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D E C I S I O N
of 13 August 2002

Case Number: T 0324/99 - 3.4.3

Application Number: 92109097.3

Publication Number: 0516148

IPC: H01L 39/12

Language of the proceedings: EN

Title of invention:

Oxide superconductive material and method of preparing the same

Applicant:

SUMITOMO ELECTRIC INDUSTRIES, LIMITED, et al

Opponent:

-

Headword:

-

Relevant legal provisions:

EPC Art. 84 and 56

Keyword:

"Clarity: yes (after amendments)"

"Inventive stop: yes (after amendments)"

Decisions cited:

-

Catchword:

-



Case Number: T 0324/99 - 3.4.3

D E C I S I O N
of the Technical Board of Appeal 3.4.3
of 13 August 2002

Appellant:

SUMITOMO ELECTRIC INDUSTRIES, LIMITED
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Chuo-Ku
Osaka 541 (JP)

Representative:

Winter, Brandl, FURNISS, Hübner, Röss,
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Partnerschaft
Patent- und Rechtsanwaltskanzlei
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Decision under appeal:

Decision of the Examining Division of the
European Patent Office posted 23 July 1997
refusing European patent application
No. 92 109 097.3 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: R. K. Shukla
Members: M. Chomentowski
M. J. Vogel

Summary of Facts and Submissions

- I. The European patent application 92 109 097.3 (Publication No. 0 516 148) was refused in a decision of the examining division dispatched on 23 July 1997 on the ground that independent claims of the applicant's main and auxiliary request lacked clarity.

Claim 1 of the main request and independent claim 2 of the auxiliary request forming the basis of the decision were directed to an oxide superconductive material and to such a material obtainable by specific method steps, respectively, whereas claim 1 of the auxiliary request was identical with independent method claim 2 of the main request.

Claim 2 of the main request reads as follows:

"2. A method of preparing an oxide superconductive material being expressed in the following formula:



where x, y and z are in relations satisfying $1.5 \leq x \leq 2.0$, $2.0 \leq y \leq 2.5$, $x + y = 4.0$ and $9.0 \leq z \leq 11.0$, said method comprising the steps of:

mixing powder metal oxides in blending ratios for satisfying said composition formula;

sintering the as-formed mixed powder in oxygen gas or in air to obtain a sintered body; and

annealing said sintered body in a decompressed sealed tube at 700 to 800°C for at least 10 hours."

II. Most of the objections concerning lack of clarity in the decision are in respect of independent claims relating to oxide superconductive materials and are also applicable to the method claims. Since as set forth hereunder only method claims form the basis of the present decision, the relevant reasoning in the appealed decision is essentially as follows:

There is an inconsistency between the only embodiment of the invention in the description (cf. page 5, line 25) with oxygen content $z = 11.85$ in the composition formula and the claimed composition range of oxygen $9.0 \leq z \leq 11.0$, since the value $z = 11.85$ falls outside the claimed range. The applicant's argument in this respect about the composition of the Ba oxide starting material appearing in Example 1 being erroneous is considered as not convincing in view of the teaching of document

D1 = Physical Review Letters, vol. 60, no. 24, 13 June 1988, pages 2539 to 2542, S. S. P. Parkin et al.: "Bulk Superconductivity at 125 K in $Tl_2 Ca_2 Ba_2 Cu_3 O_x$ "

that different compositions of Ba oxide starting materials may be used in a preparation method.

Moreover, the wording "blending ratio for satisfying said composition formula" merely means ratios suitable for the desired composition of the final products and is unclear. In the description (cf. page 8, lines 4 to 9), the terms "similar manner" may have a meaning different from that of terms such as "same manner"; thus, the application seems to include only one embodiment (Example 1), and this embodiment is different from the subject of the claim.

Further objections concern lack of essential features, e.g. the lattice constants with respect to the different compositions, and the wording in form of a result to be achieved, e.g. the transition temperature / zero resistance temperature exceeding 125 K, in some of the claims.

Insofar as the method claim 2 of the main request (method claim 1 of the auxiliary request) can be understood, it does not involve an inventive step having regard to document

D2 = Physica C, vol. 171, no. 5 & 6, 15 November 1990, pages 543 to 546, Seiji Adachi et al.: "Synthesis of Tl - Ba - Ca - Cu - O ceramics showing Meissner effect over 125 K",

since the only difference from the prior art was in the use of oxygen containing atmosphere (during sintering) which was within the routine expertise of the skilled person.

III. The applicant lodged an appeal against this decision on 1 October 1997 paying the appeal fee on the same day. A statement setting out the grounds of the appeal was filed on 1 December 1997.

IV. In response to communications from the Board pursuant to Article 11(2) of the rules of procedures of the Boards of Appeal, the appellant filed new claims 1 and 2 with a letter dated 8 August 2002 and requested that the decision under appeal be set aside and a European patent be granted on the basis of the following patent application documents:

Description:

Pages 1, 5 and 7 to 9 filed with the letter dated 12 October 1992;

Pages 2 to 4, 6, 10 and 11 filed with the letter dated 8 August 2002;

Claims:

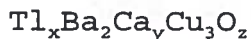
Claims 1 and 2 filed with the letter dated 8 August 2002;

Drawings:

Sheets 1/6 to 6/6 as filed with the letter dated 12 October 1992.

Claim 1 of the appellant's request reads as follows:

"1. A method of preparing an oxide superconductive material being expressed in the following composition formula:



where x, y and z are in relations satisfying $1.5 \leq x \leq 2.0$, $2.0 \leq y \leq 2.5$, $x + y = 4.0$ and $9.0 \leq z \leq 11.0$, and comprising a tetragonal system superconducting phase having lattice constants of $a = 0.385$ to 0.386 nm and $c = 3.575$ to 3.580 nm,

said oxide superconductive material exhibiting superconductivity under a temperature of at least 125 K

said method comprising the steps of:

mixing powder raw materials in blending ratios for satisfying said composition formula;

sintering the as-formed mixed powder in flowing oxygen gas to obtain a sintered body; and

annealing said sintered body under vacuum at **about 750°C** for **more than** 10 hours."

(Differences by addition or substitution with respect to the text of the method claim forming the basis of the appealed decision are emphasized by the Board.)

Claim 2 is a dependent method claim.

V. The appellant submitted the following arguments in support of his requests:

Claim 1 and dependent claim 2 are method claims. Claim 1 contains all the features essential to the performance of the invention and there is no inconsistency between its text stating a composition formula $Tl_xBa_2Ca_yCu_3O_z$ where $9.0 \leq z \leq 11.$, and Example 1 in the description with a blending composition (and not a composition formula), of $z = 11.85$. Therefore, claim 1 is clear.

Documents D1 and D2 are the only prior art documents in the sense of Art. 54(2) EPC. The exact value of the amount of oxygen in the formula of the superconductive materials in these documents is not disclosed.

Moreover, the method steps in these documents are different from each other and also different from those of the claimed method. Therefore, the subject-matter of claim 1 is not obvious having regard thereto and involves an inventive step.

Reasons for the Decision

1. The appeal is admissible.
2. *Admissibility of the amendments*

Claim 1 is based on independent method claim 3 of the application as filed. It contains also features of the prepared materials, especially its lattice constants, based on the other independent claims and on the whole content of the application as filed. Moreover, present claim 1 contains features of the method based on Example 1 and the other Examples prepared in a "similar manner". Thus, in the application as filed, the specific feature that sintering of the as-formed mixed powder takes place in **flowing** oxygen gas is based on the subject-matter of the claim 3 ("in an oxygen jet in the atmosphere"), and on the disclosure on page 2, line 25 to page 3, line 3, that the sintering is "in flowing oxygen", whereby the wording "in following oxygen" is evidently a typographic error. The further specific feature that annealing the sintered body takes place under **vacuum** is also based on this claim 3 ("in a closed atmosphere") and the description of Example 1, that annealing is done in a silica tube which was decompressed with a vacuum pump until its internal pressure was 10^{-4} Torr and was sealed. With respect to the present feature "about 750°C", it is to be noted that, in the application as filed, whereas claim 3 discloses an annealing temperature of 700 to 800°C, Example 1 is disclosed with an annealing temperature of 750°C.

Claim 2, concerning a method for preparing an oxide superconductive material wherein $x = 1.7$ and $y = 2.3$, is based on the composition of Example 1 in the description.

The amendments in the description are for adaptation to the new claim 1.

Therefore, the Board is satisfied that the application complies with the requirement of Article 123(2) EPC that a European patent application may not be amended in such a way that it contains subject-matter which extends beyond the content of the application as filed.

3. *Clarity of the claims*

As convincingly argued by the appellant, there is no inconsistency between the claim, stating that the amount z of oxygen in the composition formula is $9.0 \leq z \leq 11.0$, and the description of Example 1 wherein the blending ratios are "for attaining a blending composition of $Tl_{1.7} Ba_2 Ca_{2.3} Cu_3 O_{11.85}$ ", insofar as these ratios can evidently be different without corresponding to different products; such a wording is also currently used in the relevant technical field as can be seen from the cited prior art documents, so that there is no unclarity there.

Moreover, as a result of the amendments provided by the appellant, claim 1 includes the features, i.e., the ratios in the composition formula, the lattice constants, the particular environments for sintering and for annealing, which, in agreement with the statements in the description, are necessary to arrive at the aimed superconducting temperature of 125 K.

Thus, the objections in the decision under appeal are met by the amendments to claim 1.

It is also to be noted that it is directly and unambiguously derivable from the content of Table 1 that only the samples 5 and 11 to 14, for which both the Zero Resistance Temperature and the Diamagnetic Signal Starting Temperature are at least 125 K, correspond to products of the method of present claim 1.

Therefore, in the Board's judgement, claim 1 is clear in the sense of Article 84 EPC.

4. *Patentability of the claims*

The subject-matter of claim 1 is not comprised in any of the documents D1 and D2 and is thus new in the sense of Article 54 EPC.

Although $Tl_2Ba_2Ca_2Cu_3O_z$ materials are mentioned in document D2, there is no clear teaching therein about the amount z of oxygen as being in the range 9 to 11. Indeed, it is only "speculated" that the material is $Tl_2Ba_2Ca_2Cu_3O_{10}$, i.e., $z = 10$, with excess oxygen located at interstitial sites in Tl_2O_2 layers (cf. page 546, first column, first full paragraph). Moreover, in this known method (cf. page 543, the paragraph bridging the two columns), there is no sintering in flowing oxygen and, although the rectangular bars preformed from the mixed powders are "fired" in an evacuated quartz tube, there is no indication that this heat treatment is an annealing step.

In document D1 (page 2539, the paragraph bridging the two columns), although the sintering takes place in oxygen, this is however done in a sealed quartz tube, and not in flowing oxygen. Moreover, there is no clear teaching about annealing, and especially not in vacuum.

Therefore, a combination of the teachings of documents D1 and D2 does not lead in an obvious way to the method of present claim 1, so that the subject-matter of claim 1 involves an inventive step in the sense of Article 56 EPC.

Consequently, claim 1 is patentable in the sense of Article 52(1) EPC.

Claim 2 concerns a particular embodiment of claim 1 and is also patentable for the same reasons.

Therefore, a patent may be granted on this basis (Art. 97(2) EPC).

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the department of first instance with the order to grant a patent with the following patent application documents:

Description:

Pages 1, 5 and 7 to 9 filed with the letter dated 12 October 1992;

Pages 2 to 4, 6, 10 and 11 filed with the letter dated 8 August 2002;

Claims:

Claims 1 and 2 filed with the letter dated 8 August 2002;

Drawings:

Sheets 1/6 to 6/6 as filed with the letter dated 12 October 1992.

The Registrar:

The Chairman:

R. Schumacher

R. K. Shukla