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**D E C I S I O N**  
**of 21 November 2001**

**Case Number:** T 0946/98 - 3.4.3

**Application Number:** 89115862.8

**Publication Number:** 0356969

**IPC:** H01L 39/14

**Language of the proceedings:** EN

**Title of invention:**

Method of producing oxide superconductor

**Patentee:**

SUMITOMO ELECTRIC INDUSTRIES, LTD.

**Opponent:**

Siemens AG

**Headword:**

Bi-Pb-Sr-Ca-Cu-O superconductor/SUMITOMO

**Relevant legal provisions:**

EPC Art. 100, 54, 56

**Keyword:**

"Late filed document (admitted) - useful for understanding a  
prior art document"

"Inventive step (yes)"

**Decisions cited:**

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**Catchword:**

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Case Number: T 0946/98 - 3.4.3

**D E C I S I O N**  
**of the Technical Board of Appeal 3.4.3**  
**of 21 November 2001**

**Appellant:** Siemens AG  
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**Representative:** Schmuckermaier, Bernhard  
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**Respondent:** SUMITOMO ELECTRIC INDUSTRIES, LTD.  
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**Representative:** Winter, Brandl, Fürniss, Hübner, Röss,  
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**Decision under appeal:** Decision of the Opposition Division of the  
European Patent Office posted 28 July 1998  
rejecting the opposition filed against European  
patent No. 0 356 969 pursuant to Article 102(2)  
EPC.

**Composition of the Board:**

**Chairman:** R. K. Shukla  
**Members:** G. L. Eliasson  
M. B. Günzel

## Summary of Facts and Submissions

I. This appeal lies from the decision of the opposition division dated 28 July 1998 rejecting the opposition against European patent No. 0 356 969 pursuant to Article 102(2) EPC.

Claim 1 as granted reads as follows:

"1. A method of producing an oxide superconductor of Bi-Pb-Sr-Ca-Cu by thermally treating raw material, said method comprising the steps of:

preparing said raw material to mainly generate superconducting phases having a low critical temperature;

charging said raw material in a metallic sheath;  
performing first plastic deformation by at least one of rolling and pressing on said raw material charged in said sheath to make said sheath into a tape-like shape and increase density of said material;

performing first heat treatment on said material being subjected to said first plastic deformation within a temperature range of 780 °C to 860 °C to generate superconducting phases having a high critical temperature;

performing second plastic deformation with reduction of sectional area of at least 10 % by at least one of rolling and pressing on said material cooled after said first heat treatment to further increase density of said material; and

performing second heat treatment on said material subjected to said second plastic deformation within a temperature range of 780 °C to 860 °C to more strongly bonding crystal grains

in said material."

Claims 2 to 4 as granted are dependent claims.

II. The opposition was filed against the patent as a whole and based on Article 100(a), (b), and (c) EPC. Of the documents cited in the opposition proceedings, the following document was disregarded by the opposition division under Article 114(2) EPC:

D13: YAMAMOTO et al. "Effect of the Fabricating Process on the Superconducting Properties of Bi-Pb-Sr-Ca-Cu-O Tapes by the Powder-in-Tube Method" In: Ishiguro-Kajimura (Eds.) Advances in Superconductivity II: Proceedings of the 2nd International Symposium on Superconductivity (ISS 89) 14 to 17 November 1989, Tsukuba (JP), Springer Verlag 1990.

III. The appellant (opponent) filed the notice of appeal on 15 September 1998, paying the appeal fee the same day. A statement of the grounds of appeal was filed on 25 November 1998 together with the following new documents, published after the earliest priority date of the patent in suit, as evidence supporting the appellant's submissions:

D14: Superconductor Science and Technology, vol. 4 (1991), pages 165 to 171; and

D15: Superconductivity News, vol. 4, No. 6, 19 December 1988, pages 1 to 5.

IV. With a letter dated 24 February 1999, the respondent (patentee) filed new claims 1 to 4 forming an auxiliary

request.

- V. At the oral proceedings held on 21 November 2001, the appellant stated that he no longer maintained the grounds for opposition under Article 100(b) and (c) EPC. As to the remaining ground for opposition under Article 100(a) EPC, the appellant stated that he would only maintain the arguments demonstrating lack of inventive step having regard to the the following document D3 cited during the opposition proceedings and document D13:

D3: SEKINE et al. "Studies on the Non Rare Earth Oxide Superconductors Fabricated by Sintering" In: 1988 Applied Superconductivity Conference", 21 to 25 August 1988, San Francisco, CA (US), IEEE Transactions on Magnetics, vol. 25, No. 2, March 1989, pages 2164 to 2167.

- VI. The parties made the following requests:

The appellant requested that the decision under appeal be set aside and that the European patent No. 0 356 969 be revoked.

The respondent requested as a main request that the appeal be dismissed, and as an auxiliary request, that the decision under appeal be set aside and the patent be maintained with the claims of the auxiliary request filed with his letter dated 24 February 1999.

- VII. The arguments made by appellant can be summarized as follows:

(a) Document D13 which was not admitted into the

opposition proceedings by the opposition division, is relevant in the consideration of inventive step, since it shows the phase diagram of the Bi-Pb-Sr-Ca-Cu-O superconductor, and thereby establishes which superconducting phases were present in the samples disclosed in document D3.

- (b) The sample (a) referred to in Figure 5 of document D3 was prepared by calcining the precursor powder at 800°C for 14 hours before the powder was charged in a silver tube (cf. page 2166, left hand column). It follows from the phase diagram shown in Figure 1 of document D13 that, after such a heating step at 800°C, the low critical temperature phase (2212) will *inevitably* form. The high critical temperature phase (2223), on the other hand, will only form during sintering at about 845°C for much longer periods of time than 14 hours.
- (c) Although the sample (a) in document D3 did not show a superconducting transition, the authors of document D3 offered the explanation that sample (a) contained too much lead (cf. page 2166, left hand column, last paragraph). Since the method of preparing sample (a), in contrast to that of sample (b), does not involve a long sintering step of the precursor powder lasting for hundred hours before charging the powder in the silver tube, the skilled person would consider the method used for producing sample (a) to be advantageous. Therefore, the skilled person would try to improve the method of sample (a) by controlling the lead content more carefully.

(d) It furthermore appears that the raw material of sample (b) of document D3 contained mainly the superconducting phase 2212 having a low critical temperature, since the raw material was subject to sintering at 845°C for only 100 hours after the above-mentioned calcination step (800°C for 14 hours). From Table 3 of the patent in suit, it appears that a sintering time of about 200 to 400 hours is required to convert the raw material so that it contains mainly the high critical temperature phase 2223 (cf. Table 3, Reference Examples IV-1 and IV-2), whereas a sintering time of only 8 to 20 hours would yield a raw material containing mainly the low critical temperature phase 2212 (cf. Table 3, Examples IV-1 to IV-4). As is apparent from the phase diagram of Figure 1 of document D13, the formation of the high critical temperature phase 2223 takes place *by first forming the low critical temperature phase 2212*. Since the sintering time of 100 hours for sample (b) of document D3 lies in the middle between the values of 20 hours and 200 hours disclosed in the patent in suit for sintering times in order to form the low critical temperature phase 2212 and the high critical temperature phase 2223, respectively, it appears that the raw material of sample (b) of document D3 must contain at least a significant amount of the low critical temperature phase 2212.

VIII. The arguments of the respondent can be summarized as follows:

(a) The respondent does not agree to the introduction of document D13 into the appeal proceedings, since

firstly, it was late filed and therefore not admitted into the opposition proceedings. Secondly, it is published after the priority date of the patent in suit. Thirdly, since the phase diagram shown in Figure 1 did not belong to the common knowledge in the art at the priority date of the patent in suit, any argument on inventive step using the phase diagram amounts to an unfair, ex-post facto analysis of the claimed invention.

- (b) The method according to claim 1 contains the crucial step of having the raw material so prepared that it mainly contains the low critical temperature phase (2212) before it is charged into a metallic sheath. As demonstrated in Table 3 of the patent in suit, the claimed method has the advantage that the superconductors produced according to the method as claimed in the patent in suit, have a critical current density  $J_c$  which is substantially increased compared to the current density of the superconductors produced with a method where the raw material contains mainly the high critical temperature phase (2223) (cf. D3, abstract).
  
- (c) As to the disclosure in document D3, sample (a) does not show a superconducting transition (cf. Figure 5). Therefore, the raw material of sample (a) could not have contained mainly the low critical temperature phase (2212), but was formed mainly of non-superconducting phases. Sample (b) of document D3 was subject to sintering at 845°C for 100 hours in addition to the 14 hours of calcination which was used in sample (a). Therefore, the raw material of sample (b) which



was charged into the silver tube must have contained mainly the high critical temperature phase (2223). This is also evident from the fact that the highest critical current density  $J_c$  reported in document D3 is 1100 A/cm<sup>2</sup> which should be compared to the critical current density of about 10000 A/cm<sup>2</sup> obtained from samples produced according to the claimed method (cf. the patent in suit, Table 3; D3, abstract).

### **Reasons for the Decision**

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is therefore admissible.
  
2. *Late filed documents*
  - 2.1 Document D13 was filed after the expiry of the opposition period as laid down in Article 99(1) EPC in conjunction with Rule 55(c) EPC. In the decision under appeal, it was not admitted into the opposition proceedings, since having regard to the nominal composition of the Bi-Pb-Sr-Ca-Cu-O superconductor disclosed in D3 which is different from the composition used in document D13 (cf. D3, page 2164, right hand column, first paragraph; D13, Figure 1), the latter was not considered to be prima facie relevant.

The Board agrees however with the appellant that document D13 is relevant for understanding the disclosure in document D3 regarding the phases of the Bi-Pb-Sr-Ca-Cu-O superconductor which were likely to be present in the samples disclosed in document D3.

It has however to be kept in mind, as the respondent correctly pointed out (cf. item VIII(a) above), that the phase diagram of Bi-Pb-Sr-Ca-Cu-O as disclosed in document D13 did not form part of the general technical knowledge at the priority date of the patent in suit, since the content of document D13 was disclosed at a conference held on 14 to 17 November 1989 which is later than all the claimed priority dates. Therefore, the phase diagram can only be used as evidence to support submissions regarding the presence of various phases which would inevitably be present following the prior art methods disclosed in document D3.

Keeping the above reservations in mind, the Board therefore decides to admit document D13 into the appeal proceedings.

3. *Inventive step - Main request*

The only substantive issue remaining in the present case is that of inventive step having regard to document D3.

- 3.1 Document D3 discloses a method of forming superconducting wires made of Bi-Pb-Sr-Ca-Cu-O packed in silver tubes or sheaths. Before the raw material is charged in the metal tube or sheath, it is calcined and/or sintered. The calcination is carried out at 800°C for 14 hours and the first sintering step is carried out at 845°C for 100 hours (cf. page 2164 "Experimental"). In Figure 5, the transition curves of two samples, "(a)" and "(b)", are presented: Sample (a) was made of raw material which was calcined but not sintered before being charged in a silver tube, whereas the raw material of sample (b) was subject to both

calcination and sintering. After the raw material was charged into the silver tube, both samples (a) and (b) were drawn into wires, i.e. plastically deformed, and sintered at 845°C for 50 hours (cf. page 2166, right hand column). As shown in Figure 5, however, sample (a) failed to show any superconducting transition above 50 K. As an explanation for the absence of a superconducting transition, a too high content of lead was conjectured as a possible cause. Sample (b) which was sintered before being charged into a silver tube showed a transition at about 110 K.

In addition, document D3 discloses other samples which were repeatedly subject to plastic deformation and sintering.

3.2 The appellant argued that since sample (a) of document D3 was subjected only to a calcination treatment at 800°C for 14 hours, the raw material put into the silver tube **inevitably** contained mainly the low critical temperature phase 2212 (cf. item VII(b) above). This, according to the appellant, was apparent from the phase diagram shown in Figure 1 of document D13, from which it appeared that the high critical temperature phase 2223 could not form during a sintering treatment at 800°C. The high critical temperature phase 2223, on the other hand, required sintering at about 850°C for a very long time (of the order of hundred hours).

3.2.1 As the respondent has pointed out, however, no superconducting transition above 50 K can be seen for sample (a) in Figure 5 of document D3, where the A.C. susceptibility is shown for samples (a) and (b) as a function of temperature (cf. item VIII(c) above). A too

high content of lead was conjectured in document D3 as a possible explanation for the absence of a superconducting transition (cf. D3, page 2166, left hand column, last paragraph).

On the other hand, It is disclosed in the patent in suit that the raw material containing mainly the low critical temperature phase 2212 was not only calcined for 8 to 24 hours at 800°C, but was also sintered for 8 to 20 hours at 860°C (cf. Table 3), i.e. a substantially longer heating treatment than that used for the raw material of sample (a) of document D3.

3.2.2 Since it was speculated in document D3 that sample (a) contained too much lead, the Board also has doubts whether the phase diagram of the Bi-Pb-Sr-Ca-Cu-O compound disclosed in Figure 1 of document D13 is of any relevance for the sample (a) of document D3, since the phase diagram shown in Figure 1 of document D13 is constructed from a mixture having the Bi/Pb proportion equal to 1.4/0.6, a lead content which evidently is not too high.

3.2.3 Thus, in accordance with the respondent's submissions, and in the light of the apparent differences in the treatment of the raw materials as disclosed in the patent in suit and in document D3, the Board comes to the conclusion that the raw material of sample (a) of document D3 which was charged into the silver tube contained mainly non-superconducting phases.

3.3 The appellant further argued that the raw material of sample (b) of document D3 would contain mainly the low critical temperature phase 2212, since the raw material was subject to sintering at 845°C for "only" 100 hours

after the above-mentioned calcination step (800°C for 14 hours). According to the patent in suit, however, a sintering time of about 200 to 400 hours is required for converting the raw material to the high critical temperature phase 2223 (cf. item VII(d) above). Since the sintering time of 100 hours for sample (b) of document D3 lies in the middle between values of 20 hours and 200 hours disclosed in patent in suit for sintering times in order to form the low critical temperature phase 2212 and the high critical temperature phase 2223, respectively, it appears that the raw material of sample (b) of document D3 must have contained at least a significant amount of the low critical temperature phase 2212.

- 3.3.1 Although the Board accepts the argument that the raw material of sample (b) of document D3 may have contained **some** low critical temperature phase 2212 after 100 hours of sintering, the appellant has failed to show that the raw material of sample (b) contained **mainly** the low critical temperature phase, and the Board sees no arguments or facts that would support this contention. On the contrary, Figure 5 of document D3 shows a sharp transition near 100 K indicating that sample (b) as a finished product contained mainly the high critical temperature phase 2223. It is furthermore an undisputed fact that a long sintering treatment at about 850°C is required in order to obtain a sample containing mainly the high critical temperature phase 2223. For example, the sintering times disclosed in the patent in suit for converting the superconductor to the high critical temperature phase 2223 are between 200 and 400 hours for this treatment (cf. Table 3). In contrast, sample (b) of document D3 was subject to a sintering treatment for only 50 hours at 845°C after

the raw material was charged into the silver tube and the sample was drawn into a wire (cf. D3, page 2166, left hand column, last paragraph). Thus, if as submitted by the appellant, the raw material of sample (b) mainly contained the low critical temperature phase 2212 when it was charged into the silver tube, then, in the Board's view, the subsequent single sintering treatment for 50 hours would not be sufficient to convert most of the raw material into the high critical temperature sample 2223.

3.3.2 This conclusion that the raw material of sample (b) of document D3 contained mainly the high critical temperature phase 2223 when it was charged into the silver tube is also confirmed by the results disclosed in document D13. Thus, in Table I, it is disclosed that a sintering/calcination treatment for 100 hours at 835°C yielded a raw material having the high critical temperature phase 2223 (cf. Table I, samples A, B), whereas a sintering/calcination treatment for 40 hours at 800°C yielded a material containing mainly the low critical temperature phase 2212 (cf. Table I, samples C, D).

3.4 From the above, it follows that document D3 does not disclose a method containing the step of preparing the raw material to mainly generate superconducting phases having a low critical temperature.

3.5 The technical problem addressed by the patent in suit relates to preparing a high critical temperature superconductor of the Bi-Pb-Sr-Ca-Cu-O type having a high critical temperature and a high critical current density (cf. patent in suit, patent in suit, page 2, lines 56 to 58). From the description on page 5,

lines 5 to 12 and lines 15 to 36, and the results shown in Table 3 of the patent in suit, it follows that by preparing the raw material so that it contains mainly superconducting phases having a low critical temperature, and then subjecting the prepared raw material in a metallic sheath to heat treatments and plastic deformations as set out in claim 1 of the patent in suit, the claimed method results in the formation of superconducting phases having a high critical temperature and high critical current density.

3.6 In document D3, the preparation of the raw material was only discussed with reference to the samples (a) and (b), but without any analysis of the raw materials subjected to the different preparation methods. Only sample (b) exhibited a high superconducting transition temperature, but this sample was prepared using a raw material which contained mainly the high critical temperature phase 2223. Thus, the teaching in document D3 concerning sample (b) does not render obvious the use of raw material containing mainly a low critical temperature phase 2212.

3.6.1 Also the appellant's argument that a skilled person would consider the method used for producing sample (a) for the reason that the preparation time is shortened, does not convince the Board (cf. item VII(c) above), since following the short sintering time employed in the preparation of sample (a), the sample does not show a superconducting transition. Also in connection with sample (a), document D3 merely suggests to control the lead content with a view to obtaining a high critical temperature phase. There is however no suggestion to use a raw material containing mainly a low critical temperature phase and to subject the raw material to

heating and plastic deformation as set out in the claimed method with a view to obtaining a substantially high critical current density.

Furthermore, as acknowledged by both parties, the sintering time for converting material to the high critical temperature phase 2223 is very long. Therefore, the skilled person would recognize that if the raw material was not subject to a long sintering step, the sample would nevertheless have to be sintered for a long time after being charged into a metal sheath, so that the final product will have the desired high transition temperature.

- 3.7 Therefore, in the Board's judgement, the subject matter of claim 1 according to the respondents main request involves an inventive step within the meaning of Article 56 EPC. The requirements of Article 52(1) EPC are therefore met.

## **Order**

### **For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

The Chairman:

D. Spigarelli

R. K. Shukla