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D E C I S I O N
of 3 December 2002

Case Number: T 0589/99 - 3.4.3

Application Number: 88103367.4

Publication Number: 0282839

IPC: H01L 39/24

Language of the proceedings: EN

Title of invention:
Films of high Tc oxide superconductors and method for making the same

Patentee:
International Business Machines Corporation

Opponent:
Siemens AG

Headword:
-

Relevant legal provisions:
EPC Art. 56, 83, 100(a) and 100(b)

Keyword:
"Main and first auxiliary request - disclosure not sufficient to carry out the invention over the claimed range of composition"
"Second auxiliary request - sufficiency (yes)"
"Inventive step - (yes)"

Decisions cited:
-

Catchword:

-

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Case Number: T 0589/99 - 3.4.3

D E C I S I O N
of the Technical Board of Appeal 3.4.3
of 3 December 2002

Appellant: (Opponent)
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Representative:
-

Respondent: (Proprietor of the patent)
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Decision under appeal:
Interlocutory decision of the Opposition Division of the European Patent Office posted 4 May 1999 concerning maintenance of European patent No. 0 282 839 in amended form.

Composition of the Board:

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

Summary of Facts and Submissions

I. The European patent No. 0 282 839 was granted on the basis of the European patent application No. 88 103 367.4, claiming the priority from a US application No. 27584 of 18 March 1987.

II. The appellant filed an opposition against the European patent on the grounds mentioned in Article 100(a) and 100(b) EPC that it did not disclose a process for obtaining a continuous smooth film according to the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art, and that it did not involve an inventive step having regard *inter alia* to the prior art documents

D1: Nature, vol. 235, 19 February 1987, pages 664 and 665, M. Strongin et al., "Superconductivity at high temperatures in doped oxides";

D2: IEEE Transactions on Magnetics, vol. MAG-11, No. 2, March 1975, pages 201 to 207, R. H. Hammond, "Electron beam evaporation synthesis of A15 superconducting compounds: Accomplishments and Prospects";

D3: Journal of Vacuum Science and Technology, vol. 15, No. 2, March/April 1978, pages 382 to 385, R. H. Hammond, "Synthesis and physical properties of superconducting compound films formed by the electron-beam codeposition of the elements"; and

D8: Phys. Rev. Letters, vol. 58, No. 9, 2 March 1987, pages 908 to 910, M. K. Wu et al.,

"Superconductivity at 93 K in a New Mixed-Phase Y-Ba-Cu-O Compound System at Ambient Pressure".

III. The European patent was maintained in amended form by an interlocutory decision dated 4 May 1999 of the opposition division.

Claim 1 was the only independent claim of the set of 8 claims of the patent as maintained in amended form, and it had the following text:

"1. A process of making a crystalline film exhibiting superconductivity at temperature is (sic) excess of 40 K, the film being a single transition metal oxide phase having the general formula

$A_{1\pm x}\ M_{2\pm x}\ Cu_3\ O_y$

where A is Y or a combination of Y, La, Lu, Sc or Yb;
M is Ba or a combination of Ba, Sr or Ca;
x is between 0 and 0.5 and y is sufficient to satisfy the valence demands of the material,

said process including the steps of:

employing an evaporation process to transport metal atoms from separate metal sources to a substrate;

providing an oxygen ambient at said substrate while said metal atoms are arriving there at, said metal atoms and said oxygen reacting to form a transition metal oxide film on said substrate;

annealing the as-deposited oxide film in an oxygen environment;

and slowly cooling said annealed film to room temperature."

IV. The reasoning of the opposition division can be summarized as follows:

Taking documents

D9: Phys. Rev. B, vol. 35, No. 16, 1 June 1987, pages 8821 to 8823, R. B. Laibovitz et al., "Thin superconducting oxide films"; and

D17: Phys. Rev. Letters, vol. 58, No. 16, 20 April 1987, pages 1676 to 1679, R. J. Cava et al., "Bulk superconductivity at 91 K in single-phase oxygen-deficient perovskite $Ba_2YCu_3O_{9-ä}$",

which were published after the priority date of the patent in suit, as providing evidence of the common general knowledge in the art at the priority date of the patent in suit, the skilled person had all the parameters needed to carry out the method to produce a continuous and smooth crystalline film as specified in claim 1 (Art. 100(b) EPC).

None of the superconductor materials disclosed in document D1 is a single transition metal oxide phase having the particular general formula as claimed. Although the metallic components Y, Ba and Cu which form the typical 123 (i.e. $Y_1Ba_2Cu_3$ oxide) material were known, the investigated material of a similar compounds showed multiple phases and the skilled person neither knew any exact composition nor was he unambiguously directed to try to manufacture a single phase material.

Moreover, the process of manufacturing did not follow from the evaporation techniques known in particular from documents D2 and D3 in a straightforward manner, whereby in particular the requirements for a correct oxygen content of the ceramic and thus the necessary oxygen ambient during deposition and annealing were not disclosed. Furthermore, the effect of these process steps together with the slow cooling, which is described in the patent in sufficient detail, for forming a single film as in the claim was not predictable.

Therefore, the subject-matter of claim 1 involved an inventive step (Art. 56 EPC).

V. The opponent lodged an appeal against this decision on 28 May 1999 paying the appeal fee on the same day. A statement setting out the grounds of the appeal was filed on 25 August 1999.

VI. In response to a communication annexed to summons to oral proceedings, the respondent filed on 18 September 2002 three sets of claims according to a main request, first auxiliary request and second auxiliary request, respectively.

In relation to claim 1 as maintained in the decision under appeal, in claim 1 of the **main request**, the indices for the elements A and M in the general formula of the single transition metal oxide phase of the crystalline film are expressed differently, i.e., in

$A_{1+x}\ M_{2+z}\ Cu_3\ O_y$

A is Y or a combination of Y, La, Lu, Sc or Yb;

M is Ba or a combination of Ba, Sr or Ca;
x and z are the same or different and are between 0 and 0.5 and 0 and -0.5 and y is sufficient to satisfy the valence demands of the material.

Moreover, in the last step of cooling of the annealed film, the statement "slowly cooling said annealed film to room temperature" in claim 1 as maintained by the opposition division is replaced by "over several hours slowly cooling said annealed film to room temperature".

In claim 1 of the **first auxiliary request**, A is restricted to Y (yttrium) and B is restricted to Ba (barium), these being the only differences with respect to the main request.

Claim 1 of the **second auxiliary request** reads as follows:

"1. A process of making a crystalline film exhibiting superconductivity at temperature in excess of 40 K, the film being a single transition metal oxide phase having the general formula

$Y\ Ba_2\ Cu_3\ O_y$

where y is sufficient to satisfy the valence demands of the material,

said process including the steps of:

employing an evaporation process to transport metal atoms from separate metal sources to a substrate;

providing an oxygen ambient at said substrate while

said metal atoms are arriving there at, said metal atoms and said oxygen reacting to form a transition metal oxide film on said substrate;

annealing the as-deposited oxide film in an oxygen environment;

and over several hours slowly cooling said annealed film to room temperature."

Claims 2 to 7 of the second auxiliary request are dependent claims.

VII. Oral proceedings took place on 3 December 2002.

VIII. The respondent requested that the decision under appeal be set aside and that the patent be maintained on the basis of the main, the first or second auxiliary request, with the description to be adapted. He submitted the following arguments in respect of his requests:

Crystalline, single phase thin films of the superconductive materials mentioned in claim 1 of the main request and of the first auxiliary request, which are of the same compounds as those described in their bulk form in document

D10: EP-A-0 281 753 = European application No. 88 101 321.3 cited as copending application in the patent in suit and published after the priority date of the patent in suit,

can be made by the claimed process, and this is in particular the case for the material $Y\ Ba_2\ Cu_3\ O_y$.

On the basis of the information provided in the patent in suit, a person skilled in the art was in a position to produce a single phase superconducting thin film having a composition extending over the entire range of elements and their proportions as claimed without undue burden.

In particular, for the thin film material of $Y\ Ba_2\ Cu_3\ O_y$ it is generally known that it forms a single crystalline superconducting phase for which the content in oxygen can be adjusted easily by the skilled person in a routine manner. Therefore, the invention of the second auxiliary request is sufficiently disclosed (Art. 100(b) EPC).

At the priority date of the patent in suit, a compound with the specific composition $Y\ Ba_2\ Cu_3\ O_y$ was not known, and there was thus no incentive for looking for a process of fabricating thin films of such materials. Documents D2 and D3 were primarily directed at the co-deposition of alloys, and were not concerned with forming oxides of such alloys. Therefore, it is only with hindsight that the subject-matter of the claim can be considered obvious, so that it involves an inventive step (Art. 56 EPC).

IX. The appellant requested that the decision under appeal be set aside and that the patent be revoked. His arguments can be summarized as follows:

It follows from the following standard book

D21: the book " Materialwissenschaften", Spektrum Akademischer Verlag Heidelberg et al. 1996, page 217,

that single phase crystalline material must have a definite composition, and this is not the case for the broad range of products defined in claim 1 of the main and first auxiliary request. Thus document D9, although published after the priority date of the patent in suit, provides evidence that the results of measurements on a film with the particular composition $Y_{0.87}$ $Ba_{1.53}$ Cu_3 O_y show the existence of a second phase It is also generally known, e.g. from documents D9 and D17, that superconductors of this category must have a perovskite crystalline structure, and that is not possible to vary the composition as indicated in these requests. Therefore the inventions of the main and the first auxiliary request are not sufficiently disclosed.

The invention of the second auxiliary request is also insufficiently disclosed in that, since the amount of oxygen in the composition is not defined and since the terms "several hours slowly cooling" are not clear, it is only with undue burden that the skilled person can carry out the invention.

Once it was known from document D8 that the Y-Ba-Cu-O compound system presented superconductivity at temperatures as high as 93K, it was obvious to the skilled person to look for the available relevant fabrication techniques, such as the evaporation techniques of documents D2 and D3, which also teach the production of oxides such as Al_2O_3. This was particularly the case since it was known from e.g. document D1 that other materials having similar compositions were superconductive. Therefore, the process of the second auxiliary request lacks an inventive step.

Reasons for the Decision

1. The appeal is admissible.

2. *Sufficiency (Art. 83 EPC) - Main and first auxiliary request*

2.1 The invention as defined in claim 1 of the main request is a process of making a crystalline film exhibiting superconductivity at temperature in excess of 40 K, the film being a single transition metal oxide phase having the general formula

$A_{1+x} M_{2+z} Cu_3 O_y$

where A is Y or a combination of Y, La, Lu, Sc or Yb;

M is Ba or a combination of Ba, Sr or Ca;

x and z are the same or different and are between 0 and 0.5 and 0 and -0.5 and y is sufficient to satisfy the valence demands of the material.

The claimed process includes the steps of:

employing an evaporation process to transport metal atoms from separate metal sources to a substrate;

providing an oxygen ambient at said substrate while said metal atoms are arriving there at, said metal atoms and said oxygen reacting to form a transition metal oxide film on said substrate;

annealing the as-deposited oxide film in an oxygen environment;

and slowly cooling said annealed film to room temperature.

Contrary to the invention as defined in claim 1 of the main request, wherein A is Y or a combination of Y, La, Lu, Sc or Yb and M is Ba or a combination of Ba, Sr or Ca, the process as defined in claim 1 of the first auxiliary request is restricted to the making of crystalline film with the same mentioned properties but with compositions restricted to A = Y and M = Ba.

2.1.1 In the statement setting out the grounds of appeal, i.e., in the letter dated 24 August 1999, the appellant contended that the patent in suit does not disclose sufficiently the invention for making thin films with the large range of variation of the indices for the elements such as Y and Ba complying also with the claimed requirement of single phase structure. There can already be seen a contradiction in the definition of the invention since it is generally known in the art, for instance from document D21, that a single phase of a crystalline material is determined by a specific crystalline structure and a specific stoichiometry, and this is evidently not the case for a compound with such a wide range of compositions.

Document D21 was published in 1996, i.e., 9 years after the priority date of the patent in suit and reflects the general knowledge of the skilled person concerning the science and engineering of materials, which in this respect has not changed, since the priority date.

Moreover, in the statement of grounds, and also during the oral proceedings, it was contended that the embodiment in the patent as maintained (see column 7,

line 49 to column 8, line 3), concerning a Y-Ba-Cu-O film with the composition $Y_{0.87}$ $Ba_{1.53}$ Cu_3 O_y, thus falling within the range defined in claim 1 and derivable as being a product of the claimed process, is not a single phase material.

In this respect, reference was first made to document D17 (see the abstract; page 1676, left-hand column, second paragraph, lines 1 to 10), which concerns bulk superconductivity at 91K in single phase oxygen-deficient perovskite Y Ba_2 Cu_3 O_y, i.e., a material according to the general formula and which states that in the Y-Ba-Cu-O system the superconducting crystalline phase is the 1-2-3 phase (Y_1 Ba_2 Cu_3 O_y). This document, which was published more than one month after the priority date of the patent in suit, provides thus evidence of a single phase superconducting material only for the 1-2-3 phase.

Reference was also made to document D9, which reports on thin superconducting oxide films made by an evaporation method, and in particular on Y-Ba-Cu-O films with different compositions. According to document D9 (see page 8822, the paragraph bridging both columns and Figure 1(b); see also 8823, right-hand column, second paragraph), for a composition of the film with the same composition, i.e., $Y_{0.87}$ $Ba_{1.53}$ Cu_3 O_y, it is stated that the results indicate the existence of a second phase. The further materials mentioned in the document (see Fig. 1(a)), e.g. $Y_{0.75}$ $Ba_{1.35}$ Cu_3 O_y , i.e. z = - 0.65, are not comprised in the range defined in claim 1. Incidentally, it is to be noted that this further composition has been disclaimed in the patent as maintained (see column 7, lines 47 to 55). In document D9 , it is concluded with respect to the grown

thin films that, as the film fabrication develops and uniform films of the correct composition and single phase are developed, the supraconductive properties and applications of such films should be increased.

However, although the document does not provide all the details of the preparation process, e.g. the slow cooling, it is a document of the respondent which was published more than two months after the priority date and which was received by the publisher exactly on the priority date.

2.1.2 Moreover, concerning the crystallographic structure of the single phase crystalline material produced by the disputed process, it is also known, for instance from document D17 (see the abstract; page 1677, the paragraph bridging both columns; page 1679, first paragraph), that the crystalline single phase materials of this type of materials exhibiting superconductivity at 91K and thus at temperatures in excess of 40 K, the superconductive crystalline structure is a perovskite type structure. In this late document, it is assumed that the single phase Cu perovskite structure has a specific arrangement of the planes of YO and BaO.

According to document D10 (see column 1, line 35 to column 2, line 17), which concerns the same compositions as those of the patent in suit, it is stated that said compositions are single phase bulk electrical superconductors above 77K which have a perovskite-like structure. This document is the copending application mentioned in the patent in suit (see column 1, lines 25 to 35), by the same applicant, has a priority date of 11 March 1987, that is, a few days earlier than the patent in suit, and was published

on 14 September 1988.

In this respect, the appellant's argument that the perovskite structure did not allow wide variations of the composition as those mentioned in the claim is convincing since, as shown at least in document D9, the $Y_{0.87}$ $Ba_{1.53}$ Cu_3 O_y composition contains a second phase in the film.

2.1.3 The question, whether with the information in the patent in suit if was feasible to make thin films with the claimed method within the whole range of the compositions of the materials defined in the claim, was raised already before the oral proceedings. During the oral proceedings, the question has also been raised, more particularly with respect to the indices x and z, in particular in the Y-Ba-Cu-O system. The respondent stated that thin films of compositions other than Y_1 Ba_2 Cu_3 O_y could be made, but did not provide any evidence showing that a superconducting phase other than the 123 phase existed.

Thus, it is not established, 15 years after the priority date of the patent in suit, whether the disclosure in the patent in suit allows the invention to be performed in the whole range claimed (cf. the decisions **T 409/91**, OJ 1994, 653, item 3.5 of the reasons and **T** 435/91, OJ 1995, 188, item 2.2.3 of the reasons). On the contrary, the available evidence, in form of the documents cited here above, indicates that for at least one composition in the claimed range a single phase superconducting sample is not produced.

2.1.4 Therefore, in the Board's judgement, the patent in suit does not disclose the invention as claimed in the main

request in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art (Art. 100(b) and 83 EPC).

2.2 Since the above reasoning concerning the main request has been made by taking into consideration only compositions restricted to A = Y and M = Ba in the general formula of the single transition metal oxide phase crystalline film exhibiting superconductivity at temperature in excess of 40 K made by the process of the invention, it also applies to the first auxiliary request.

Therefore, also the first auxiliary request is not allowable (Art. 100(b) and 83 EPC).

3. *Second auxiliary request*

3.1 The Board is satisfied that the amendments contained in the claims are admissible, and no objections in this respect were raised by the appellant pursuant to Art. 123(3) and 123(2) EPC. In this respect, it is to be noted that objections of the appellant concerning the amendments proposed by the respondent as a non-appealing party, which would have put the appellant in a worse situation than if he had not appealed ("reformatio in peius") were not directed against the second auxiliary request.

3.2 Clarity and sufficiency

The appellant has objected that claim 1 in dispute does not specify the amount of oxygen to be incorporated in the $Y\ Ba_2\ Cu_3\ O_y$ thin single phase crystalline film exhibiting superconductivity at temperature in excess

of 40 K, that moreover it does not indicate the duration of the slow cooling of the annealed film; therefore, the claim lacks clarity and, since the description does not provide supplementary information in this respect, the patent does not sufficiently disclose the invention.

As regards the amount of oxygen to be incorporated in the composition, it is indeed correct that the claim does not specify the exact amount. The Board however does not find that this introduces any ambiguity in the definition of the subject-matter for which protection is sought.

Also, the expression "over **several** hours slowly cooling...to room temperature" is also clear in the overall context of the claimed subject-matter, in that the cooling has to be slow lasting over two or more hours with a view to providing a single phase superconducting film.

Therefore, in the Board's judgement, the claims are clear in the sense of Article 84 EPC.

Moreover, concerning the objection of insufficiency of disclosure, it is to be noted that it has not been disputed that the material Y_1 Ba_2 Cu_3 O_y can be prepared, and there is no evidence that there would have been an undue burden for the skilled person to prepare them in the thin film form defined in claim 1.

Therefore, in the Board's judgement, the patent discloses the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art (Art. 100(b) and 83 EPC).

3.3 Novelty and inventive step

The novelty of claim 1 has not been disputed (Art. 54 EPC).

The closest prior art in the sense of Art. 54(2) EPC is document D8 (see the abstract) concerning superconductivity between 80 and 93K in a new mixed-phase Y-Ba-Cu-O compound system. However, in the document, neither the specific composition Y Ba_2 Cu_3 O_y nor the thin single phase crystalline film structure exhibiting superconductivity at temperature in excess of 40K is suggested.

The appellant has argued that, starting from document D8, the person skilled in the art knowing that for many applications it is necessary to prepare such superconductive compounds in form of thin films, would refer to already well established deposition techniques using evaporation from separate metal sources, e.g. documents D2 and D3, thereby arriving at the compound having the required properties, i.e., the compound with the composition mentioned in the claim. This is in particular the case since in document D2 (see the paragraph bridging pages 203 and 204) aluminum oxide is deposited.

However, as convincingly argued by the respondent, at the priority date of the patent in suit, a compound with the specific composition Y Ba_2 Cu_3 O_y was not known, so that there was no incentive for looking for a process of fabricating thin films of such materials. Moreover, documents D2 and D3 were primarily directed at the co-deposition of alloys, and were not concerned with forming oxides of such alloys.

The further prior art documents either concern superconductors with other different compositions, materials in forms other than thin-films or made by different processes.

Therefore, in the Board's judgement, to the person skilled in the art, having regard to the state of the art, the subject-matter of claim 1 was not obvious, so that it involves an inventive step in the sense of Article 56 EPC.

3.4 Consequently, claim 1 is patentable in the sense of Article 52(1) EPC and the patent can be maintained on this basis, with the description to be adapted (Art. 102(3) 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 with the order to maintain the patent on the basis of claims 1 to 7 of appellant's auxiliary request 2 filed with letter dated 18 September 2002, with the description to be adapted.

The Registrar The Chairman

P. Martorana R. K. Shukla