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**Datasheet for the decision  
of 15 March 2012**

**Case Number:** T 0573/10 - 3.2.03  
**Application Number:** 02762773.6  
**Publication Number:** 1418012  
**IPC:** B22F 9/24, H01G 9/052  
**Language of the proceedings:** EN

**Title of invention:**

Nitrogen containing metal powder and method for producing the same, and porous sintered product and solid electrolytic capacitor using the same

**Applicant:**

Cabot Super Metals K.K.

**Opponent:**

-

**Headword:**

-

**Relevant legal provisions:**

EPC Art. 123(2), 54, 56

**Keyword:**

"Amendments - broadening of claim (no)"

"Novelty (yes)"

"Inventive step (no)"

**Decisions cited:**

-

**Catchword:**

-



Case Number: T 0573/10 - 3.2.03

**D E C I S I O N**  
of the Technical Board of Appeal 3.2.03  
of 15 March 2012

**Appellant:**  
(Applicant)

Cabot Super Metals K.K.  
9-5, Shibakouen 2-chome  
Minato-ku  
Tokyo 105-0011 (JP)

**Representative:**

Grünecker, Kinkeldey  
Stockmair & Schwanhäusser  
Anwaltssozietät  
Leopoldstrasse 4  
D-80802 München (DE)

**Decision under appeal:**

Decision of the Examining Division of the  
European Patent Office posted 19 October 2009  
refusing European patent application  
No. 02762773.6 pursuant to Article 97(2) EPC.

**Composition of the Board:**

**Chairman:** I. Beckedorf  
**Members:** G. Ashley  
C. Donnelly

## Summary of Facts and Submissions

I. This appeal arises out of the decision of the examining division to refuse European patent application No. 02 762 773.6 (EP-A-1 418 012) for added subject-matter contrary to Article 123(2) EPC.

II. The decision was posted by the examining division on 19 October 2009. The appellant (the applicant) filed notice of appeal on 21 December 2009, paying the appeal fee on the same day; a statement containing the grounds of appeal was filed on 26 February 2010.

III. Oral proceedings were held on 15 March 2012.

IV. Requests

The appellant requested that the decision under appeal be set aside, and that a patent be granted on the basis of one of the sets of claims filed as main request and as first and second auxiliary requests with the letter of 26 February 2010 and filed as new second auxiliary request during the oral proceedings before the examining division on 3 June 2009 at 10:30 am (third auxiliary request).

V. Claims

(a) Claim 1 of the main request reads as follows:

"1. A production process for a nitrogen containing metal powder, wherein said metal is at least one metal selected from a group consisting of niobium, tantalum, and niobium-tantalum alloy, wherein the ratio W/S

between the nitrogen content  $W[\text{ppm}]$  and the specific surface area  $S[\text{m}^2/\text{g}]$ , as measured by a BET method, is within the range 500 to 2000, wherein

a metal salt containing said metal is reacted with a reducing agent and undergoes reduction within a molten diluent salt, thereby generating said metal, and

a nitrogen-containing gas is introduced into the space contacting the reaction melt comprising said metal salt, said reducing agent and said diluent salt, and onto the top of the reaction melt while the reduction reaction of the metal is carried out,

thereby generating said metal and incorporating said nitrogen within said metal."

Dependent claims 2 to 4 concern preferred embodiments of the process of claim 1.

(b) Claim 5 is directed to a metal powder, and claim 6 relates to a use for the powder; these claims read as follows:

"5. A nitrogen containing metal powder having a specific surface area  $S[\text{m}^2/\text{g}]$  of at least 1.0, as measured by a BET method, wherein a ratio  $W/S$  between a nitrogen content  $W[\text{ppm}]$ , and the specific surface area  $S$  is within a range from 500 to 2000, wherein said metal is tantalum and wherein said nitrogen exists as a solid solution within said metal."

"6. Use of the nitrogen containing metal powder according to claim 5 for the production of a porous sintered body."

(c) Claims 1 to 3 of the application as originally filed (EP-A-1 418 012) are as follows:

"1. A nitrogen containing metal powder comprising a metal powder that contains nitrogen, wherein a ratio W/S between a nitrogen content W[ppm], and the specific surface area S[m<sup>2</sup>/g], as measured by a BET method, is within a range from 500 to 3000.

2. A nitrogen containing metal powder according to claim 1, wherein said nitrogen exists as a solid solution within said metal.

3. A nitrogen containing metal powder according to claims 1 or 2, wherein said metal is at least one metal selected from a group consisting of niobium, tantalum and niobium-tantalum alloy."

VI. Prior Art

- D5: US-B1-6 238 456
- D5-1: Analytical experiments concerning examples 4 and 7 of D5, filed during examination proceedings with the letter of 21 May 2008.
- D14: WO-A98/37248
- D15: Declaration of Dr J. W. Koenitzer, filed with the letter of 27 January 2008.

VII. Submissions of the Appellant

(a) Article 123(2) EPC

The appellant submitted that present claim 5 is based on claims 1 to 3 and page 8, lines 13 and 14, of the application as originally filed, with the claim being limited to tantalum.

(b) Novelty (Article 54 EPC)

The appellant submitted that the wording of claim 5, and in particular the expression "said nitrogen" indicates that all nitrogen is in solid solution; other forms of nitrogen in the claimed metal powder, such as nitrides are excluded from the wording of claim 5.

The examining division had referred to the disclosure on page 17, second full paragraph, of the application, where it is said that "almost no crystalline nitrides" are contained in the powder, as indicating that minor amounts are allowable in the claimed powder. To this, the appellant argued that the cited passage relates to a powder in which the W/S ratio is between 500 and 3000, whereas in the present claim 5 it is limited to between 500 and 2000, ie to a lower nitrogen content. At lower nitrogen contents, the likelihood of forming crystalline nitrides is much less, and claim 5 expressly requires that all nitrogen is in solid solution. Hence the expression "almost no crystalline nitrides" does not apply to the powder of present claim 5.

The appellant said that, in practice, the feature that all nitrogen exists in solid solution relates to the ability to detect the presence of crystalline compounds using X-ray diffraction analysis by a means known at the priority date. Should some crystalline nitrides be present, but in such a small quantity that they could not be detected by X-ray diffraction analysis, the powder is considered to contain all nitrogen in solid solution.

- Document D5

Example 7 has been cited as being the most relevant example in D5. An analysis of the powder produced according to example 7 (submitted as document D5-1) shows that there are minor amounts of tantalum nitride in the powder. Thus the powder of claim 5 is novel over D5.

- Document D14

The appellant argued that the reaction used to produce the tantalum powder of D14 is highly exothermic, reaching white heat (page 4, lines 2 to 3), and nitrogen is only added during this reaction phase. At the high temperatures associated with this process, tantalum nitride is formed rather than the nitrogen entering solid solution.

The examining division considered that D14 also discloses (page 7, lines 8 to 10) a low temperature method for producing nitrogen-containing tantalum powder. The appellant responded by arguing that the passage cited by the examining division refers to the

"adjustment" of the nitrogen content, implying that nitrogen has already been introduced into the powder during the reaction phase.

(c) Inventive Step

Starting from the cited prior art, the appellant sees the objective problem as providing an improved tantalum powder, and in particular reducing the leakage current of a capacitor made from such a powder.

The objective problem is solved by ensuring that all of the nitrogen in the powder is in solid solution. This is achieved by the process described in the application, which enables the nitrogen to be added carefully under controlled conditions.

Since there is no indication in the cited documents as to how a powder containing nitrogen only in solid solution could be achieved, the claimed subject-matter has an inventive step.

**Reasons for the Decision**

1. The appeal is admissible.
2. Article 123(2) EPC
  - 2.1 The examining division held that the subject-matter of product claim 5 (which was claim 6 of the main request before the examining division) did not meet the requirements of Article 123(2) EPC (see Reasons, page 2, point 2).



Product claim 6 before the examining division was directed to "a nitrogen containing metal powder comprising a metal powder that contains nitrogen... wherein said metal is tantalum...". The examining division considered that the claim referred to metal powders in general, but only niobium, tantalum and niobium-tantalum alloys were disclosed in the original application, hence the amended claim was an unallowable generalisation contrary to Article 123(2) EPC.

2.2 The Board does not agree with the examining division, as the claim is limited to tantalum, which is disclosed in dependent claim 3 of the original application (see V(c) above). In addition, the specific surface area  $S$  of the metal powder is stated on page 9, lines 6 to 7 of the original description as being preferably at least  $1.0 \text{ m}^2/\text{g}$ , and the preferred W/S range is given on page 8, lines 13 to 14 as being 500 to 2000. The subject-matter of present claim 5 can thus be derived from the original application.

2.3 The examining division also commented, albeit *obiter*, that the introduction of the feature:

"...a nitrogen-containing gas is introduced into a space on top of the melt..."

into the definition of the process in claim 1 has been taken out of context, and thus also amounts to an unallowable generalisation (see point 1 of the *obiter dictum* of the contested decision).

The production process for obtaining the metal powder of the invention is summarised on page 8, lines 15 to 20 of the application, where it is said that the nitrogen-containing gas is introduced into a space contacting the reaction melt comprising the metal salt, the reducing agent and the diluent salt. Figure 1 and accompanying description (page 12, lines 1 to 4) also show nitrogen being introduced onto the top of the reaction melt. This feature is both disclosed in the original application and is essential to the invention, hence the amendment is not contrary to Article 123(2) EPC.

2.4 The claimed subject-matter of the main request therefore meets the requirements of Article 123(2) EPC.

3. Novelty (Article 54 EPC)

3.1 Novelty and inventive step of the claimed process (claims 1 to 4) have not been questioned by the examining division. However, as an *obiter* to its decision, the examining division expressed the view that the metal powder of claim 5 lacks novelty with respect to D5 or D14.

*Document D5*

3.2 Example 7 (column 11 and Table 5) of D5 discloses a tantalum powder having a specific surface area of 1.54 m<sup>2</sup>/g (BET) and a nitrogen content of 2750 ppm, giving a W/S ratio of 1786. These figures fall within the claimed ranges.

3.3 The question is whether or not the nitrogen in the powder of example 7 exists in solid solution, as defined in claim 5. D5 does not discuss the form of the nitrogen in the powder, however, the appellant has submitted experimental evidence that includes an X-ray diffraction analysis of example 7; the results are presented in document D5-1. The X-ray analysis shows the powder to be tantalum with traces of tantalum nitride  $Ta_2N$ , ie all nitrogen except for a trace amount is in solid solution.

3.4 The examining division was of the opinion that the wording of claim 5 does not exclude the presence of minor amounts of tantalum nitride in the powder, hence the claimed powder lacked novelty in light of example 7.

The wording of claim 5 is "A nitrogen containing metal powder... wherein said nitrogen exists as a solid solution within said metal.". The expression "said nitrogen" refers to the nitrogen contained in the metal powder. The Board thus agrees with the appellant's construction of the claim, that use of the word "said" indicates that all of the nitrogen contained in the powder is in solid solution.

3.5 However, it is important to consider what the skilled person would understand by the feature "all of the nitrogen is in solid solution". The presence or absence of crystalline phases containing nitrogen in the powder is determined by X-ray diffraction analysis (see page 20 of the application, first paragraph). By using this analysis technique, no crystalline phases were observed in examples 1 to 3, which relate to tantalum powders of the invention. The claimed feature that "all of the

nitrogen is in solid solution" therefore has to be understood in light of the description, ie all of the nitrogen is in solid solution, insofar as no crystalline phases of nitrogen can be detected using X-ray diffraction.

- 3.6 The examining division referred to the third paragraph on page 17 of the application, which states that the powder contains "almost no crystalline nitrides", ie the invention as defined in claim 1 also allows for trace amounts.
- 3.7 The cited paragraph is discussing powders in which the W/S ratio falls within the range 500 to 3000. The significance of this range is described on page 8, lines 9 to 14, where it is said that if the W/S ratio exceeds 3000 the quantity of nitrogen becomes excessive, which "increases the possibility of the generation of crystalline nitrides". In other words, at the higher end of the range, around 3000, there is more likelihood of crystalline nitrides being formed. Hence, the reference to "almost no crystalline nitrides" in the paragraph cited by the examining division is referring to the upper part of the range where there is a possibility that some crystalline nitrides may form.
- 3.8 Present claim 1 defines the upper limit of the range as 2000, which implies that there is less likelihood that crystalline nitrides will form. Claim 1 requires *inter alia* a powder both to have a W/S ratio in the defined range and to have no detectable crystalline nitrides. Since some nitrides can be detected in the powder of example 7 of D5, the subject-matter of claim 5 is novel over D5.

*Document D14*

- 3.9 As with D5, document D14 discloses a tantalum powder meeting the requirements of claim 5, save that it is not clear if all of the nitrogen is in solid solution.
- 3.10 According to the process of D14, a the tantalum powder is formed by igniting a mixture containing tantalum pentachloride, magnesium hydride and a nitrogen-containing compound such as ammonium chloride or air (D14, page 6, lines 1 to 2 and 16 to 19; examples 4 and 9 to 16). As argued by the appellant, the reaction is highly exothermic and reaches white heat (page 4, lines 2 to 3); nitrogen is added during this reaction phase and at these high temperatures tantalum nitride is formed rather than the nitrogen entering solid solution (see first page, fourth paragraph of the declaration D15 filed by Dr Koenitzer). This appears to be the case, as was accepted by the examining division, but the examining division also considered that D14 discloses a low temperature method (page 7, line 8 to 10) in which the nitrogen would be in solid solution. This, however, concerns the "adjustment" of the nitrogen content, which as argued by the appellant, implies that nitrogen has already been introduced into the powder during the reaction phase, particularly as none of the examples show nitrogen being added at the latter stage. There is no clear indication in D14 that the nitrogen is inevitably in solid solution, and on this basis the subject matter of claim 5 is novel with respect to D14.

4. Inventive Step

4.1 Both D5 and D14 disclose tantalum powders containing nitrogen and hence can be considered as appropriate starting points for assessing inventive step. The metal powder of claim 5 differs from either of these documents in that all of the nitrogen is defined as being in solid solution.

4.2 Starting from either D5 or D14, the objective problem to be solved is to improve the tantalum powder, and in particular, the properties of a capacitor having an anode made from the tantalum powder.

4.3 This is achieved by ensuring that the nitrogen in the powder exists in solid solution.

It is well known to improve the properties of capacitors by doping tantalum powders with nitrogen, of which D5 and D14 are two examples. However, as set out above, neither D5 nor D14 discloses a powder in which all of the nitrogen is in solid solution. This effect is achieved in the present application by introducing a nitrogen-containing gas into the space on top of the reaction melt, while the reduction reaction of metal salts to tantalum takes place. This allows better control of the incorporation of nitrogen than is possible with the known prior art methods (see second paragraph on page 13 of the application), with the result that no detectable crystalline nitrides form.

4.4 Since none of the cited documents suggest a tantalum powder in which all the nitrogen is in solid solution, or a process by which such a powder could be produced, the claimed subject-matter has an inventive step.

5. Auxiliary Requests

Since the claims of the main request are allowable, there is no reason to consider those of the auxiliary requests.

## Order

### For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the department of first instance with the order to grant a patent in the following version:

Claims: 1 to 6, filed as the main request with the letter of 26 February 2010;

Description: Pages 1 to 5, 10 to 12, 14 to 16 and 18 to 25, as originally filed;

pages 6 to 9, 13, 17, 26 and 27, filed during the oral proceedings;

Figures: 1 and 2, as originally filed.

The Registrar:

The Chairman:

D. Hampe

I. Beckedorf