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D E C I S I O N
of 28 September 2001

Case Number: T 0408/97 - 3.3.5

Application Number: 91111685.3

Publication Number: 0467238

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Language of the proceedings: EN

Title of invention:

Method for preparing bismuth superconductor

Patentee:

SUMITOMO ELECTRIC INDUSTRIES, LIMITED

Opponent:

Siemens AG

Headword:

Superconductor preparation/SUMITOMO

Relevant legal provisions:

EPC Art. 54(3), 56

Keyword:

"Novelty - yes (after amendment)"

"Inventive step - yes (after amendment)"

Decisions cited:

-

Catchword:

-



Case Number: T 0408/97 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 28 September 2001

Appellant:
(Opponent)

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Respondent:
(Proprietor of the patent)

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Representative:

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Decision under appeal:

Interlocutory decision of the Opposition Division
of the European Patent Office posted 3 February
1997 concerning maintenance of European patent
No. 0 467 238 in amended form.

Composition of the Board:

Chairman: R. K. Spangenberg
Members: B. P. Czech
J. H. Van Moer

Summary of Facts and Submissions

- I. The appeal is from a decision of the opposition division maintaining the patent in amended form.
- II. In the contested decision, the opposition division considered ten documents, including the following:

D2: Cryogenics, Vol. 30 (1990), pp. 581 - 585

D6: Japanese Journal of Applied Physics, Vol. 29 (1990), No. 4, pp. L576 - L579

D8: Japanese Journal of Applied Physics, Vol. 29 (1990), No. 3, pp. L412 - L415

D9: Japanese Journal of Applied Physics, Vol. 29 (1990), No. 2, pp. L254 - L257

D10: EP-A-0 447 994

The opposition division came to the conclusions

- that the claims as amended met the requirements of Article 123(2) and (3) EPC,
- that the subject-matter of the amended claims was novel over the disclosure of D10,
- and that, taking the process disclosed in D2 as the closest prior art, and considering the disclosures of D6 and D8, the method of claim 1 was not obvious.

III. The appellant (opponent) maintained his earlier objections, i.e.

- lack of novelty over the disclosure of D10
- lack of inventive step over the method disclosed in D2 in combination with the disclosure of D6 and the common general knowledge as illustrated by D8 and D9.

He filed five further documents to support his view that D10 at least implicitly disclosed all of the features of claim 1 of the patent as maintained.

IV. In response to a communication accompanying the summons to oral proceedings, the respondent (patent proprietor) filed two sets of amended claims as basis for a main and an auxiliary request. Claim 1 according to the first set (main request) reads as follows:

"A method of preparing a bismuth superconductor comprising a 2223 phase with a longitudinally oriented a-b plane as a matrix, while a superconducting phase mainly composed of a 2212 phase and/or non-superconducting phases are dispersed along the a-b plane of the 2223 phase, comprising the steps of mixing raw materials for forming a bismuth superconductor with each other in a grinding system to obtain mixed powder, heat treating said mixed powder, pulverizing said mixed powder and then covering said mixed powder with a metal sheath, and then performing deformation processing and heat treatment, said mixed powder to be covered with said metal sheath being prepared to have a 2223 composition in a composition of Bi-Sr-Ca-Cu or (Bi,Pb)-Sr-Ca-Cu and to contain a superconducting phase

being mainly composed of a 2212 phase, and pulverized into a mean particle diameter of not more than 1 μm in such a time that 2212 phase is not converted to an amorphous state."

V. Oral proceedings took place on 28 September 2001.

VI. The appellant's oral and written submissions may be summarised as follows:

Concerning novelty over D10, he argued that in the technical field concerned blending would **usually** be carried out in grinding systems. D10 would therefore induce the skilled person having common general knowledge to use such systems.

Concerning inventive step, he accepted the technical problem as stated in the contested patent and acknowledged that the results reported in the examples can be achieved by the claimed method. He argued that starting from the method of D2, the use of particles ground to submicron size would be obvious in view of D6. Although D6 refers to a mean particle size of 1.8 μm , the skilled person would not consider this value as an absolute lower limit. D2 and D9 would illustrate that in the technical field in question, it was generally desirable to use as fine powders as possible, and to avoid transformations into the amorphous state. Moreover, in view of D2 and D8, the skilled person would be aware of the possible consequences of grinding on the crystallinity of the superconducting phases. D8 would show that the duration of grinding was not critical with respect to amorphisation in the case of 2212 powder.

VII. The respondent contested these arguments.

Concerning D10, he argued that the feature of "mixing the raw materials in a grinding system" was not derivable directly and unambiguously from that document, since "blending" does not necessarily imply "grinding".

Concerning inventive step, he submitted

- that the distinguishing features of claim 1, i.e. the pulverisation to submicron particles "in such a time that 2212 phase is not converted to an amorphous state", is not disclosed or made obvious by D2, D6 or D8;
- that although it is known from D9 that fine powders are required to obtain dense and uniform sintered bodies, both D8 and D9 would teach away from carrying out drastic size reduction, in order to avoid amorphisation; and
- that the criticality of the duration of the pulverisation was demonstrated by the comparative examples of the patent, and
- that due to the non-obvious features of the method of claim 1, the critical current density of the products obtained by the claimed method is surprisingly superior to the ones obtained according to the prior art.

VIII. The appellant requested that the decision under appeal be set aside and that the patent be revoked.

The respondent requested that the appeal be dismissed and that the patent be maintained on the basis of the claims submitted as main or auxiliary request, respectively, with the letter of 27 August 2001.

Reasons for the Decision

1. Since all of the amendments are based on the documents as originally filed, and since the amendments carried out after the grant of the patent restrict the scope of the claims, the board is satisfied that the requirements of Articles 123(2) and (3) are met. This not being in dispute, no reasons for this finding need to be given.

2. *Novelty (main request)*
 - 2.1 Document D10 has been published (25 September 1991) after the filing date (15 March 1991) of the contested patent but has earlier priority dates (16 March 1990 and 27 March 1990) than the contested patent (16 July 1990). The validity of the priorities claimed by D10 has never been contested and is also accepted by the board. The same contracting states are designated in D10 and in the contested patent. D10 thus belongs to the state of the art according to Article 54(3) and (4) EPC and may only be considered relating to novelty.

 - 2.2 The processes disclosed in D10 comprise the preparation of a Bi(Pb)-Sr-Ca-Cu "raw material", from a blend of powders present in amounts corresponding to a 2223

composition. This "raw material", which comprises a superconducting phase mainly consisting of a superconducting 2212 phase and a non superconducting phase, is then filled in a silver pipe and subjected to mechanical and thermal treatments (claims 6 and 8, column 3, line 52 to column 4, line 5). According to the experimental examples 1 and 2 of D10, the powder to be filled in the silver pipe is obtained by blending Bi_2O_3 , Pb or PbO, SrCO_3 , CaCO_3 and CuO in a first step, followed by three heat treatments at different temperatures from 700 to 800°C. The blend is pulverised after each of these heat-treatments. The powder obtained is further pulverised with a ball mill to obtain a submicron powder (the "raw material") which is filled in the silver pipe (see also column 4, lines 15 to 19).

- 2.3 According to the contested patent, the term "raw materials" is used to designate the individual starting components such as oxides or carbonates. The term "mixed powder" is used in claim 1 of the contested patent to designate the material obtained after mixing the blend of the starting components in a grinding system. It is this "mixed powder" which is then further subjected to heat treatments and pulverisation. See column 2, lines 57 to column 3, line 3, column 3, lines 22 to 40 and column 4, lines 22 to 26.

On the other hand, according to D10, the "raw material" to be filled into the silver pipe comprises a superconducting 2212 phase. It is clear that this 2212 phase is formed during the high temperature heat treatments of the starting components described in the examples by a solid state reaction (see e.g. D2, page 582, "Experimental details", the first two

sentences). The appellant's mere allegation, during oral proceedings, that the heat treatments of the blend as described in the examples of D10 only serve to improve the rheological properties of the powders as starting components is thus in contradiction with the fact that the powder to be filled in the pipe comprises 2212 phase and cannot, therefore, be accepted.

The pulverisation steps described in the examples of D10 are carried out on the heat treated blend of the starting components, which cannot - as alleged by the appellant - be considered as "raw materials" in the sense of claim 1 for the reasons given here above. Therefore, the pulverisation of the heat treated blend is not identical with "mixing raw materials in a grinding system", even if it were to be accepted that a pulverisation device can always be considered as a kind of grinding system.

- 2.4 The appellant also argued that the first "blending" step mentioned in the examples of D10 would be understood by the skilled person as referring to the use of a mill, for mixing and grinding the starting powder materials in one and the same step. Although the board accepts that, as repeatedly pointed out by the appellant, blending is **often** carried out in some kind of grinding equipment in the technical field concerned, and that any grinding leads to an improved mixing of the ground components, this does not necessarily imply that blending will **usually**, let alone always be carried out in a grinding system. This is underlined by the fact that two different terms ("blending" and "pulverising") are used in the examples of both the patent and D10. Simply blending powders of appropriate particle size will require less time and energy than

grinding them.

Hence, although the appellant's submission that "blending" as mentioned in the examples of D10 **can** be carried out in a grinding system is accepted, the board cannot accept that D10 comprises a clear and unambiguous implicit technical teaching of performing the step comprising the blending of the "raw materials" (in the sense of the contested patent) in a grinding system.

2.5 Since the features of "mixing of the raw materials for forming a bismuth superconductor with each other in a grinding system" in the sense of claim 1 are not disclosed in D10, the subject-matter of this claim is novel in view of D10 (Article 54(1) and 54(3) EPC).

3. *Inventive step (main request)*

3.1 In agreement with the parties, D2 can be considered to represent the closest prior art. D2 discloses a method for obtaining a superconducting tape by the so-called "powder-in-tube" technique. The superconducting material obtained is based on a Bi/Pb-Sr-Ca-Cu composition comprising mainly the "2223 phase" ("high T_c phase", "110 K phase"). According to the "Experimental details" given in D2, a calcined powder composed of a 2212 phase ("low T_c phase", "80 K phase") and obtained by a solid state reaction from oxides and carbonates at 800°C was filled in a silver tube. The filled tube was drawn and then subjected to further mechanical and thermal treatment. The desired 2223 phase was formed through a partial melt reaction occurring during the final heat treatment of the filled tube. The products obtained according to D2 have critical current

densities (" J_c " in the following) of 2500 A/cm² and have an a-b plane oriented structure composed mainly of the 110 K phase and including minor amounts of the 80 K and unknown phases. See in particular the abstract on page 581, page 582, left-hand column, first and second full paragraphs, Figure 3 and the corresponding text in the paragraph bridging pages 582 and 583. D2 does not address any kind of grinding or size reduction of the powder materials used before their filling into the silver tube.

- 3.2 As indicated in the contested patent (column 1, lines 52 to 56 and column 2, lines 51 to 54), and in agreement with the parties, the technical problem to be solved by the present invention can be seen in providing a "powder-in-tube" method for preparing bismuth superconductors having higher critical current densities than the prior art superconductors disclosed in D2, particularly in a magnetic field.

It has not been contested that the method as claimed, comprising as a distinctive feature the pulverisation of the material to be filled in the tube to an average particle diameter of below 1 μ m without amorphisation of the superconducting 2212 phase, leads to the superior properties evidenced by the examples of the patent (J_c values of 19000 to 58000 A/cm²). Hence, it remains to be seen whether the claimed solution of the stated technical problem could be derived in an obvious manner from the prior art.

- 3.3 D2 itself only refers to submicron particles in connection with a less preferred preparation method comprising swaging, rather than drawing, the filled silver tube (page 582, left-hand column, section

"Experimental details", 2nd paragraph). This swaging leads to submicron particles and induces the formation of an amorphous phase. The corresponding final product showed less texturing structure and a smaller J_c value than the other products obtained. Although this reference to submicron particles does not relate to a pulverisation of the material to be filled into the tube, the products obtained are described as having undesirably low J_c values. Therefore, the board holds that the skilled person would not, in view of D2 alone, consider the pulverisation of the material to be filled in the tube to an average particle diameter of below $1 \mu\text{m}$ as a possible way of improving the current density values of the final products.

- 3.4 Document D6 does not relate to a "powder-in-tube" method, but is concerned with the preparation of high T_c (Bi,Pb)-Sr-Ca-Cu superconductors by the conventional ceramic technique. The preparation method comprises preparing a mixed oxide by a solid state reaction involving mixing, milling and heat treating oxide and carbonate powders at 800 to 820°C in amounts corresponding to a 2223 composition. This obtained powder is further ball-milled for size reduction to obtain an average particle size of $1.8 \mu\text{m}$. The powder is then pelletised and sintered at 845°C to form the 110 K phase. See page L576, section "Experimental Techniques", 1st paragraph. Whereas the products obtained according to the contested patent have been found to require the presence of dispersed low T_c phase and/or non superconducting phases in order to achieve high J_c values (see contested patent, column 2, lines 37 to 40 and column 5, lines 1 to 7), the aim of D6 is to obtain dense single phase high T_c materials with no other phases present. Table I and figures 2,4 and 5,

together with the corresponding text show that the J_c values, the high T_c phase content and the J_c value of the final product increase with increasing sintering time, the product with the highest T_c content and J_c value being a single phase product consisting of the high T_c superconductor. However, the optimum J_c values are reported to range from only 1000 to 1200 A/cm² (see the abstract on page L576 and the "Conclusions" on page L579). Hence, the skilled person could not derive from D6 that by reducing the average particle size of the 2212 phase material to be filled in the tube in a method according to D2 to about half the average particle size disclosed in D6, the specific structures and the very high corresponding J_c values as reported in the contested patent could be obtained.

- 3.5 Documents D8 and D9 are both concerned with the effects of mechanical grinding on powders comprising mainly the 2223 phase. According to both documents, it was found that the accumulation of crystal strain leads to the amorphisation of the 2223 phase during the grinding of the powders, and hence to a loss in superconductivity. It was concluded that grinding is not a suitable method for obtaining fine 2223 phase superconducting powders to be used in the preparation of high T_c bodies. See D8, the abstract on page L412 and page L415, right-hand column, lines 4 to 10 from the bottom, and D9, the abstract on page L254 and the "Conclusions" on page L256.

More particularly, it can be taken from D8 that both the (main) 2223 and the (minor) 2212 phase are subject to amorphisation during extended grinding, with the 2223 phase being more sensitive to mechanical grinding than the 2212 phase (figures 5 and 6 and

corresponding text). Figure 8 of D8 does not, as argued by the appellant, indicate that the minor 2212 component would be less prone to amorphisation. Rather, this figure appears to show that in specific compositions, and under specific annealing conditions, the amorphous minor 2212 component may recover its superconducting properties to a larger degree than the amorphous main 2223 component, irrespective of the duration of the previous grinding.

D9 confirms that the amorphisation of the 2223 phase after prolonged grinding is difficult to reverse by an annealing heat treatment. See §2 and §3 on pages L254 to L256, and more particularly the discussion of figures 7 and 8 on page L256.

D8 and D9 do not mention the "powder-in-tube" method, they do not report any J_c values, and they investigate a different approach to superconductor fabrication based on starting powders essentially comprising the 2223 phase, and not - as in the claimed process - essentially comprising the 2212 phase. Moreover, the comparative examples of the contested patent show that the properties of the final products deteriorate with prolonged grinding. Hence, although the need for fine powdered starting materials is mentioned in D8 and D9, as well as experimental size reductions down to submicron particles, and although it appears that the amorphisation of the 2212 phase can, under certain conditions, be reversed, it cannot be inferred from these documents without the benefit of hindsight that, in a process as described in D2, the pulverisation of the powders mainly comprising 2212 phase to a very low average particle diameter of less than 1 μm without amorphisation, before the filling thereof in a metal

sheath, would lead to the superior current densities of the final products evidenced by the examples of the patent.

3.6 The board is convinced, and it was not disputed, that the other documents cited by the appellant do not come closer to the invention and do not contain any more relevant information.

3.7 Therefore, the claimed method, and more particularly the feature of "pulverising (the mixed powder) into a mean particle diameter of not more than 1 μm in such a time that 2212 phase is not converted to an amorphous state", is not obvious in the light of the documents cited, taken alone or in combination, and the subject-matter of claim 1 is thus based on an inventive step (Articles 52(1) and 56 EPC).

4. The dependent claims 2 and 3 according to the respondent's main request are narrower in scope than claim 1 and concern specific embodiments of the invention. Their subject-matter is thus novel and inventive as well.

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 with the following

documents:

- claims 1 to 3 submitted as main request with letter of 27 August 2001,
- a description to be adapted.

The Registrar:

The Chairman:

C. Eickhoff

R. Spangenberg