3

DECISION
of the Technical Board of Appeal 3.4.3
of 12 December 2001

Appellant: HOECHST AG
(Opponent) Zentrale Patentabteilung
D-65926 Frankfurt am Main

Representative: Ahrens, Gabriele, Dr.
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Respondent: MATSUSHITA ELECTRIC INDUSTRIAL CO., LTD
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Composition of the Board:
Chairman: R. K. Shukla
Members: M. Chomentowski
          M. J. Vogel
Summary of Facts and Submissions

I. The European patent No. 0 331 292 was granted on the basis of the European patent application No. 89 301 057.9, filed on 3 February 1989 and claiming a priority of three applications 26128/88, 26129/88 and 26130/88 filed on 5 February 1988 in Japan.

II. An opposition was filed against the patent. During the opposition proceedings, the following documents filed by the opponent were particularly taken into consideration:


E2: "New High-Temperature Oxide Superconductor Has Been Found That Has Excellent Characteristics and Does Not Contain any Rare Earth Element", press release of the National Research Institute for Metals (NRIM), Japan, January 21, 1988;

E3: Magazine "Nikkei Chodendo", January 25, 1988, page 1;

E4 "UH Physicists Produce Superconductor Without Rare Earth Element", press release of the University of Houston, USA, January 25, 1988;


E12: Science, Volume 239, 26 February 1988, Report pages 1015 to 1017; M. A. Subramanian et al.: "A new high-Temperature Superconductor:
III. The opposition division maintained the European patent in amended form according to the auxiliary request of the patent proprietor. Claims 1 and 2 of the auxiliary request had the following wording:

"1. A superconductive oxide material which substantially consists of a single phase ABiCuO, where A is a mixture of at least one of Mg and Ca an at least one of Sr and Ba, the ABiCuO forming a crystal phase in which the atomic ratio of A/Bi/Cu is substantially 3/2/2 and the material having a superconductive transition temperature above 80K."

"2. A multi-layered superconductive oxide material comprising artificial alternate layers of a superconductive oxide material according to claim 1, and a crystal phase consisting of ACuO, where A has the meaning stated in claim 1 and the atomic ratio of A/Cu is substantially 1/2".

IV. Concerning the maintenance of the patent as amended, it was argued essentially as follows in the decision of the opposition division:
Sufficiency (Article 100(b) EPC)

The description of the patent in suit discloses at least one example for making a substantially single phase high-Tc ABiCuO superconducting material by indicating the starting composition and the appropriate calcination and sintering temperatures (see in particular compositions 1 and 9 in Table 2 and page 4, lines 41 to 44).

This point of view is confirmed by the documents E14 (see page 7, left-hand column, lines 14 to 17 and right-hand column, paragraph 2), or E17 (see page 9382, right-hand column, last paragraph), taken as an expert's opinion.

Although the duration of the heating steps is not explicitly mentioned in the description of the contested patent, it is clear that the skilled person would apply usual heating periods of the order of one to several days to produce superconductive oxide materials such as the ABiCuO materials (see page 8, Table 3 and page 4, Table 2 of document E14).

Concerning the opponent's comparative experiments, it is to be noted that only the third example (1/2) was relevant to claim 1. According to the opponent's assessment, the material contained only minor portions of other phases, although the sintering time was as short as 6 hours (see the table annexed to the letter dated 23 August 1997). This comparative experiment is therefore not in contradiction to the findings of the patent in suit.

Therefore, the invention in the patent in suit is
disclosed in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art.

Novelty and inventive step

The subject-matter of claim 1 is novel over the prior art documents E1 to E5 because a superconductive material consisting substantially of a single 3/2/2 ABiCuO crystal phase is nowhere disclosed or directly and unambiguously derivable therefrom.

Document E5, which is a European patent application claiming a priority anterior to the priority of the patent in suit is a prior art document in the sense of Article 54(3) EPC and is relevant only for novelty (Article 56 EPC).

The selection of a substantially 3/2/2 ABiCuO composition as defined in claim 1 is not suggested in any of the cited prior art documents so that the subject-matter of claim 1 involves an inventive step.

Claim 2 defines a superconductive oxide material which comprises the single phase material of claim 1 in the form of an artificially produced layer of a multilayered structure. In the patent in suit, the artificially produced 3/2/2 material layer is fabricated by use of a specific step such as magnetron sputtering, causing preferred orientation of the 3/2/2 material in the form of a thin film. Such a specific step is neither disclosed nor fairly suggested by the available prior art documents, so that the multilayered structure of claim 2 is therefore novel and inventive.
V. The opponent lodged an appeal against this decision on 5 February 1998, paying the appeal fee on the same day. A statement setting out the grounds of the appeal was filed on 7 April 1998.

VI. Following the communication accompanying the summons to oral proceedings, the respondent (patent proprietor) filed on 3 December 2001 three sets of claims each set containing two claims. Claim 2 of all the requests are identical with claim 2 maintained by the decision of the opposition division.

Claim 1 of the main request differs from claim 1 maintained by the decision of the opposition division in that the expression "where A is a mixture of at least one of Mg and Ca an at least one of Sr and Ba" is replaced by

"where A is a mixture of:

(1) Ca and Sr and at least one of Mg and Ba, or

(2) Ca and Sr".

Claim 1 of the auxiliary request 1 differs from claim 1 of the main request in that the alternative (1) of A is replaced by

"(1) Ca, Sr, Mg and Ba, or"

In claim 1 of the auxiliary request 2, the alternative (1) of A of the main request is deleted, so that A is a mixture of Ca and Sr.

(Emphasis added by the Board).
VII. During the oral proceedings of 12 December 2001, the respondent filed new pages 2, 6 and 7 of the description according to the auxiliary request 2 and requested that the patent be maintained on the basis of any one of the following requests:

Main request and Auxiliary request 1:

Claims 1 and 2 according to the main or the auxiliary request 1, respectively, as filed on 3 December 2001;

Description:

Pages 1, 2 according to the main or the auxiliary request 1, respectively, as filed on 3 December 2001 and Pages 3 to 10 of the auxiliary request forming the basis of the decision under appeal;

Auxiliary request 2:

Claims 1 and 2 according to the auxiliary request 2 as filed on 3 December 2001;

Description:

Page 1 according to auxiliary request 2 as filed on 3 December 2001;

Pages 2, 6, 7 as filed on 12 December 2001; and

Pages 3 to 5 and 8 to 10 of the auxiliary request forming the basis of the decision under appeal.

VIII. The appellant (opponent) requested that the decision under appeal be set aside and the patent be revoked. He
argued essentially as follows in support of his request:

The appellant disputes that at the priority date of the patent in suit which was a few days after the first public announcement of the new high temperature Bi-Sr-Ca-Cu-O (= BSCCO) superconductor material by Maeda in Japan, it was possible for a person skilled in the art to produce the high temperature superconductor oxide materials forming the subject-matter of the patent in suit. The results of the comparative measurements supplied by the appellant during the opposition proceedings with the letter dated 23 August 1997 clearly demonstrate that under the conditions specified in the patent in suit it is not possible to reproduce a substantially single phase superconductive material as claimed.

Moreover, the new 322 phase is not sufficiently and correctly characterised in the X-ray data and its final composition is represented as Bi₂(Sr, Ca)₃Cu₂ without indicating the necessary range for the Sr:Ca ratio. Also, there is no indication regarding the content of oxygen in the composition, although it is generally known that the 322 phase of A BiCu exhibits high temperature superconductivity only at a specific minimum oxygen content which is higher than the normal stoichiometric oxygen content.

Claim 1 of the main request and the auxiliary request 1 include compositions containing Mg or/and Ba. It is however known in the art that replacement of Ca and Sr with Mg or Ba has negative influence on the high temperature superconductivity and their incorporation can lead to a total absence of superconductivity at a
high temperature.

The priority document (JP 63-26130) reports on page 7, first paragraph that a 322 phase was obtained over a wide composition range. The document however does not state that a substantially single 322 sample having a Tc of about 83 K was obtained. Consequently, the claims under consideration are not entitled to the priority date of 5 February 1988, and the published documents, in particular document E17 takes away the novelty of the subject-matter as claimed in claim 1 of all the requests.

The auxiliary request 2 is not sufficiently disclosed in the patent in suit. Thus, there is no information concerning the preparation of the materials, for instance the durations of different thermal treatments, or concerning the obtained materials, for instance the oxygen content or the crystallographic dimensions of the crystalline phase as well as the indications about the X-ray method used or about the error of measurement accepted in the measurement.

Moreover, this auxiliary request 2 is also not sufficiently disclosed in that sense that the limited information in the patent in suit, which concern only Example 1 of Table 2, would not lead to the claimed material, as can be seen from the technical documents presented, from the results of experiments provided during the opposition proceedings and from contradictory results reported for Example 7 of the priority document 26130/88, and Example 1 of Table 2 of the patent in suit, which concerns the same material as the Example 7.
These objections are particularly important in view of the breadth of the scope defined by claim 1; the information made available by the patent does not enable the skilled person to achieve the envisaged result within the whole ambit of claim 1 of the auxiliary request 2.

In any case, the subject-matter of claim 2 is anticipated by document E5, Example 1 of Table 1 thereof having the same composition as Example 3 of Table 3 of the patent in suit.

Therefore, the auxiliary request 2 is also to be rejected.

IX. The respondent's arguments in support of his requests can be summarized as follows:

The superconductive oxide materials according to the patent in suit belong to the general technical field of ceramics which have been prepared already for decades and, as single phase superconductive materials with high Tc, their preparation, although difficult, was also known to skilled persons, as can be seen for instance from the documents E12 (see page 1015, right-hand column, first full paragraph), E14 (see Tables 3 and 6) or E17 (see page 9382, last paragraph to page 9383, right-hand column, first paragraph), which concern such materials and their preparation.

Concerning the differences of temperature in the thermal treatments, 850°C in the patent in suit as compared to 865°C ± 10°C in document E17, it is to be noted that document E14 shows 2 examples in Table 3, at 1113 K and 1108 K, ie, 840°C and 835°C,
respectively. Therefore, the materials can be prepared using the indications about the temperature in the patent in suit.

It is also to be noted that the content of oxygen is reached automatically since the thermal treatments are in open air. Concerning the experiments made by the opponent, it is to be noted that it is a single experiment and, additionally, that sintering was carried out for 6 hours, this being much less than what is indicated in the relevant literature. Indeed, Example 7 of the priority document 26130/88 is in contradiction with the results for Example 1; however, it was realized at the time of filing the European patent application that the incorporation of Example 7 was an error. Accordingly, this example was not included in the European patent application.

On the other hand, Examples 1 and 9 of Table 2 of the patent in suit give information about prepared compositions falling within the scope of the main request and the auxiliary request 1, Example 1 concerning specifically a material according to auxiliary request 2.

Therefore, the invention is disclosed in a manner sufficiently clear and complete for it to be carried out by a skilled person.

Moreover, since the auxiliary request 2 concerns the same invention as the priority document 26130/88, the patent is entitled to the priority and the subject-matter of this request is new and inventive having regard to the state of the art.
Reasons for the Decision

1. **Admissibility of the appeal**

   The respondent (patent proprietor) had submitted that the appeal was not admissible because it did not specify the extent to which amendment or cancellation of the contested decision was requested, as required by Rule 64(b) EPC.

   The Board, in the communication accompanying the summons to the oral proceedings, pointed out that the notice of appeal met the requirements of Rule 64 EPC, and that the appeal was therefore admissible.

   In his last letter, filed on 3 December 2001, the respondent declared that he did not wish any more to contest the admissibility of the appeal. The issue of admissibility of the appeal therefore does not require any further discussion and the appeal is admissible.

2. **Admissibility of the requests of the respondent**

   The appellant objected that, since the requests of the respondent had not been filed within the time limit of one month before the oral proceedings mentioned in the communication of the Board, these requests were not filed in due time and were not admissible.

   However, these new requests address objections expressed in the Board's communication and they do not contain any new matter with respect to the former request. Therefore, the Board, in the exercise of its discretion under Article 114(2) EPC, admits them into the proceedings.
3. **Main request and the auxiliary request 1**

Claim 1 of the main request concerns a superconductive oxide material which substantially consists of a single phase $\text{ABiCuO}$, where $A$ is a mixture of:

(1) Ca and Sr and at least one of Mg and Ba, or

(2) Ca and Sr,

the $\text{ABiCuO}$ forming a crystal phase in which the atomic ratio of $A/\text{Bi/Cu}$ is substantially $3/2/2$ and the material having a superconductive transition temperature above $80 \text{ K}$.

In contrast to claim 1 of the main request, component $A$ of the superconductive material of claim 1 of the auxiliary request 1 contains both Mg and Ba in alternative (1). Such a composition is disclosed in Example 9 of Table 2 in the patent in suit.

As pointed out by the appellant in the patent in suit, in particular in Table 2, there is no example of a superconductive oxide material which substantially consists of a single phase $\text{ABiCuO}$, where $A$ is either

a mixture of Ca, Sr and Mg (thus without Ba) or

a mixture of Ca, Sr and Ba, (thus without Mg)

and having the further properties mentioned in the claim.

It was also contended by the appellant in the statement setting out the grounds of appeal that it was known in
the art that amounts of Mg and Ba as substituents of Sr and Ca have a negative effect on the high temperature properties and can thus lead to materials which show no supraconductivity. Moreover, according to the appellant, it has not been possible till to date to produce a high temperature ABiCuO single phase 322 superconductive material containing either Mg or Ba.

The appellant submitted in connection with the above Example 9, which is a Sr/Ca/Ba/Mg/Bi/Cu composition in the proportion 1.0/1.0/0.5/0.5/2/2, that even a minimal amount of Mg was known to have detrimental effect on the high temperature superconductive properties. It was therefore contested that it was possible to produce a substantially single phase high temperature superconductive material containing a relatively high amount of Mg as reported in Example 9. In the event that the Board considered it necessary, the appellant would provide a published document supporting his contention.

The question of feasibility of the ABiCu single phase composition containing Mg or Ba was raised by the appellant at the beginning of the appeal proceedings. However, the respondent, apart from stating that such a composition in Example 9 of the patent in suit was produced, did not make any further submissions refuting the allegations of lack of feasibility or reproducibility of the composition.

Also, at the oral proceedings, the respondent, when asked by the Board whether he was aware of any published document reporting a single phase ABiCuO high temperature superconductor containing Mg or Ba, stated that he was not aware of such a document.
In view of the above, the Board is persuaded by the appellant's arguments and concludes that the composition of Example 9 corresponds to an isolated unconfirmed result which has not been reproduced in the art following the ceramic method disclosed in the patent in suit.

Therefore, the Board concludes that the invention as defined in claim 1 of the main or auxiliary request 1 is not sufficiently disclosed (Article 100(b) and 83 EPC).

4. **Auxiliary request 2**

4.1 **Formal requirements**

There were no objections by the appellant under Article 84 EPC, Article 123(2) EPC or Article 123(3) EPC against claim 1.

It is to be noted that claim 2 is identical with claim 2 of the text of the European patent application maintained by the decision of the opposition division.

The Board is also satisfied that claim 1 as amended is clear (Article 84 EPC) and complies with Article 123(2) and 123(3) EPC.

4.2 ** Sufficiency of disclosure**

As to the objection under Article 100(b) EPC, the question which needs to be considered is whether taking into account the information provided in the patent in suit and the common general knowledge in the field of ceramic oxide superconductive materials, it would have
been possible for a person skilled in the art to obtain a superconductive oxide material as set out in claim 1.

The only information in the patent in suit regarding the manufacture of the superconductive oxide material in question is provided in the two paragraphs preceding Table 2. It follows from this disclosure that for Example 1 of Table 2, a mixture of oxides of Sr, Ca, Bi and Cu was formed so as to give atomic ratio $\text{A/Bi/Cu}$ of $3/2/2$, the oxides were thoroughly mixed, calcined at 800 to 850°C, crushed, formed and finally sintered at 850°C. In the paragraphs following Table 2, it is further disclosed that compositions 1 to 5, 9 and 10 had a transition temperature in the range of 80 to 85 K and that in case of Example 1 X-ray analysis confirmed the presence of a single crystalline phase with the $\text{A/Bi/Cu}$ ratio of $3/2/2$.

With regard to the issue of sufficiency of disclosure, the Board accepts the submissions of the respondent (see item IX above) that at the priority date of the patent in suit, the ceramic method was well known in the art for the manufacture of ceramic oxides including the known high temperature superconductive oxides. Consequently, once the skilled person was informed of the formation of a single phase of $\text{A/Bi/Cu}$ in the ratio $3/2/2$, as in the patent in suit, it was within his routine expertise to adjust various process parameters such as cooling rate, oxygen content, etc..., to reproduce the high temperature superconductive oxide as claimed. This view is supported by the disclosure in document E17 which was published on 1 June 1988, only four months after the priority date of the patent in suit. It follows from the disclosure on page 9382, last paragraph of document
E17 that a single phase compound is formed at the composition $\text{Bi}_4\text{Sr}_3\text{Ca}_3\text{Cu}_4\text{O}_y$ (4-3-3-4), ie having A/Bi/Cu ratio of 3/2/2, using a ceramic method similar to the one employed in the patent in suit. Although the lower limit (855°C) of the sintering temperature range (865°C ± 10°C) reported in the document for the formation of the single phase is 5°C higher than the sintering temperature of 850°C employed in the patent in suit, the narrow temperature range in the Board's view cannot be regarded as the necessary requirement. This, as pointed out by the respondent, follows from the disclosure in document E14, which was published after the publication of document E17, wherein it is reported that a single phase compound Bi-Sr-Ca-Cu-O having a A/Bi/Cu ratio of 3/2/2 is formed when the sintering was carried out at 1108 K (835°C) and 1113 K (840°C) (see Table 3, compositions in lines 6 and 7).

The appellant has also referred to the arguments and the results of comparative measurements in his letter dated 23 August 1997 during the opposition proceedings; in particular, for the third example, which corresponds to Example 1 of Table 2 of the patent in suit, with $\text{BiSrCaCuO}$ (BSSCO) with proportions 2 / 1.5 /1.5 / 2, the superconductor is shown as presenting no definite single phase structure and as having a $T_c$ of mainly 77 K, ie under the 80 K required by claim 1 in dispute.

However, as convincingly argued by the respondent, the duration of the heat treatments mentioned in the comparative measurement are in hours, whereas durations in days are generally known to the skilled person (see for instance the cited examples and examples of similar compositions in Table 3 of document E14). Therefore, the results of the comparative measurements are not...
conclusive and thus not clearly in contradiction to the findings of the patent in suit.

The appellant has also pointed out that although Example 1 in the priority document relevant for 3/2/2 materials, ie, No. 26130/88, is identical with Example 1 of Table 2 of the patent in suit, there is however in the same priority document another example, ie, Example 7, which is disclosed as Example 1 having regard to composition and preparation, but with no single phase of the obtained material; thus, there is a doubt, whether the invention can be carried out in a repetitive way.

The following is to be noted in this respect:

One of the inventors, Dr. Kugiyima, was asked by the Board at the oral proceedings for clarification of this point and stated that Example 7 of the Table of this priority document was wrong and its inclusion was an obvious mistake. This example was therefore not contained in the European application as filed.

The Board finds that this is a plausible explanation so that the above inconsistency cannot be regarded as indicative of nonreproducibility of Example 1 of Table 2.

In any case according to document E14 (see Table 3) materials of the same type as Example 1 have been prepared, so that the appellant's argument is not considered as convincing.

The appellant has also objected that the patent in suit does not disclose the exact formula of the obtained
materials, in particular the concentration of oxygen, or the X-ray parameters of said materials. However, as explained by the respondent, the disclosure of the lattice constants in the patent is correct, and the lattice constants \( a, b \) and \( c \) are close to the values given in documents E12 and E14. Also, the patent specification (cf. page 5, lines 26 to 30) explains how the values \( a = 1.53 \) and \( b = c = 2.29 \) are obtained. As regards the oxygen content, it was stated by the respondent that sintering was carried out in the air, as is common in the ceramic method.

Thus, notwithstanding the fact that the preparation of the claimed materials was not easy, the skilled person, taking into account the information in the patent in suit and his general knowledge, was in a position to prepare materials of this type without undue burden (see the decision T 409/91, OJ EPO 1994, 653, in particular point 3.5 of the reasons; see also the decision T 435/91, OJ EPO 1995, 188, points 2.2 to 2.2.3 and 4.1 to 4.2).

Consequently, the Board concludes that the invention according to claim 1 of the auxiliary request 2 is sufficiently disclosed in the sense of Article 100(b) and 83 EPC.

4.3 Novelty

4.3.1 For determining the extent of the state of the art in the sense of Article 54(2) EPC, it is necessary to consider whether the patent in suit is entitled to the claimed priority.

The priority document 26130/88 discloses in the first
example of the Table a material with identical composition and identical results and properties as Example 1 of Table 2 of the patent in suit. Thus, the auxiliary request 2 represents the same invention as in the priority document. Moreover, since the information in the priority document 26130/88 is in substance very close to the auxiliary request 2 and since as set forth above this request has been found to disclose the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art, the disclosure in the priority document is considered as being enabling.

Therefore, the Board concludes that the auxiliary request 2 is entitled to the priority of, in particular, the priority document 26130/88 (Article 87(1) EPC).

The documents E13 to E17 published after the priority date of the patent in suit are therefore not comprised in the state of the art according to Article 54(2) EPC.

4.3.2 In the opinion of the Board, none of the prior art documents in the sense of Article 54(2) EPC discloses or suggests a material according to claim 1 of the auxiliary request 2, so that the subject-matter of claim 1 is new. This has not been disputed by the appellant.

It is to be noted that the appellant had submitted that the subject-matter of claim 2 was not new in the sense of Article 54(3) EPC having regard to the content of document E5, which is a European patent application claiming a priority anterior to the priority of the patent in suit, in particular having regard to
Example 1 of Table 1 thereof, which is identical to Example 3 of Table 3 of the patent in suit. However, since according to the auxiliary request 2 none of the examples of Table 3 of the patent in suit falls within the scope of the invention, and since moreover document E5 in particular does not disclose a multi-layered superconductive oxide material in accordance with claim 2, this claim is new.

4.4 Inventive step

As mentioned above, document E5 belongs to the state of the art according to Article 54(3) EPC. This document is therefore not relevant for the consideration of inventive step.

Document E3 discloses a high temperature superconductive oxide having Bi-Sr-Ca-Cu$_2$O$_x$ composition which comprises at least two high temperature superconducting phases. There is no suggestion in any of the remaining cited prior art documents regarding a substantially single phase ABiCuO composition as set out in claim 1 of the patent in suit, so that it was not obvious to the skilled person to arrive at the claimed invention in view of the cited prior art.

4.5 For the foregoing reasons, the Board concludes that claims 1 and 2 are new in the sense of Article 54 EPC and that they involve an inventive step in the sense of Article 56 EPC.

Order
For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance to maintain the patent on the following documents:

   Claims 1 and 2 according to the auxiliary request 2, filed on 3 December 2001;

   Description:

   Page 1 according to the auxiliary request 2, filed on 3 December 2001;

   Pages 3, 4, 5 and 8 to 10 filed during the oral proceedings before the opposition division on 28 October 1997; and

   Page 2, 6 and 7 filed during the oral proceedings on 12 December 2001.

The Registrar:                  The Chairman:

D. Spigarelli                  R. K. Shukla