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**Datasheet for the decision
of 13 February 2008**

Case Number: T 0210/06 - 3.2.02

Application Number: 00957081.3

Publication Number: 1255872

IPC: C22C 1/04

Language of the proceedings: EN

Title of invention:

Production of nitrogen-containing tantalum or niobium powder
and solid electrolytic capacitor

Patentee:

Cabot Super Metal K.K.

Opponent:

H.C. Starck GmbH

Headword:

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Relevant legal provisions:

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Relevant legal provisions (EPC 1973):

EPC Art. 100(a), 115, 54(1), 56

Keyword:

"Novelty (no)"
"Inventive step (no)"

Decisions cited:

-

Catchword:

-



Case Number: T 0210/06 - 3.2.02

D E C I S I O N
of the Technical Board of Appeal 3.2.02
of 13 February 2008

Appellant:
(Opponent)

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Respondent:
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Decision under appeal:

Interlocutory decision of the Opposition
Division of the European Patent Office posted
8 December 2005 concerning maintenance of
European patent No. 1255872 in amended form.

Composition of the Board:

Chairman: T. Kriner
Members: R. Ries
A. Pignatelli

Summary of Facts and Submissions

- I. Opposition was filed against European patent No. 1 255 872 as a whole by the present appellant and was based on Article 100(a) EPC (1973) (lack of novelty and lack of inventive step).

In its interlocutory decision dispatched on 8 December 2005, the opposition division held that the subject matter of the claims according to the first auxiliary request then on file met the requirements of the EPC and maintained the patent in amended form.

- II. On 8 February 2006 the opponent (appellant) lodged an appeal against this decision and paid the appeal fee at the same date. The statement setting out the grounds of appeal was received on 13 April 2006.

In support of his submissions, the appellant essentially referred to the following documents:

D2: WO-A1-98/37249

D8: US-A-3 825 802

D9: US-A-5 442 978

- III. Observations of a third party under Article 115 EPC (1973) were submitted on 18 October 2007 referring to

D10: WO-A1-98/37248.

This document was considered as relevant prior art with particular respect to the novelty and inventive step of

the method claims 2 to 8 according to the set of claims upheld by the opposition division.

IV. At the end of the oral proceedings held before the Board on 13 February 2008, the following requests forming the basis of the present decision were made:

The appellant (opponent) requested that the decision under appeal be set aside and the European patent No. 1 255 872 be revoked.

The respondent (patent proprietor) requested that the European patent be maintained

- on the basis of claims 1 to 14 filed on 3 January 2007 as Main Request or, alternatively,
- on the basis of the claims filed as first to fourth auxiliary requests on 3 January 2007, or
- as fifth to twelfth auxiliary requests filed on 8 January 2008, or
- as thirteenth or fourteenth auxiliary requests filed during the oral proceedings.

The independent claim 2 of the main request reads as follows:

"A method for producing nitrogen-containing metallic powder, characterized in that while a tantalum compound is reduced with a reducing agent, a nitrogen-containing gas is introduced into a reaction system to thereby form tantalum, and nitrogen is simultaneously incorporated into the tantalum."

Method claim 2 of the main request is also present in the set of claims according to the first and second

auxiliary requests and as claim 1 in the third and fourth auxiliary requests.

Independent claim 2 according to the fifth and seventh and claim 1 of the ninth, and eleventh auxiliary request read (the additional feature to claim 1 of the main request in bold):

"A method for producing nitrogen-containing metallic powder, characterized in that while a tantalum compound is reduced with a reducing agent **in a liquid phase system**, a nitrogen-containing gas is introduced into a reaction system to thereby form tantalum, and nitrogen is simultaneously incorporated into the tantalum."

Independent method claim 2 according to the sixth and eighth as well as claim 1 of the tenth and twelfth auxiliary requests read (the additional features to claim 1 of the main request in bold) :

"A method for producing nitrogen-containing metallic powder characterized in that while a **halide of tantalum** is reduced with a reducing agent **in a molten salt at 700 to 1000°C**, a nitrogen-containing gas is introduced into the molten salt to thereby form tantalum, and nitrogen is simultaneously incorporated into the tantalum."

In addition to this claim, independent claim 1 of the thirteenth and fourteenth requests further comprises the wording (in bold):

"A method for ... and nitrogen is simultaneously incorporated into the tantalum, **wherein the nitrogen-containing powder is a solid solution comprising 50-20,000 ppm nitrogen.**"

V. The appellant's arguments can be summarized as follows:

A method for producing fine Ta-powder of the type claimed in the patent at issue was disclosed in document D10. The amount of nitrogen incorporated in the Ta-powder ranged from 100 to 15000 ppm, with at least 500 ppm N being preferred (see D10, page 5, penultimate paragraph). As specifically emphasised in D10, example 4, the Ta-powder was nitrided during the reduction and, therefore, the technical disclosure of this document anticipated the process set out in the claims of the main request and first to fourth auxiliary requests.

Moreover, the method in document D9 provided that the doping of Ta-powder with nitrogen (and/or with P or Si) could be performed during the original reduction step, in particular during the reduction of K_2TaF_7 comprised in a molten salt mass at about 975° by Na. This method was also used in the patent at issue. Doping the Ta-powder with P, S or N during the reduction step was, therefore, well known to the skilled person.

The claimed process and the one known from the prior art being identical, the product obtained by such a process must be expected to exhibit the same physical and chemical properties. In case of Ta-powder doped with nitrogen, it must be postulated that in both processes a body-centered cubic structure was formed

having nitrogen interstitially incorporated as solid solution therewith, as it was pointed out for instance in document D8, column 3, lines 1 to 4.

VI. The respondent's arguments can be summarized as follows:

In the process described in D10, example 4, the technical term "Nitridierung" showed that the nitrogen was incorporated in the reduced Ta-powder in the form a "tantalum nitride" rather than as a interstitial solid solution, as was brought about by the claimed method. The process set out in claim 2 of the main, first and second auxiliary requests and in claim 1 of the third and fourth auxiliary requests was therefore novel over D10.

Document D9 actually disclosed in column 3 the reduction of K_2TaF_7 in a molten salt at $975^\circ C$. However, this document merely described, in a very general manner, the possible modification of the primary or secondary Ta-powders with phosphorus or silicon or nitrogen. The modification could be carried out either at primary or secondary stages of the process. Moreover, the document pointed out that the dopant additive(s) could be supplied either in elemental form (P, Si, or N) or as a compound source thereof, and if the additive(s) were provided during reduction, oxidising conditions in the reactor should be strictly avoided. In order to arrive at the process claimed in the patent at issue the skilled person, putting into practice the teaching of document D9, had to make several choices. Specifically, he had to select (a) nitrogen or (b) a nitrogen bearing compound and (c) to provide it during the reduction under (d) non-oxidising conditions.

Adding nitrogen gas (N_2) to the molten salt bath would mean creating oxidising conditions in the chemical understanding of the term "oxidation" since the oxidant (N^0) was reduced to (N^{3-}) in the salt bath. Hence document D9 dissuaded from adding nitrogen gas to the salt bath during the reduction step.

Moreover, neither D9 nor any other document taught or suggested that nitrogen, when added during the reduction step, could be uniformly incorporated as a solid solution in the Ta-powder. In the absence of such a technical teaching, a skilled person had no reason for making the above-mentioned selection. Despite the fact that document D9 acknowledged D2 as prior art on page 4, last paragraph, a skilled person would not combine both documents, since in document D2 the Ta powder was doped with nitrogen after the reduction merely by diffusion in a NH_4Cl atmosphere (cf. e.g. D2, page 5, 4th paragraph, example 7), and not during the reduction step as disclosed in D9. Neither D9 nor D2 provided nitrogen bubbling through the molten salt during the reduction step to provide a solid solution of nitrogen in the Ta-matrix, as set out in the patent at issue. The combined teaching of documents D9 and D2 therefore did not lead a skilled person to the claimed process.

The method stipulated in the independent process claim according to the fifth to fourteenth auxiliary requests was, therefore, novel and involved an inventive step.

Reasons for the Decision

1. The appeal is admissible.
2. Main request, first to fourth auxiliary requests; novelty; Article 54(1) EPC (1973)

Claim 2 of the main, first and second auxiliary requests and claim 1 of the third and fourth auxiliary requests all relate to the same method of producing a nitrogen containing tantalum powder comprising the steps of introducing a nitrogen containing gas into the reaction system while a Ta-compound is reduced with a reducing agent. By this process, nitrogen is simultaneously incorporated into the tantalum.

According to the process set out in example 4 of document D10, a tantalum compound (TaCl_5 , melting point: 216°C , boiling point 242°C) is mixed with a reducing agent (MgH_2) and red phosphorus as a dopant. The physical constants of TaCl_5 are generally known to the expert e.g. from the Handbook of Chemistry and Physics, 56th edition, 1975/1976, CRC Press Inc, Cleveland Ohio, 44128, page B148, or alternatively from wikipedia.org. These references are cited for corroboration only and do not constitute further evidence.

On being electrically ignited, the MgH_2 in the hot mixture reduces the tantalum compound to elemental Ta (cf. D10, page 4, first full paragraph). The example specifically mentions that the reduced tantalum powder is simultaneously nitrided at least in part during the reduction step by nitrogen (gas) contained in the ambient air and that the resulting Ta-powder comprises 1.2 wt. % nitrogen (12000 ppm N; cf. D10, page 15).

This concentration of the dopant nitrogen falls within the concentration range of 50 to 20000 ppm N stipulated in the patent at issue.

The patent proprietor's argument that the technical term "Nitridierung" in example 4 of D10 means the "formation of tantalum nitride" (TaN) has no bearing on the matter since "nitridieren" is the chemically correct term for the frequently used expression "nitrieren" which in fact means introducing nitrogen in a solid alloy by holding it at a suitable temperature in contact with a nitrogenous material (usually ammonia). Reference is made in this context to <http://de.wikipedia.org/wiki/Nitridierung> which is cited only for corroboration and not as further evidence. Moreover, the claimed method does not specify any distinct type or form in which the nitrogen is to be incorporated in the tantalum powder.

Hence, the subject matter of claim 2 of the main request, first and second auxiliary requests and of claim 1 of the third and fourth auxiliary requests lacks novelty over the disclosure of document D10.

2. Fifth to twelfth auxiliary requests:

- 2.1 In the method set out in claim 2 of the sixth and eighth auxiliary requests and in claim 1 of the tenth and twelfth auxiliary requests, a nitrogen containing gas is introduced into a molten salt while reducing a halide of tantalum by a reducing agent in the molten salt at 700 to 1000°C. As previously mentioned, the additional feature that nitrogen is simultaneously incorporated into the tantalum does not represent a

separate and independent process step, but rather qualifies the result obtained by this process.

Claim 2 of the fifth and seventh and claim 1 of the ninth and eleventh auxiliary requests are even broader in scope given that a tantalum compound (rather than a halide of Ta) is reduced in a liquid phase system (rather than a molten salt) without defining a temperature range at all.

- 2.2 Doping tantalum with a dopant, such as N, Si, P or B, is well known in the art to form an electrode which improves the properties of a resultant capacitor (e.g. voltage breakdown, storage energy). Like the patent at issue, document D9 is concerned with the production of tantalum powder via the reduction of K_2TaF_7 in a diluent salt melt. When the temperature of the molten K_2TaF_7 -NaCl charge has been adjusted to $980^\circ C$, the reducing agent (sodium) is added to reduce the potassium-tantalum fluoride to tantalum powder which after screening and blending may be used as a "primary powder", or may be agglomerated into a porous mass to form a "secondary powder" (cf. D9, column 2, line 60 to column 3, line 5; column 3, lines 21 to 30). D9 goes on saying in column 3, lines 31 to 38 that *"primary and secondary powders can be modified by additions of other materials (e.g. phosphorus, silicon, nitrogen) at primary or secondary stages (or during the original reduction). If the additives (P, Si, N) or compound sources thereof are provided during reduction, it must be done in a way to avoid creating an oxidising condition in the reactor."*

When putting into practice this clear technical teaching the person skilled in the art, faced with the problem of doping the tantalum powder with nitrogen, would seriously contemplate carrying out the doping during the original reduction, i.e. when reducing the K_2TaF_7 with Na in the molten salt bath at $980^\circ C$. In so doing, he is further taught to add nitrogen to the molten salt either in elemental form (N) or as a nitrogen-bearing compound such as ammonia, ammonium salts, urea etc. The use of such compounds as a nitrogen source is commonly known in the art. Reference is made in this context e.g. to document D2, page 5, paragraph 4 and page 4, last paragraph, specifically disclosing NH_4Cl .

- 2.3 It is true, as alleged by the patent proprietor, that document D9 points out several stages and constituents that could be chosen to dope the primary and secondary powders with P, N or Si, as argued by the patent proprietor. However, given that document D9 makes particular reference to providing the dopant additive(s) during the original reduction and to adhere to non-oxidising conditions in the reactor, a skilled person's attention would be preferably drawn to this technique as a promising method for doping Ta.

Contrary to the patent proprietor's position, the Board also considers that the passage "to avoid creating oxidising conditions" in D9 has to be interpreted as essentially avoiding the addition of oxygen-bearing compounds (as for instance $NH_4(OH)$) rather than excluding the addition of nitrogen gas in general.

2.4 The process set out in claim 2 of the fifth to eighth auxiliary requests and in claim 1 of the ninth to twelfth auxiliary requests is therefore obvious from document D9 taken individually. Consequently, the subject matter of these claims does not involve an inventive step.

3. Thirteenth and fourteenth auxiliary requests:

3.1 Claim 1 of the thirteenth and fourteenth auxiliary requests further comprise the feature "*wherein the nitrogen containing metallic powder is a solid solution comprising 50-20,000 ppm nitrogen*". In the patent proprietor's view, this feature is neither disclosed in nor made obvious from any of the cited documents.

3.2 Apart from being clearly reflected in the patent specification itself, it is beyond dispute that the feature of incorporating nitrogen as a dopant in the form of a solid solution into the Ta-matrix per se does not represent an independent technical feature, but merely qualifies the result that is achieved by adding nitrogen or a nitrogen compound during the reduction of K_2TaF_7 to the molten salt (cf. the patent specification, paragraphs [0020], [0056], [0057], [0080]). Hence, this feature does not establish a patentable distinction over the disclosure of document D9, all the more so since it could be correctly inferred that nitrogen doping generally results in an interstitial solid solution at least until reaching the solubility limit for N in Ta-metal for a specific temperature (cf. for instance D8, column 3, lines 39 to 47; column 3, lines 1 to 5). Moreover, the concentrations for nitrogen-doping typically range from 4700 to 27100 ppm,

as mentioned in document D8, column 3, lines 17 to 19. As can be seen also from D10, page 5, lines 23 to 26 or from D2, claim 5, the tantalum powder generally comprises 30 to 15000 ppm N. Selecting the claimed amounts of nitrogen interstitially dissolved as a dopant in the Ta-powder, therefore, does not involve an inventive step either.

3.3 Given that the claimed process per se was obvious to a person skilled in the art, as has been shown in the previous sections, it is concluded that the feature which defines the properties of the final product resulting from this process does not add patentable matter to the claimed process and hence does not involve an inventive step.

4. Since none of the claims set out in any of the main request or the first to fourteenth auxiliary requests relate to novel and inventive subject matter, the patent has to be revoked.

Order

For these reasons it is decided that:

The patent is revoked.

The Registrar:

The Chairman:

V. Commare

T. Kriner