

**Internal distribution code:**

- (A) [ ] Publication in OJ  
(B) [ ] To Chairmen and Members  
(C) [X] To Chairmen  
(D) [ ] No distribution

**D E C I S I O N**  
**of 7 May 2003**

**Case Number:** T 0238/98 - 3.3.5

**Application Number:** 89103220.3

**Publication Number:** 0330214

**IPC:** C04B 35/00

**Language of the proceedings:** EN

**Title of invention:**

High-Tc oxide superconductor and method for producing the same

**Patentee:**

KABUSHIKI KAISHA TOSHIBA

**Opponent:**

Siemens AG  
Hoechst AG  
Sumitomo Chemical Co., Ltd.

**Headword:**

Superconductor/TOSHIBA

**Relevant legal provisions:**

EPC Art. 123(2), 54, 56

**Keyword:**

"Public availability of pre-print (no) - not proven beyond any reasonable doubt"

**Decisions cited:**

T 0150/82, T 0248/85, T 0011/99, T 0219/83, T 0642/92

**Catchword:**

-



Case Number: T 0238/98 - 3.3.5

**DECISION**  
of the Technical Board of Appeal 3.3.5  
of 7 May 2003

**Appellant 1:** Siemens AG  
(Opponent 1) Postfach 22 16 34  
D-80506 München (DE)

**Representative:** -

**Appellant 2:** Hoechst AG  
(Opponent 2) Patent- und Lizenzbteilung, Geb. K801  
D-65926 Frankfurt am Main (DE)

**Representative:** Ahrens, Gabriele, Dr  
Patentanwälte Einsel & Kollegen  
Jasperallee 1A  
D-38102 Braunschweig (DE)

**Opponent 3:** Sumitomo Chemical Co., Ltd.  
(Opponent 3) 5-33. Kitahama 4-chome  
Chuo-ku  
Osaka-shi  
Osaka 541 (JP)

**Representative:** Boeters, Hans Dietrich, Dr  
Patentanwälte Boeters & Bauer  
Bereiteranger 15  
D-81541 München (DE)

**Appellant 3:**  
(Proprietor of the patent)      KABUSHIKI KAISHA TOSHIBA  
72, Horikawa-cho,  
Saiwai-ku  
Kawasaki-shi,  
Kanagawa-ken 210-8572      (JP)

**Representative:**      Hansen, Bernd, Dr. Dipl.-Chem.  
Hoffmann Eitle  
Patent- und Rechtsanwälte  
Postfach 81 04 20  
D-81904 München      (DE)

**Decision under appeal:**      Interlocutory decision of the Opposition  
Division of the European Patent Office posted  
15 January 1998 concerning maintenance of  
European patent No. 0330214 in amended form.

**Composition of the Board:**

**Chairman:**      R. Spangenberg  
**Members:**      M. M. Eberhard  
                  M. B. Günzel

## Summary of Facts and Submissions

- I. The three appeals are from the decision of the opposition division according to which European patent No. 0 330 214 in amended form meets the requirements of the CBE. The patent claiming the priority date of 24 February 1988 was granted in response to European application No. 89 103 220.3. The decision was based on the amended set of claims filed with the letter dated 2 October 1996, as the main request, and on two sets of claims submitted on 10 October 1997 as the 1st and 2nd auxiliary requests.
- II. During the opposition proceedings appellants 1 and 2 (opponents 1 and 2 respectively) relied *inter alia* on the following documents:
- D1: EP-A-0 332 291
  - D4: Solid State Communications, Vol. 17, pages 27 to 28, 1975, A.W. Sleight et al.
  - D7: Japanese Journal of Applied Physics, Vol. 27, No. 2, 20 February 1988, pages L209 to L210, H. Maeda et al.
  - D7a: Maeda's pre-print
  - D8: Applied Physics Letters, 51, No. 22, 1987, pages 1854 to 1856
  - D10: Pre-print "Structure and Physical Properties of Single Crystals of the 84K Superconductor  $\text{Bi}_{2.2}\text{Sr}_2\text{Ca}_{0.8}\text{Cu}_2\text{O}_{8+}$ ", S.A. Sunshine et al., 15 pages
  - D10a: First page of pre-print D10
  - D13: Superconductor Week, Vol. 2, No. 6, 8 February 1988, page 1

D18: Europ. Workshop on High  $T_c$  Superconductors and potential applications, Proceedings, 1-3 July 1987, Genova, pages 45, 48, 54, B. Raveau, C. Michel

The opposition division took the view that claim 1 of the main request did not meet the requirements of Article 123(2) EPC and that the subject-matter of claim 9 according to the 1st auxiliary request lacked novelty over the disclosure of D1. The claims of the 2nd auxiliary request were considered to involve an inventive step. There was no sufficient evidence that the Sunshine et al. pre-print D10 was available to the public before the priority date and therefore D10 could not be taken into consideration. It was not obvious in view of D18 to substitute Bi with Pb in the method for forming Bi-Sr-Ca-Cu-O superconductors known before the priority date.

III. Appellant 3 (proprietor of the patent) filed amended claims 1 to 13 with their grounds of appeal dated 22 May 1998, as a basis for three different requests. An additional set of amended claims was filed with the letter dated 15 December 1998 as the main request; the first, second and third auxiliary requests being based on the claims filed with the letter of 22 May 1998, namely claims 1 to 13, claims 1 to 9 and claims 1 to 8 respectively. A fourth and fifth set of auxiliary requests were filed on 14 March 2003. Claim 1 of the main request reads as follows:

"1. A method for preparing an oxide superconductor including Bi, Sr, Ca, Cu, Pb and having a  $T_c$  of greater than 77K comprising the step of :

mixing appropriate amounts of chemical compounds of Bi, Sr, Ca, Cu, Pb and O to obtain a chemical composition in which the molar ratio of (Bi+ Pb):Sr:Ca is 1:1:1 and the molar ratio of Pb to Bi is in the range of 5:95-85:15 to prepare a mixture; calcining the mixture; grinding and then cold-pressing the calcined mixture; and sintering the pressed mixture."

Claims 1, 3 and 9 of the 1st and 2nd auxiliary requests have the following wording:

"1. A method for preparing an oxide superconductor including Bi, Sr, Ca, Cu, Pb and O having a  $T_c$  of greater than 77K and in which the molar ratio of Pb to Bi is in the range of 5:95-85:15 comprising the step of:

mixing appropriate amounts of powdered  $\text{Bi}_2\text{O}_3$ ,  $\text{Sr}_2\text{CO}_3$ ,  $\text{CaCO}_3$ ,  $\text{PbO}$  and  $\text{CuO}$  to obtain a mixture; calcining the mixture at 750 to 900°C; grinding and then cold-pressing the calcined mixture; and sintering the pressed mixture at 750-900°C."

"3. A method for preparing an oxide superconductor including Bi, Sr, Ca, Cu, Pb and O having a  $T_c$  of greater than 77K and in which the molar ratio of Pb to Bi is in the range of 5:95-85:15 comprising the step of:

mixing appropriate amounts of powdered  $\text{Bi}_2\text{O}_3$ ,  $\text{Sr}_2\text{CO}_3$ ,  $\text{CaCO}_3$ ,  $\text{PbO}$  and  $\text{CuO}$  to obtain a mixture; calcining the mixture at 750 to 900°C; melting the calcined mixture at a temperature higher than its melting point to obtain a melted material; and annealing the melted material at a temperature below the melting point of the melted material."

"9. An oxide superconductor including Bi, Sr, Ca, Cu, Pb and O obtainable by a method according to any of Claims 1-8"

The independent claims 1 and 3 of the 3rd auxiliary request are identical to those and the 1st and 2nd auxiliary requests; however claims 9 to 13 have been deleted.

IV. At the appeal stage appellants 1 and 2 relied, *inter alia*, upon the following additional documents:

D31: Physical Review B, Vol. 35, No. 10, 1 April 1987, pages 5350-5352

D32: Europ. Workshop on High  $T_c$  Superconductors and potential applications, Programme, 1-3 July 1987, Genova, P-51, pages 377 to 378

D33: KfK Nachrichten, Jahrgang 19, 3/87, pages 119 to 129

D34: Science Vol. 238, Research News, 18 December 1987, pages 1655 to 1656

Opponent 3 withdrew its opposition by a letter dated 4 September 2002. Appellants 1 and 2 requested that the two witnesses offered by appellant 1 be heard in order to establish that the Sunshine et al. pre-print cited by appellant 2 had been made available to the public before the priority date.

V. In a first communication the board questioned the allowability of the amendments in the claims of the main request. In an annex to the summons to oral proceedings the board invited appellant 1 to provide

for the appearance at the oral proceedings of the two witnesses offered by them. Oral proceedings took place on 7 May 2003. Appellant 1 was accompanied by Prof. Dr H.E. Hoenig, one of the two witnesses they requested to be heard. At the beginning of the oral proceedings, the board decided that evidence should be taken from Prof. Dr Hoenig as a witness. Concerning the details of the witness testimony, reference is made to the minutes of the taking of evidence. These minutes are referred to hereinafter as D40.

VI. Appellants 1 and 2 requested that the decision under appeal be set aside and that the patent be revoked. As a main request, appellant 3 requested that the decision under appeal be set aside and that the patent be maintained with claims 1 to 15 of the "Main Petition" filed with appellant 3's letter dated 15 December 1998. As first, second and third auxiliary requests, appellant 3 requested maintenance of the patent on the basis of any of the first to third "Auxiliary Petitions" of the same letter. As the fourth and fifth auxiliary requests appellant 3 requested maintenance of the patent with the claims of any of the fourth and fifth auxiliary requests filed with the letter dated 14 March 2003.

VII. Appellants 1 and 2' arguments can be summarised as follows:

The Sunshine et al. pre-print was sent per fax by the Kernforschungszentrum Karlsruhe (hereinafter KfK) to Dr Hoenig (at that time an employee of appellant 1) on 23 February 1988, and a copy of this fax was transmitted per post to appellant 2. The original fax



received by Dr Hoenig was on thermopaper and could no longer be found. Appellant 2 could only have received the copy of the fax from appellant 1 since the copy D10 submitted by appellant 2 with their notice of opposition showed the same discontinuous page numbering as the copy D10b handed over by appellant 1 at the oral proceedings before the board and which was found in Prof. Hoenig's files remaining at Siemens after he had left this company. The missing pages 1 to 6 of the fax concerned other documents sent by KfK. The date stamp and fax number stamp imprinted on D10 showed that the fax was sent by KfK on 23 February 1988. According to his testimony Prof. Dr Hoenig remembered that he received the faxed pre-print directly from Dr Halbritter and transmitted a copy of the fax within the firm Siemens as well as outside according to a distribution list. In his testimony Prof. Hoenig had also confirmed that the distribution of pre-prints was a means for very rapidly informing an unlimited circle of persons about their contents and was a usual practice at that time. According to decision T 11/99 information was already made available to the public when only one person of the public had the possibility to gain knowledge of the information and to understand it and was not bound by a confidentiality obligation. This was the case for Prof. Dr Hoenig who further transmitted copies of the pre-print to universities and other firms according to an agreement between Dr Halbritter and Prof. Dr Hoenig concerning the distribution of pre-prints. The fax machine stamp confirmed in any case that Dr Halbritter gained knowledge of the pre-print at the latest on 23 February 1988, ie before the priority date. As he was also a member of the public and there was no evidence that a

confidentiality agreement existed between the co-author of the pre-print and Dr Halbritter, the pre-print had been made available to the public at this date. The co-author of the pre-print had no reason to maintain confidential the content of the pre-print since the latter was already with the publisher on 12 February 1988. Testimony by Dr Halbritter, the second witness initially offered by appellant 1, was not considered necessary since Prof. Dr Hoenig was able to testify to the same facts as Dr Halbritter.

Claim 1 of the main request did not meet the requirements of Article 123(2) EPC. Claim 1 of the first auxiliary request also contravened Article 123(2) EPC since the molar ratio Pb:Bi stated in this claim for the end product was not disclosed in the description as filed. The composition of the end product differed from that of the starting composition because of the partial evaporation of Bi and Pb during the heating step. Claim 12 as filed was not consistent with the description as filed. The product according to claim 9 of the first auxiliary request lacked novelty over the disclosure of D1. As the mixture was calcined and sintered at a temperature of about 850°C, no residue from the nitrate remained in the end product. Appellant 3's allegation that the end product contained a residue was contrary to the expectation of the skilled person, therefore the burden of proof lay with appellant 3 in this respect. Furthermore, the opposition division considered the product-by process claim as lacking novelty and thus appellant 3 had the burden of proving the contrary. D1 also destroyed the novelty of the process according to claim 1 of the first auxiliary request. D1 taught that the nitrate

used in the examples could be replaced by a carbonate and that Bi could be substituted with Pb. The process steps were the same whatever the kind of starting products. Furthermore the claimed amounts of Pb and the amounts stated in D1 overlapped.

The process according to claim 1 of the first auxiliary request further lacked an inventive step in view of the cited prior art. D7 published on 20 February 1988 was the closest prior art. The claimed process differed therefrom only by the substitution of part of the bismuth with lead. However lead was already known as a possible substituent in high- $T_c$  Y-Ba-Cu-O superconductors as shown by D32, which further disclosed that the substitution of rare earth with Pb led to an increase in the  $T_c$ . The partial substitution with Pb was also disclosed in D31 which concerned La-M-Cu-O (M= La, Ba, Sr, and Pb) superconductors. D31 further established a link between the classical Ba-Pb-Bi-O perovskite superconductors and the new materials La-M-Cu-O. The substitution with Pb in La-Sr-Cu-O superconductors was also confirmed by the teaching of D33 which also made reference to the low- $T_c$   $BaPb_{1-x}Bi_xO_3$  superconductors. The skilled person would therefore have considered using Pb as a partial substituent for Bi in the new Bi-Sr-Ca-Cu-O superconductors of D7. D4 showed that the Ba-Pb-Bi-O superconductors were prepared by sintering nitrates or carbonates of the components and all these different superconductors had the same structure, namely the perovskite structure.

According to another line of argument submitted by appellant 2, the chemical substitution of lead in many bismuth oxides was well-known to the skilled person as

stated in D10. D4 showed this substitution in  $BaPb_{1-x}Bi_xO_3$  superconductors. D32 further disclosed the partial substitution of yttrium with Pb in Y-Ba-Cu-O superconductors. Therefore, in view of the teaching in D4, D32 and common general knowledge, the skilled person would have tried to substitute Bi with Pb in the new Bi-Sr-Ca-Cu-O superconductors of D7. The skilled person would have explored all possible known substitutions in the recently discovered superconductors of the Bi-Sr-Ca-Cu-O system.

Appellant 1 objected that claim 1 contained a generalisation of the disclosure in the examples which in fact showed an improvement of  $T_c$  only for specific compositions, some of them not being disclosed in the priority document.

Concerning inventive step of the process of claim 3 of the first auxiliary request, appellants 1 and 2 argued that a melting step was already known from D8 and D34 for Y-Ba-Cu-O superconductors, and D34 taught that melting of the material improved the superconductive properties. D13 also mentioned a melting step for Bi-Sr-Ca-Cu-O materials. In view of this prior art it was obvious to use a melting step also in the case of Bi-Sr-Ca-Cu-O materials in which Bi was partially substituted with Pb. Appellant 2 submitted that claim 3 was unclear in the absence of any definition for the terms "melting point".

VIII. Appellant 3 presented essentially the following arguments:

Appellants 1 and 2 had failed to prove the public availability of the Sunshine et al. pre-print before the priority date. It was clear from Prof. Hoenig's testimony that he could not say when he received the fax from KfK. Appellants 1 and 2's allegation that Prof. Dr Hoenig was in possession of the pre-print on 23 February 1988 was not proved. Neither D10, D10a and D10b, nor the witness testimony evidenced that the pre-print was transmitted to members of the public before the priority date. It could be inferred from the circumstances of the case that a confidentiality agreement existed between Dr Halbritter and the co-author B. Batlogg of the pre-print. The European patent application D1 from the company AT&T corresponding to the content of the pre-print claimed the priority date of two US applications, the first one filed on 12 February 1988 and the second one on 26 February 1988. As this company had a commercial interest in this invention, it was in its interest not to transmit the content of the pre-print to the public at least until the filing of the second US patent application. Furthermore, the hand-written note from the said co-author on D10a also showed his personal relations with Dr Halbritter from which a confidentiality agreement could be derived. As appellant 1 withdrew their offer to hear the second witness Dr Halbritter, it was unknown what the co-author of the pre-print and Dr Halbritter had agreed on. The copy D10 of the fax was incomplete since the first six pages were missing and it was not known where and to whom the fax was sent. Information were also missing as to whom received the fax and when. It could have been received by someone down the chain. There was thus no reliable

evidence establishing beyond any doubt the date at which the pre-print was made available to the public.

Claim 9 of the 1st auxiliary request was novel over D1 as the carbonate starting materials would leave an inevitable fingerprint in the final superconductors which distinguished them from superconductors produced from nitrates. The calcination step did not completely eliminate the nitrogen or the carbon contained in the nitrate or the carbonate respectively. It was standard chemical knowledge that no reaction went to completion. A residue would remain in the calcined products which distinguished them. In case of doubt, the board had to decide in favour of the patent proprietor according to T 219/83. The process according to claim 1 of the first auxiliary request was novel with respect to D1 since starting from example 1 the skilled person had to make at least four alterations to arrive at the claimed subject-matter.

It was admitted that D7 was prior art. Starting therefrom the technical problem was to improve the  $T_c$  of the known Bi-Sr-Ca-Cu-O superconductor. The solution to this problem, namely the partial substitution of some content of Bi with Pb in the Bi-Sr-Ca-Cu-O-type superconductors in order to increase their  $T_c$  was not rendered obvious in view of D31, D32, D33 or D4. First of all the two types of known superconductors, ie Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O materials, had different crystal structures and their superconductivity arose from quite different crystallographic planes. Furthermore, D31 showed that the inclusion of lead into the superconductor removed the superconductivity. D32 also taught that the substitution with Pb reduced

the  $T_c$ . These two documents led away from the claimed invention. D33 taught in no way that the superconductivity was increased by substitution with Pb. Substitution of La or Cu with Pb led on the contrary to a reduction of  $T_c$ . The very general statement in D10 lacked any context. The superconductors of D4 were entirely different materials with a  $T_c$  of only 10K and had a different structure. Pb was an essential element to induce superconductivity contrary to the Bi-Sr-Ca-Cu-O superconductors of D7. D4 did not teach to add Pb in order to boost the superconductivity. There was no incentive to try the substitution with Pb since there was no expectation of success.

### **Reasons for the Decision**

1. The appeal is admissible
2. Public availability of the Sunshine et al. pre-print

Appellant 3 contested that it could be inferred from D10, D10a and D10b and from the witness testimony that Prof. Dr Hoenig, appellant 1, and/or appellant 2 received the pre-print before the priority date, ie before 24 February 1988.

- 2.1 D10 is a copy of a fax which bears the following data imprinted by the fax machine of KfK: "FEB 23 '88 11.55 KfK POSTST. FAX 07247/825070" on the title page of the pre-print. Appellant 1 did not cite D10 in their notice of opposition but argued in their letter dated 2 September 1997 that the co-author B. Batlogg of this

pre-print sent a copy thereof to Dr Halbritter in the "Forschungszentrum Karlsruhe" (KfK) on 12 February 1988 and signed this copy "with best regards Bertram B 2/12/1988" as shown by D10a, that Dr Halbritter then sent this pre-print to appellant 1 (to Prof. Dr H. E. Hoenig) by fax on 23 February 1988 while asking to further transmit the pre-print to interested persons, and that Prof. Dr Hoenig accordingly transmitted the pre-print among others to appellant 2. The question was raised by appellant 3 at the oral proceedings whether the date imprinted by the KfK fax machine on 23 February 1988 was the receipt date of a fax at KfK or the date at which KfK sent a fax to an addressee, ie a sender date. The witness testimony does not include any precise information in this respect. However the file contains a copy of another faxed pre-print, namely D7a which is a copy of the Maeda's pre-print sent by fax on 12 February 1988. In this case Dr Halbritter from KfK sent the pre-print on 12 February 1988 per fax to Prof. Dr Hoenig at Siemens using the same fax machine with the number 07247/825070. Identification of both the sender and the recipient is possible in view of the cover page of the fax (page 1 imprinted by the fax machine). In this case it is clear that the date imprinted by the KfK fax machine was the sender date. It cannot be completely excluded that this has not been changed between February 12, 1988 and February 23, 1988; however the board assumes in favour of appellants 1 and 2, that the date imprinted by the KfK fax machine in the case of the Sunshine et al. pre-print was also the date at which KfK sent the said pre-print.



From the page numbering imprinted by the KfK fax machine on the Sunshine et al. pre-print it can be seen that the title page of the pre-print is in fact **page 7 of the fax**. The fifteen pages of D10 were numbered pages 7 to 11 and 13 to 23 by the KfK fax machine. The copy D10a, which includes only the title page of the pre-print, also bears the same fax number, date, time, and page numbering (ie page 7) as the title page of D10. The copy D10b handed over by appellant 1 during the oral proceedings before the board contains the fifteen pages of the pre-print which have the same page numbering imprinted by the KfK fax machine, ie pages 7 to 11 and 13 to 23, and thus pages 1 to 6 of the fax are also missing. Although in the board's communication dated 14 May 2001 appellant 1 was asked to bring the original fax corresponding to D10a or D10 to the oral proceedings, this original document was not provided since, according to appellant 1 and to the witness Prof. Dr Hoenig, it could not be found (see minutes D40 page 4, first paragraph). The file contains no evidence from which it could be inferred **to whom the fax sent by KfK on 23 February 1988 was addressed**. The copies D10, D10a and D10b of the fax contain no copy of a possible cover page which might help to identify the addressee of the fax. They also contain no receipt stamp of the recipient fax machine. Although appellant 1 alleged that they received the fax, they could not provide the original fax and the copies D10, D10a or D10b do not prove beyond any reasonable doubt that they received it. This is because it cannot be excluded that Dr Halbritter sent the fax of 23 February 1988 to an addressee X other than Prof. Dr Hoenig, appellant 1 or appellant 2 and that this other addressee X then sent the incomplete copies D10 or D10b to Prof. Dr Hoenig,

appellant 1 or appellant 2 per post. In this case they would not have received the information before the priority date of 24 February 1988. In these circumstances, the addressee X would himself have received the information before the priority date and would have had the possibility to gain knowledge of it before the said date; however it would still remain to prove that there existed no confidentiality agreement between Dr Halbritter and the addressee X. Such a confidentiality agreement might have existed at least up to February 26, 1988 taking into account that two US patent applications concerning the same subject-matter as the pre-print have been filed by AT&T on 12 February 1988 and 26 February 1988 (see the two priority dates and documents indicated in D1). In this context it is observed that Prof. Dr Hoenig could not exclude having received the Sunshine et al. pre-print a second time from elsewhere (see page 4 of D40, last sentence).

- 2.2 Appellants 1 and 2 made reference to the declaration of Prof. Dr Hoenig that he himself received the Sunshine et al. pre-print directly from Dr Halbritter. According to the witness testimony, Prof. Dr Hoenig received the pre-print directly from Dr Halbritter and was able to recognise a fax coming from Dr Halbritter by a handwritten identification on it. However Prof. Dr Hoenig could not say **on which exact date** he received the pre-print (see Minutes D40 page 3, 4th paragraph; page 5, 4th and 6th paragraphs; page 6, 3rd paragraph). Therefore, it cannot be excluded that Prof. Dr Hoenig received a fax containing the Sunshine et al. pre-print from Dr Halbritter on a date later than February 23, 1988. In particular it cannot be inferred from Prof. Hoenig's testimony that the faxed pre-print

received by Prof. Dr Hoenig directly from Dr Halbritter was a fax bearing the date February 23, 1988.

Appellant 1 has not provided the original fax received by Prof. Dr Hoenig in order to support their contested allegation that Prof. Dr Hoenig actually received the faxed Sunshine et al. pre-print on 23 February 1988 although the burden of proof lies on appellant 1 or appellant 2 in this respect. For the preceding reasons and in particular in the absence of the required evidence the board is not convinced by the appellants' allegation that Prof. Dr Hoenig received the Sunshine et al. pre-print before the priority date.

2.3 Appellants 1 and 2 further argued that Dr Halbritter was in possession of the pre-print on 23 February 1988 and that the content thereof was publicly available since Dr Halbritter represented a member of the public and was not bound by a confidentiality agreement which would restrict the transmission of the pre-print to other members of the public. The board cannot follow these arguments for the following reasons. Assuming in favour of the appellants that the date imprinted by the KfK fax machine was the sender date, ie the date at which Dr Halbritter sent the Sunshine et al. pre-print to an addressee (see point 2.1 above), then Dr Halbritter had indeed the possibility to gain knowledge of the content of the pre-print before the priority date. However, it is not clear what the circumstances of the relationship between Dr Halbritter and B. Batlogg (co-author of the pre-print) and/or the company AT&T were. It is therefore also not clear that this relationship has not been governed by an obligation of Dr Halbritter to keep the information confidential at least until a certain point in time.

Appellant 3 argued at the oral proceedings that a confidentiality agreement could be derived from the fact that the company AT&T had a commercial interest in the invention and that it was in its interest not to disclose the content of the pre-print at least until the filing of the second US application on 26 February 1988. In the board's judgment it is plausible in view of the statement in D1 about the economic interest of the new materials (see column 2, lines 1 to 4) and of the priority dates of 12 February 1988 and 26 February 1988 stated in D1 that this company saw a commercial interest in the invention and was interested in filing US patent applications on the subject-matter of the pre-print. In these circumstances, it cannot be excluded that an obligation to keep the content of the pre-print confidential might have existed at least for a short period of time in February 1988 between the said co-author of the pre-print and Dr Halbritter. Although the burden of proof rested on appellants 1 and 2 as regards their allegation that there were no circumstances obliging Dr Halbritter to some kind of confidentiality, appellants 1 and 2 filed no evidence in this respect. Despite the invitation of the board to provide for the appearance of Dr Halbritter at the oral proceedings for a possible hearing as a witness who could possibly have enlightened the board as to the nature of his relationship with B. Batlogg and the firm AT&T, and in particular the circumstances of the transmission of the pre-print D10, appellants 1 and 2 deliberately chose not to present Dr Halbritter as a witness. The lack of sufficient proof resulting from the failure to do so goes to the detriment of appellants 1 and 2 who had the burden of proof. For the preceding reasons the board cannot consider on the

basis of the evidence submitted that it has been proven beyond any reasonable doubt that the Sunshine et al. pre-print was made available to the public before the priority date.

*Main request*

3. Claim 1 of this request does not meet the requirements of Article 123(2) EPC for the following reasons. According to amended claim 1 of the main request a mixture is prepared by mixing appropriate amounts of chemical compounds of Bi, Sr, Ca, Cu, Pb and O. However, the application as filed discloses mixing appropriate amounts of  $\text{Bi}_2\text{O}_3$ ,  $\text{SrCO}_3$ ,  $\text{CaCO}_3$ ,  $\text{PbO}$  and  $\text{CuO}$  as starting compounds to obtain the mixture to be calcined. It is not directly and unambiguously derivable from the application as filed that compounds other than the specific compounds indicated above might be used to obtain the desired superconductor having a  $T_c$  greater than  $77^\circ\text{K}$ . Furthermore, according to amended claim 1 a chemical composition is obtained in which the molar ratio of  $(\text{Bi}+\text{Pb}):\text{Sr}:\text{Ca}$  is 1:1:1 and the molar ratio of Pb to Bi is in the range of 5:95-85:15. Compositions having ratios of  $(\text{Bi}+\text{Pb}):\text{Sr}:\text{Ca}$  equal to 1:1:1 are indeed disclosed in the application as filed (see for example page 4a, page 6, lines 8 and 17; page 7, line 31; page 8, lines 18 and 29; page 9, line 11; claims 5 and 6) but always in combination with a specific amount of copper. It cannot be directly and unambiguously derived from the application as filed that the use of any amount of copper in combination with the molar ratio  $(\text{Bi}+\text{Pb}):\text{Sr}:\text{Ca}$  of 1:1:1 would lead to an oxide superconductor having a  $T_c$  greater than  $77\text{K}$ . As claim 1 of the main request contravenes the

provisions of Article 123(2) EPC, the main request must fail.

*First and second auxiliary requests*

4. The question arises whether the superconductor as defined in claim 9 of both requests, a product-by-process claim, is new with respect to the disclosure in D1. According to the established case law of the boards of appeal, claims for products defined as having been obtained by a particular process are allowable only if the product as such satisfies the requirements of patentability. In other words the product itself has to meet the requirements of novelty and inventive step. A product is not rendered novel merely by the fact that it is produced by a new process (see for example T 150/82, OJ EPO 1984, 309; T 248/85, OE EPO 1986, 261).

D1 is a document as defined in Articles 54(3) and (4) having two priority dates, namely 12 February 1988 and 26 February 1988. Only the information of D1 entitled to the priority date of 12 February 1988 forms part of the state of the art. D1 discloses superconductors manifesting a superconductivity at a temperature of at least 77K and having the nominal composition  $X_{2+y}M_{3-y}Cu_2O_{8+\delta}$  where X is Bi or Bi and one or more of Pb, In, Ga, Sn, Gd and Sb, M is selected from divalent ions and combinations of monovalent and trivalent ions selected such that the average valence of M is substantially equal to 2, and is typically 0 to 0.5. M is preferably selected from Ca, Sr, Ba, Mg, Cd, Na, K, Y or mixtures thereof. The superconductor has in particular the nominal composition  $X_{2.2}M_{2.8}Cu_2O_{8+\delta}$  where X is for example Pb and Bi, the Pb/Bi ratio being at most 0.5. An

exemplary crystal having the composition  $\text{Bi}_{2.2}\text{Sr}_2\text{Ca}_{0.8}\text{Cu}_2\text{O}_{8+\delta}$  ( $\delta$  typically 0-0.5) shows bulk superconductivity at a temperature of about 84K. D1 further teaches that substitutions of Bi with lead in the 84K phase have yielded considerable improvement in  $T_c$ , a multiphase lead substituted sample having a  $T_c$  of 107K. Powders of  $\text{Bi}_2\text{O}_3$ ,  $\text{Sr}(\text{NO}_3)_2$ ,  $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ , and  $\text{CuO}$  are used to prepare the composition of example 1. According to the general description of the process in column 8 of D1 appropriate starting materials are mixtures of metallic oxides, hydroxides, carbonates, hydrates, oxalates or other reactive precursors in the appropriate ratio to obtain the desired final composition (see column 2, lines 50 to 53; column 3, lines 13 to 19; column 7, lines 24 to 28; column 8, lines 4 to 7; example 1; claims 1 to 3). It was not disputed that this disclosure is entitled to the priority date of 12 February 1988.

The composition and the  $T_c$  of the superconductor materials disclosed in D1 fall within the broad definition indicated in claim 9 and also in claim 1 to which claim 9 makes reference. Appellant 3 argued that the product-by-process of claim 9 was novel over the disclosure in D1 since D1 used calcium nitrate and strontium nitrate as starting materials instead of calcium and strontium carbonates in the process of claim 1 and the calcination/sintering steps disclosed in D1 did not completely eliminate the nitrogen contained in the nitrates. The claimed products would thus contain a residue from the carbonates instead of a residue from the nitrates. This was disputed by appellants 1 and 2 who, from their side, argued that under the calcination and sintering conditions

disclosed in D1 the nitrates would not leave any nitrogen-containing residue. In the board's judgment, in view of the calcination and sintering conditions disclosed in example 1 of D1, namely 840°C for 12 hours and then 860°C for 5 hours, it seems to be unlikely that a nitrogen-containing residue from the Ca and Sr nitrates would remain in detectable amounts in the end products. However, taking into account that the starting mixture does not contain only one nitrate but a mixture of five components including nitrates and oxides, the board cannot completely exclude that a detectable residue might remain. Only experiments using a starting mixture comprising the two said nitrates instead of the two carbonates would in fact permit to conclude whether or not there is a difference in the end products. However the appellants have provided no evidence in this respect. Taking into account that the claims as granted did not contain any product-by-process claims, the latter having been introduced during the opposition proceedings, the board considers that the burden of proof lies on appellant 3 in respect to their allegation that a residue from the nitrate would remain in the end product. The board also observes in this respect that the question whether or not a residue from the nitrate would remain in detectable amounts after the sintering step was neither discussed, nor decided by the opposition division in the decision under appeal. Therefore the latter also contains no arguments or reasons which might shift the burden of proof to appellants 1 and 2. Under these circumstances, in particular in the absence of evidence from appellant 3 to support the contested allegation, the board does not accept this allegation and thus cannot consider the subject-matter of claim 9 of the



1st and 2nd auxiliary requests to be new with respect to the disclosure of D1. This is in line with decision T 219/83 (OJ EPO 1986, 211, point 12 of the reasons) according to which, if the EPO is unable to establish the facts, it is the party whose arguments rest on these alleged facts who loses thereby.

*Third auxiliary request*

5. Amended claims 1 to 8 of this request meet the requirements of Article 123(2) EPC. Claim 1 is based on a combination of claims 7 and 9 as filed with features disclosed in the application as filed. The  $T_c$  of greater than 77 K and the composition ratio of Pb to Bi in the range of 5:95-85:15 in the superconductor are disclosed on page 2, lines 5 to 15, and on page 3, line 35 to page 4 line 3, of the application as filed. The molar ratio of 5:95-85-15 in the superconductor is also directly derivable from claim 12 as filed. Appellant 1's allegation that original claim 12 is not consistent with the description as filed is not convincing in view of the disclosure in the passage on page 2, lines 14 to 15, whose content is also repeated in original claim 2.

Amended claim 3 is based on the combination of original claim 13 with the calcining temperature indicated in original claim 15. The molar ratio of Pb to Bi in the range 5:95-85:15 is directly derivable from original claims 18 and 2 and from page 2, lines 11 15, of the description as filed. The features of the dependent claims 2 and 4 to 7 are disclosed in original claims 8, 14, 15, 16 and 17. Those of claim 8 find support on page 5, lines 24 to 26, of the description as filed.

The scope of protection is clearly restricted by the amended claims compared to that of the granted claims.

6. Appellant 2 raised an objection of lack of clarity against claim 3 arguing that the "melting point" was not unambiguously defined taking into account that several phases with different melting points were present in the mixture. The board observes that the expression "melting point" was already stated in the granted claims and that lack of clarity pursuant to Article 84 EPC is not a ground of opposition. Thus, this expression has to be construed taking account of possible additional information in the patent in suit. The opposition division's construction indicated on page 6, third paragraph, of the decision under appeal was not contested by the parties and can also be followed by the board.
  
7. Appellants 1 and 2 argued that the process according to claim 1 lacked novelty over the disclosure of D1. The appellants' arguments put forward in this respect are not convincing for the following reasons. The method of preparation of the superconductor having the nominal composition  $\text{Bi}_{2.2}\text{Sr}_2\text{Ca}_{0.8}\text{Cu}_2\text{O}_8$  disclosed in example 1 of D1 is entitled to the priority date of 12 February 1988. It was not contested that the method of claim 1 differs from the method of example 1 in that the starting powder mixture contains PbO, Ca and Sr carbonates are used instead of Ca and Sr nitrates, and the ratio of Pb to Bi is in the range of 5:95-85:15. D1 further teaches that appropriate starting materials are mixtures of metallic oxides, hydroxides, carbonates, hydrates oxalates or other reactive precursors. It is also disclosed in D1 that Bi may be substituted with lead in

the 84K phase yielding considerable improvement in  $T_c$ . A Pb/Bi ratio of at most 0.5 is disclosed in D1, ie a range which partially overlaps with the claimed range (see column 8, lines 1 to 7; column 7, lines 24 to 25; claim 3). This disclosure is entitled to the priority date of 12 February 1988. To arrive at the claimed process the skilled person had therefore to select from the general list of starting materials calcium carbonate and strontium carbonate, none of them being expressly mentioned in this list, to replace in the process of example 1 only the strontium nitrate and the calcium nitrate by the corresponding carbonates, to select PbO as the starting product to be added to the mixture and to choose the appropriate Pb/Bi ratio. The specific combination of starting materials as defined in claim 1 is not disclosed in D1 and confers novelty on the claimed process.

The process of claim 1 is also novel with respect to the other documents cited by the parties and published before the priority date. Claim 3 also meets the novelty requirement with respect to the cited documents. This was not in dispute so that further considerations in this respect are not necessary.

8. Turning to the issue of inventive step, D1 does not form part of the state of the art. Appellant 3 accepted at the oral proceedings that the date of publication of D7 was 20 February 1988, ie before the priority date. D7 is considered to represent the closest prior art. It discloses a method for preparing a high- $T_c$  oxide superconductor of the Bi-Sr-Ca-Cu-O system such as the oxides  $\text{BiSrCaCu}_2\text{O}_x$  having a  $T_c$  of about 105K. Appropriate amounts of powders of  $\text{Bi}_2\text{O}_3$ ,  $\text{SrCO}_3$ ,  $\text{CaCO}_3$  and

CuO were mixed, calcined at 800 to 870°C for 5h, thoroughly reground and then cold-pressed into pellets which were sintered in air or an oxygen atmosphere at about 870°C (see page L209, abstract, left-hand column, 4th paragraph).

8.1 Starting from D7, the technical problem underlying the patent in suit can be seen in the provision of a process leading to superconductors having a higher critical temperature  $T_c$ . It is proposed to solve this problem by the process as defined in claim 1, which differs from the process of D1 in that the starting mixture further contains PbO. In view of the examples of the patent in suit, it is credible that the addition of PbO, in particular the partial substitution of Bi with Pb results in an increase of  $T_c$ . Appellant 1 objected that the improvement was shown only for some specific compositions and had been generalised to all compositions encompassed in claim 1. Assuming that appellant 1's objection was that the problem was not solved over the whole ambit of claim 1, then the burden of proof lies on them in respect of this allegation. However, appellant 1 provided no evidence to support their allegation. In the absence of evidence to the contrary, the board considers that the technical problem has actually been solved by the claimed process.

8.2 Appellants 1 and 2' arguments that it was obvious to the skilled person to partially substitute Bi with Pb in the process of D7 in view of the teaching in D32, D31, D33 and/or D4 in order to solve the technical problem (see point VII above) are not convincing for the reasons indicated hereinafter.

8.3 D32 concerns high- $T_c$  superconductors of the type  $YBa_2Cu_3O_7$  ( $T_c$  of about 90K). It deals with the question whether Y can be substituted with Pb in this system. According to D32 the motivation for studying the system  $Y_{1-x}Pb_xBa_2Cu_3O_7$  was that lead oxide is cheaper than yttrium oxide and that a previous attempt to substitute the rare earth with Pb had led to an increase of  $T_c$  and the most significantly to a remarkable increase of the upper critical field  $H_c$  (see page 377, lines 1 to 11). However the variation of  $T_c$  with  $x$  shown on Figure 3 (page 378) reveals that the highest value for  $T_c$  is obtained when the superconductor contains no Pb ( $x=0$ ). In other words the partial substitution of Y with Pb leads to a decrease of  $T_c$  in the case of the  $YBa_2Cu_3O_7$  superconductor. This result would lead the skilled person away from trying Pb as a substituent in the Bi-Sr-Ca-Cu-O system of D7 rather than encourage him to make such experiments.

Furthermore the detrimental effect of the partial substitution with Pb on the critical temperature is not only shown in D32 but also in D31. This document, which relates to superconducting phase transitions in the La-M-Cu-O layered perovskite system, where M=La, Ba, Sr, and Pb, discloses that the partial replacement of La by Pb in all three samples studied, ie  $La_{2-x}Pb_xCuO_4$  with  $x=0.1, 0.2$  and  $0.3$ , resulted in a complete disappearance of the superconductivity (see page 5351, right-hand column, first complete paragraph; page 5352, table 1 and right-hand column, first complete paragraph).

In view of D31 and D32 both showing the detrimental effect of the substitution of Y or La with Pb on the

critical temperature, the skilled person would have had no reason to expect that in the case of the Bi-Sr-Ca-Cu-O system according to D7 an increase of the critical temperature might be obtained by a substitution of Bi with Pb. In the absence of any reasonable expectation of success, he would not have tried such a substitution.

8.4 D33 discloses that substitutions had been performed in oxide superconductors such as  $\text{Li}_{1+x}\text{Ti}_{2-x}\text{O}_3$  and  $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$  in order to increase their critical temperature, however they remained unsuccessful (see page 120, right-hand column, lines 1 to 17 from the bottom; page 121, paragraph bridging the right-hand column and the middle column). Concerning the superconductors in the La-Ba-Cu-O or La-Sr-Cu-O systems having higher  $T_c$  of  $> 30\text{K}$  to about  $40\text{K}$ , D33 discloses different substitutions, among others substitutions with Pb (see in particular page 124, Table 1). However it can be inferred from Table 1 that the substitution of Cu with Pb leads to a decrease of the critical temperature (compare sample Y3 with sample X24.04) and the substitution of La with Pb, and Cu with Bi also resulted in a lower  $T_c$  (compare samples L44 and L24). Therefore, substitutions with Pb in these La-Ba(Sr)-Cu-O systems having  $T_c$  of about  $40\text{K}$  do not improve the  $T_c$  (see also page 124, the sentence bridging the left-hand and middle columns). It follows therefrom that D33 contains no additional information which would give the skilled person an incentive to partially replace Bi by Pb in the material of D7 in order to increase the  $T_c$  of the superconductors.

8.5 D4 concerns superconductors of the composition  $BaPb_{1-x}Bi_xO_3$ . Although these superconductors have perovskite related structures, they belong to a different type of superconductors not containing any copper, having a different crystal structure and a superconductivity which arises from different crystallographic planes compared to the superconductors of D7. The combination of both Pb and Bi results in superconductivity properties, superconductivity having been observed from  $x=0.05-0.3$ . The addition of Pb to the  $BaBiO_3$  material which has a semi-conductive behaviour leads to a material having superconductive properties with a maximal critical temperature of 13K at  $x=0.3$ ,  $T_c$  then decreasing to 9K when the amount of Pb is further increased (see page 27, abstract and page 28, left-hand column, 1st paragraph). These superconductors have relatively low  $T_c$  of at most 13K compared to the much higher values of 90K in the Y-Ba-Cu-O system (D32) or 105K in the Bi-Sr-Ca-Cu-O system of D7. It cannot be inferred from this teaching that the substitution of Bi with Pb in the superconductors of the Bi-Sr-Ca-Cu-O system which, contrary to  $BaBiO_3$ , already exhibit superconductivity properties) and a very high  $T_c$  of 105K might result in a **further increase** of the critical temperature all the more so as substitutions with Pb in  $YBa_2Cu_3O_7$  or in  $La_{2-x}Sr_xCuO_4$  superconductors having  $T_c$  values of about 90K or about 40K respectively lead to a decrease of  $T_c$  as shown in D32 and D33. Therefore, in the absence of any reasonable expectation of success in view of the cited prior art the skilled person would not have tried substituting Bi with Pb in the superconductors of D7 in order to solve the problem stated above. The fact that chemical substitution of lead might have been well-

known in many bismuth oxides as pointed out by appellant 2 cannot in any way change the preceding conclusions in the absence of further information as to which bismuth oxides are meant in D10 (see D10, handwritten page 7, first line), whether or not they exhibit superconductivity properties and what the effect of the substitution on the material properties was. The bismuth oxides meant in D10 might be those of D4.

In the board's judgement, the fact that the high- $T_c$  superconductor of D7 was a new kind of superconductor whose substitution possibilities had not yet been explored, does not justify fully ignoring the concept of reasonable expectation of success developed in the case law. This concept is also applied in the technical field of genetic engineering where some areas have not yet been explored. For example in decision T 42/92 (OJ EPO 1997, 408, point 8.5 and ff.) where an experiment was considered to be "obvious to try" for the skilled person, it has been examined whether or not the skilled person was in a position to reasonably predict the successful conclusion of the experiment, on the basis of the existing knowledge, before starting the experiment. In the present case the skilled person was not in a position to reasonably predict that substitutions with Pb in the new superconductors of D7 would bring about an increase of  $T_c$  for the reasons given above, in particular in view of the known detrimental effect of the said substitutions in superconductors already having a relatively high  $T_c$  of for example 40K or 90K.



8.6 The considerations put forward above apply likewise to the subject-matter of claim 3. The additional documents D8, D13, and D34 were cited by appellants 1 and 2 in connection with the issue of inventive step to show that the additional melting step stated in claim 3 was known for the preparation of superconductors in the rare earth-Ba-Cu-O system (D8) and in the Y-Ba-Cu-O system (D34), or could be inferred from D13 for superconductors in the Bi-Sr-Ca-Cu-O system. They concluded that it was obvious to perform a melting step also in the case of the new Bi-Sr-Ca-Cu-O superconductors. The three cited documents indeed disclose a melting step, however they contain no information suggesting that the substitutions of any elements of the considered superconductors with Pb would increase their critical temperature. Therefore, even if the melting step were obvious to the skilled person, the subject-matter of claim 3 would still be considered to involve an inventive step for the reasons given above in connection with claim 1.

8.7 The remaining documents published before the priority date were not relied upon at the oral proceedings. These documents contain no additional information which would point towards the claimed solution as defined in claims 1 and 3.

8.8 It follows from the above that the subject-matter of independent claims 1 and 3 of the third auxiliary request meets the requirement of inventive step set out in Articles 52(1) and 56.

9. Claims 1 and 3 being allowable, the same applies to dependent claims 2 and 4 to 8, whose patentability is supported by that of claim 1.

**Order**

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

1. The decision under appeal is set aside.
2. The case is remitted to the opposition division with the order to maintain the patent with the claims of the third auxiliary request filed with the letter dated 15 December 1998, ie with claims 1 to 8 of the request filed with the letter dated 22 May 1998, and a description to be adapted.

The Registrar:



U. Bultmann

The Chairman:



R. Spangenberg

*Gu*  
*MCS*