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D E C I S I O N
of 4 April 2001

Case Number: T 0911/97 - 3.4.2

Application Number: 93307368.6

Publication Number: 0593167

IPC: H01G 4/008

Language of the proceedings: EN

Title of invention:

Internal electrode for multilayer ceramic capacitor

Patentee:

SHOEI CHEMICAL INC.

Opponent:

Degussa Ag

Headword:

-

Relevant legal provisions:

EPC Art. 123, 54, 56

Keyword:

"Claim 1 (main request) - added subject-matter (yes)"

"Claim 1 (first and second auxiliary request) - novelty (yes)"

"Claim 1 (first and second auxiliary request) - inventive step (no)"

Decisions cited:

-

Catchword:

-



Case Number: T 0911/97 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 4 April 2001

Appellant: SHOEI CHEMICAL INC.
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Decision under appeal: **Decision of the Opposition Division of the
European Patent Office posted 11 July 1997
revoking European patent No. 0 593 167 pursuant
to Article 102(1) EPC.**

Composition of the Board:

Chairman: E. Turrini
Members: M. A. Rayner
B. J. Schachenmann

Summary of Facts and Submissions

I. The appellant (=patent proprietor) has appealed against the decision of the opposition division revoking European patent number 593 167 (application number 93 307 368.6). In the proceedings before the opposition division, reference was made, amongst others, to the following documents:

D1: JP-B-63/31522 (English Translation)

D2: J. Material Science 26 (1991) pages 2477 to 2482

D3: US-A-3 815 187

D8: Declaration of inventor, also the author of the Article in document D2.

As an annex to the statement setting out the grounds of appeal the appellant filed document

D10: Condenser Review, Vol. 44, No.1, page 26, 1991
(together with an English translation of a relevant passage).

The opposition division considered a distinguishing feature of the subject matter of claim 1 to be the use of the paste known from document D1 with a particular capacitor, namely a multilayer capacitor. The claim also specified essential steps for manufacturing the same. Multilayer capacitors need a good printable film and the skilled person would recognise without being inventive that the thick film of document D1 must be useful therein. The remaining process related

features are well known in the art of manufacturing multilayer capacitors. Therefore, the subject matter claimed is obvious. It is of no further relevance, if an additional positive bonus effect such as the reduction of cracking occurs.

II. According to the appellant, the opposition division entirely ignored the problem underlying the invention and the solution. The appellant emphasised that the subject invention is concerned with a specific type of capacitor, namely a co-fired multilayered ceramic capacitor. Delamination or cracking is a long standing problem that hitherto had no solution. There is no technical teaching in document D1 relating to co-firing of a plurality of green ceramic sheets and the above mentioned problem. Furthermore, it is not readily apparent why a skilled person would be motivated to select a single crystalline powder for use in the co-fired multilayered laminated ceramic capacitor, which is not even discussed in document D1. Documents D3 and D10 make clear that the skilled person expects volume expansion to occur with a thick film paste and thus that the teaching of document D1 is not applicable to multilayered capacitors. Document D8 explains that the chemical flame method produces polycrystalline particles of Pd and accordingly the palladium powder disclosed in document D2 is a polycrystalline metallic powder exhibiting different behaviour. The powder is thus more susceptible to oxidation than single crystal powder. Therefore, document D2 is not relevant to the subject invention.

III. According to the respondent, the claim 1 in any form does not distinguish patentably from the prior art and does not reflect problems correlated with the possible

oxidation of the metal particles and associated with the manufacture of multilayer capacitors when co-firing ceramic green sheets with metallic paste printed thereon. The feature defining the metal powder as a substantially single crystal metallic powder is disclosed in or at least obvious from documents D1, D2 and D3.

- IV. The appellant requested setting aside of the decision of the opposition division and maintenance of the patent. The respondent (=opponent) requested the board to dismiss the appeal of the appellant. Oral proceedings were requested by both parties on an auxiliary basis.
- V. Oral proceedings were appointed, consequent to the auxiliary requests of the parties. In the communication attached to the summons to oral proceedings, the board expressed doubts about the admissibility in the sense of Article 123(2) EPC of a feature relating to an alloy of Pd and Ag.
- VI. During the oral proceedings, the appellant requested maintenance of the patent on the basis of a main or alternatively a first or second auxiliary request filed during the oral proceedings. The appellant argued that claim 1 according to the main request was supported by the documents as filed. In particular, while admitting that only a specific example of Pd and a specific example of Ni were shown in the description, the selection of the alloy of Pd and Ag was nevertheless supported by the paragraph: "The metallic powder to be employed as a conductor in the present invention includes Pd, Ag, Ni and Cu which have heretofore been used as the conductive components of the internal

electrodes for capacitors. The metallic powder as mentioned in the present invention is meant to include single metal powder, alloy powder and mixtures thereof." Moreover, original claim 3 recites "a mixture of at least two metal salts and the conductor is one formed from an alloy". Since the possible selections that can be made are not that great, a selection of Pd/Ag does not add subject matter.

Oxidation of Pd powder becomes a problem during co-firing of green sheets and that this is alleviated by single crystal powder cannot be derived from the citations. In particular, document D1 does not address delamination nor multilayer capacitors and refers to the desirability of thick film paste, for which document D10 (or D3) teaches agglomerated particles (partially oxidised) should be used to avoid expansion of Pd. An indication of good crystallinity does not mean single crystal and in any case there is no application to a multilayer capacitor. The upper line in Figure 8 of document D2 confirms the polycrystalline Pd powder suffers from volume expansion powder.

VI. The respondent maintained his request for dismissal of the appeal and argued during the oral proceedings that the whole of the manufacturing of multilayered capacitors could be seen in document D3, which also disclosed the underlying problem of oxidation of Pd, its cracking and a solution thereto by preoxidation. A method of producing substantially single crystal powder is shown in document D1, where a furnace is used just as in the patent in dispute and which also suggests application of the teaching to capacitors. Document D2 discloses spray pyrolysis and mentions multilayer

capacitors and also that the metal powder is well crystallised. Section 3.3 explains that oxidation-reduction of palladium is important in the manufacture of multilayer capacitors and also that the extent of palladium oxidation of powders produced by spray pyrolysis is much lower than those produced by chemical precipitation. The respondent also drew attention to the last sentence of document D2 indicating that although an actual application to electronic devices was not carried out in the study, it (spray pyrolysis) will surely exhibit excellent capabilities in major cases. So far as the powders formed are concerned, there is no difference claimed between heating in a furnace and a chemical flame and anyway both are disclosed in the prior art. The subject matter of claim 1 according to all the requests must therefore be considered obvious from a combination of these references.

- VII. Claim 1 according to the main, first and second auxiliary requests of the appellant is worded as follows:

Main Request

A multilayered ceramic capacitor having internal electrodes comprising substantially single-crystal metallic powder of Pd, or of Ni, or of an alloy of Pd and Ag as a conductor, said powder having been produced by spraying a solution containing a metal salt of Pd, or metal salts of Pd and Ag, or a metal salt of Ni to form droplets and heating said droplets to a temperature higher than the decomposition temperature of said metal salt or salts and also higher than the melting point of said Pd, alloy of Pd and Ag or Ni said

capacitor being prepared by printing a paste including said powder onto plural ceramic green sheets, laminating the green sheets and firing the laminated green sheets.

First Auxiliary Request

A multilayered ceramic capacitor having internal electrodes comprising substantially single-crystal metallic powder as a conductor, said powder having been produced by spraying a solution containing at least one metal salt to form droplets and heating said droplets to a temperature higher than the decomposition temperature of said metal salt and also higher than the melting point of said metal, said capacitor being prepared by printing a paste including said powder onto plural ceramic green sheets, laminating the green sheets and firing the laminated green sheets.

Second Auxiliary Request

A multilayered ceramic capacitor having internal electrodes comprising substantially single-crystal metallic powder of Pd, or of Ni, as a conductor, said powder having been produced by spraying a solution containing a metal salt of Pd or of Ni to form droplets and heating said droplets to a temperature higher than the decomposition temperature of said metal salt and also higher than the melting point of said Pd or Ni said capacitor being prepared by printing a paste including said powder onto plural ceramic green sheets, laminating the green sheets and firing the laminated green sheets.

VIII. At the end of the oral proceedings, the board gave its

decision.

Reasons for the Decision

1. The appeal complies with the provisions mentioned in Rule 65(1) EPC and is therefore admissible.

2. *Article 123(2) EPC - Main Request*
 - 2.1 According to the submissions of the appellant, while the selection of an alloy of Pd and Ag is not specifically disclosed in the application documents as filed, it is nevertheless derivable from the passages cited in support, since only a limited choice is available according thereto. However, the board observes that in these passages and elsewhere in the documents as filed, an alloy is mentioned in the context of the metals Pd, Ag, Ni and Cu, the only further elucidation being a reference to "at least two" alloying metals. Accordingly, two steps are needed for the skilled person starting from this disclosure to reach the Pd/Ag claimed, namely firstly to decide how many metals should be in the alloy and secondly to choose the metals concerned. This procedure has to be carried out without any guidance in the specification and despite the specific description only containing examples of single metals, one of Pd alone and one of Ni alone. In the view of the board, this two step procedure involving a component not even in the specific examples requires further guidance not present in the documents as filed and thus amounts to more than making the limited choice argued by the appellant, whose argument in support of admissibility of the amendment fails.

Accordingly, claim 1 of the main request contains subject matter which does not satisfy Article 123(2) EPC.

3. *Novelty*

First Auxiliary Request

- 3.1 Document D1 discloses a method of manufacturing metal powder, which is useful for application in thick film paste finding application in the area of electronics for parts such as resistors, capacitors and IC packages. The paste is applied on the substrate and fired at high temperature to form conductor or resistor coat. Good crystallinity with uniform orientation is desired (see item 3 on page 2). The method entails spraying a solution of one or more metal salts, the droplets being heated to a temperature higher than the decomposition temperature of the metal salt and higher than the melting point of the metal. The metals concerned can include palladium or nickel. Alloys can also be formed of two or more metal salts (see the paragraph bridging page 2 and 3 and the paragraph thereafter). The first example disclosed involves dissolving silver nitrate and heating in an electric furnace. The resulting powder has good crystallinity. In the case of example 2, a similar procedure was followed starting with a mixture of silver nitrate and palladium nitrate. The powder obtained was Ag/Pd alloy of good crystallinity.

The subject matter of claim 1 according to the first auxiliary request differs from the disclosure of document D1 by virtue of the features relating to the capacitor being prepared by printing a paste including

said powder onto plural ceramic green sheets, laminating the green sheets and firing the laminated green sheets.

- 3.2 Document D2 recognises that the oxidation-reduction of palladium is important (see the right hand column of page 2480) in the manufacture of multilayer ceramic capacitors because electrode expansion occurs during oxidation and gas evolution during reduction. Also noted is that the extent of palladium oxidation of silver-palladium alloy and pure palladium powders prepared by the spray pyrolysis technique is much lower than powders prepared by chemical precipitation. This is stated to be mainly because a metal prepared by the spray pyrolysis technique has a large crystallite size or low specific surface area, whereas in chemical precipitation it is difficult to prepare such particles (see the right hand column of page 2481). The spray pyrolysis is effected using the chemical flame method.

There is no specific reference in document D2 to substantially single-crystal powder, nor is there a reference to the green sheet firing step.

- 3.3 Document D3 relates to a method for making multilayered ceramic capacitors wherein an ink containing palladium oxide is used in the printing of electrode patterns on green ceramic sheet which is subsequently stacked and sintered. Multilayer ceramic capacitors made according to the teaching of document D3 are said not to be subject to delamination during the firing step which often occurs with the use of metallic palladium containing inks due to oxidation of the metal with concomitant volume increase. Preoxidation of the

palladium electrode material is said to avoid expansion of the electrode material in situ (see column 2, lines 50 et seq.).

Document D10 discloses that a problem to be considered is the oxidation of Pd powder, oxidation and expansion beginning at about 500°C which reaches 100% and thereafter reduces to return to the original Pd at about 900°C.

- 3.4 The subject matter of claim 1 according to the first auxiliary request differs from the disclosure of document D3 by virtue of the features relating to said powder having been produced by spraying a solution containing at least one metal salt to form droplets and heating said droplets to a temperature higher than the decomposition temperature of said metal salt and also higher than the melting point of said metal. A single-crystal powder is also not disclosed in document D3. A similar situation exists with respect to document D10, where no explicit detail of the firing process is given at least in the translated part.

The subject matter of claim 1 according to the first auxiliary request is therefore novel within the meaning of Article 54 EPC.

- 3.5 Second Auxiliary Request

The same features are novel in the case of the second auxiliary request in relation to the alternative Pd, the alternative of "nickel" only being mentioned in document D1.

4. *Inventive step*

First Auxiliary Request

4.1 From the submissions of the parties, the board has formed the view that it is common ground that the underlying problem addressed by the patent in dispute relates to cracking and delamination of multilayer ceramic capacitors. Having reviewed the available prior art, the board thus considers documents D2 and D3 to be close prior art because they, like the patent in dispute, deal with the problem of cracking and delamination caused by oxidation of palladium in relation to layers incorporating palladium as electrode material in multilayer ceramic capacitors. The problem solved by the features of claim 1 which are novel with respect to document D3 and relate to the spray pyrolysis technique and the substantially single-crystal metallic powder can therefore be seen as that of providing a solution to the problem of delamination and cracking other than that of preoxidation.

Document D2 offers another solution. In fact, although the structure of a multilayer ceramic capacitor is not given in document D2 and an actual application to electronic devices not carried out therein, the board considers its teaching to be even closer to the patent in dispute than document D3 because it explains that powder formed by spray pyrolysis reduces expansion of Pd, realising that this is important in the context of multilayer ceramic capacitors. It is expected according to document D2 that the spray pyrolysis technique will exhibit excellent capabilities. Despite no application to electronic devices being made, the scene therefor is set and the skilled person thus understands that spray pyrolysis is expected to be applied in the field of multilayered ceramic capacitors, which are explicitly

mentioned as important in relation to reduced expansion. Since document D2 is not a patent document, but a technical publication, it does not spell out the structure of a multilayer ceramic capacitor because it is assumed that the reader is fully familiar with this. Consistent with the submissions of the appellant, the expansion problem is however well known and thus even if the board were to take a generous approach towards the case of the appellant by considering the structure of multilayer ceramic capacitors and their firing not to be implicitly known from document D2, there could be no question in the light of the reference to multilayer ceramic capacitors of an inventive step being involved in finding the corresponding teaching in document D3.

- 4.3 The board considers the decisive disclosure of document D2 to occur at the end of the first paragraph of the right hand column, where it is recited that "...the extent of palladium oxidation of the silver-palladium alloy and pure palladium powders prepared by the spray pyrolysis technique is much lower than powders prepared by chemical precipitation. This is mainly because a metal particle prepared by the spray pyrolysis technique has a large crystallite size or low specific area, and low activity attributed to the fusion during precipitation". The skilled person knows that the largest size and lowest area points strongly towards a single-crystal and thus the teaching of document D2 gives such a clear hint towards a single-crystal powder that its provision is obvious.

There have been no arguments on the basis that document D2 does not teach spray pyrolysis or indeed that it does not mention multilayered ceramic capacitors and indeed arguments in this direction would not be

credible. Thus, the main line of reasoning of the appellant departs from the analysis of the board only in the last step, by submitting that not a substantially single-crystal powder but an unsuitable polycrystalline powder is produced according to the teaching of document D2 as interpreted by the declaration of document D8.

- 4.4 If this line of reasoning is pursued, the difference between the subject matter of claim 1 and the disclosure of document D2, apart from the structure of the multilayer ceramic capacitor dealt with above, is the claiming of a substantially single-crystal powder. The problem to be solved in relation to document D2 is therefore to provide an even better crystallinity powder and reduce expansion even further than shown in Figure 8 of document D2, which was already expected to be successful. Given the importance attached to crystallinity, the board is convinced that even without the "input" of document D8, the skilled person would have sought an improvement in this respect. Just such an improvement is offered according to the teaching of document D1, which refers explicitly to the desirability of a powder of good or very good crystallinity for thick film pastes (see item 3 on page 2, or the middle of pages 3, 4 or 5) and which also teaches production of powder by spray pyrolysis. The board therefore considers a combination of the teachings of these documents to have been an obvious course for the skilled person. The appellant sought to defeat this argument by submitting that good crystallinity and substantially single-crystal do not have the same meaning. Rather than becoming lost in a question of semantics, the board considers that the issue can best be resolved on a technical basis by

consideration of document D8 which was relied on by the appellant in analysing the content of document D2. According to document D8, the reason why the powders obtained according to document D2 are not single-crystalline is that it is difficult to stably and evenly heat the reaction zone by a chemical flame method, whereas an electric furnace as used in the patent in dispute makes it easy to precisely set and control the heating conditions. The board takes this to mean that the "substantially single-crystal" feature claimed implicitly means use of an electric furnace in the spray pyrolysis. This is just what is taught in document D1, an electric furnace (itself commonly used equipment and shown as 3 in the figure) being used for the spray pyrolysis. Therefore the implicitly meant feature of the patent in dispute is met by document D1. Accordingly, technical considerations reveal that in the present context "good crystallinity" and "substantially single-crystal" do indeed have the same technical meaning. Quite apart from this technical consideration, lines 40 to 41 of the patent (and the corresponding portion of the documents as filed) recite that "single-crystal powder to be used in the present invention is described in Japanese Patent Publication 31522/1988 (i.e. document D1)". The board is therefore convinced that the desideratum of a large crystallite size according to document D2 would in consideration of the good crystallinity according to document D1 have lead the skilled person to the subject matter of claim 1 without involving any inventive step.

- 4.5 When starting from document D1 as closest prior art, the appellant advanced another line of argument that documents D3 and D10 constitute a prejudice against Pd powders for multilayer ceramic capacitors. This hurdle

is however taken outside the framework of document D1 when starting from document D2 according to the above reasoning of the board because multilayer ceramic capacitors are mentioned in document D2 which indicates that the skilled person would expect a successful application of spray pyrolysis.

Second Auxiliary Request

- 4.6 Since a Pd powder, which is specified in the second auxiliary request, was associated with the lamination and cracking problem according to the teaching of document D2, the arguments in relation to inventive step apply correspondingly to the subject matter of claim 1 of the second auxiliary request.
5. Therefore, claim 1 of the main request is inadmissible because it contains subject matter infringing Article 123(2) EPC. The subject matter of the claims 1 according to the first and second auxiliary request are not considered to involve an inventive step within the meaning of Article 56 EPC.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

P. Martorana

E. Turrini