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**Datasheet for the decision
of 24 April 2012**

Case Number: T 1398/10 - 3.2.03
Application Number: 06252814.6
Publication Number: 1738848
IPC: B22F 1/02, B22F 3/00,
B22F 9/18, C23C 16/44
Language of the proceedings: EN

Title of invention:

Coating metal powder particles with metal by chemically
reducing nonmetallic precursors

Applicant:

GENERAL ELECTRIC COMPANY

Headword:

-

Relevant legal provisions:

EPC Art. 56

Keyword:

"Inventive step (no)"

Decisions cited:

-

Catchword:

-



Case Number: T 1398/10 - 3.2.03

D E C I S I O N
of the Technical Board of Appeal 3.2.03
of 24 April 2012

Appellant:
(Applicant)

GENERAL ELECTRIC COMPANY
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Representative:

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Decision under appeal:

Decision of the Examining Division of the
European Patent Office posted 23 December 2009
refusing European patent application
No. 06252814.6 pursuant to Article 97(2) EPC.

Composition of the Board:

Chairman: U. Krause
Members: G. Ashley
K. Garnett

Summary of Facts and Submissions

I. This appeal arises from the decision of the examining division to refuse European patent application No. 06 252 814.6 (EP-A-1 738 848). The examining division was of the view that the amendments made to claim 1 during the examination procedure did not fulfil the requirements of Article 123(2) EPC, that the wording of claim 1 lacked clarity (Article 84 EPC), and that the claimed subject-matter lacked novelty and an inventive step.

II. The decision was posted on 23 December 2009. The appellant (the applicant) filed notice of appeal on 1 March 2010, paying the appeal fee on the same day; a statement of the grounds of appeal was filed on 28 April 2010.

III. Oral proceedings were held on 24 April 2012.

IV. Requests

The appellant requested that the above decision be set aside and that a patent be granted on the basis of the set of claims filed as the main request during the oral proceedings before the Board.

V. Claims

Claim 1 of the appellant's sole request reads as follows:

"1. A method for making an article (20), comprising the steps of:

furnishing a plurality of powder particle substrates (80) made of a substrate metal;

providing a nonmetallic precursor of a metallic coating material, wherein the metallic coating material comprises an alloying element that is thermophysically melt incompatible with the substrate metal, such that:

- i) the alloying element has an evaporation rate of greater than 10 times that of the melt temperature of the substrate metal; or
- ii) the melting point of the alloying element is greater than or less than that of the substrate metal by more than 400°C; or
- iii) the alloying element has a density difference with the substrate metal of greater than 0.5 gram per cubic centimeter; or
- iv) the alloying element or a chemical compound formed between the alloying element and the substrate metal chemically reacts with the substrate metal in the liquid phase to form deleterious phases in the melt; or
- v) a miscibility gap exists between the alloying element and the substrate metal in the liquid phase;

contacting the powder particle substrates (80) with the nonmetallic precursor; and

chemically reducing the nonmetallic precursor to form coated particles (102) comprising the powder particle substrates (80) having a surface-enriched layer (120) of metal coating material thereon, the step of chemically reducing being performed without melting the powder particle substrates (80), and processing the

coated powder particles (102) to form the article (20) without melting the powder particle substrates;

characterized in that:

the powder particle substrates (80) are formed of a superalloy nickel-base alloy having at least two phases, including a continuous matrix phase with a face-centered-cubic crystal structure that is strengthened by both solid solution strengthening and the presence of one or more additional discrete phases that are distributed throughout the matrix phase, where the discrete phases have a different composition than the matrix phase, said alloying element being bismuth, cadmium, calcium, magnesium, gallium, hafnium, indium, lanthanum, niobium, rhenium, tantalum, tungsten, molybdenum, erbium, europium, gadolinium, nitrogen, neodymium, yttrium, silver, barium, lithium, lead or thallium."

Dependent claims 2 to 8 concern preferred embodiments of the method of claim 1.

VI. Prior Art

The following documents cited by the examining division are relevant for this decision:

D1: C. Vahlas et al., "Fluidization, Spouting, and Metal-Organic CVD of Platinum Group Metals on Powders", Chem. Vap. Deposition, Wiley-VCH Verlag, Weinheim, DE, Vol. 8, No. 4, July 2002, pages 127 to 144.

D4: US 2004/0208773 A1.

The following document was referred to by the appellant during the oral proceedings before the Board:

D5: Page 317 with the heading "Corrosion and Protection of Superalloys", said to be taken from Donachie & Donachie, "Superalloys a Technical Guide", 2002.

VII. Submissions of the Appellant

The submissions of the appellant can be summarised as follows:

Inventive Step

(a) Starting from D4

Document D4 discloses a method for incorporating thermophysically melt incompatible alloying elements into an alloy, by which melting is avoided throughout the production procedure, with the result that these elements are homogenously distributed throughout the material of D4.

Starting from D4, the problem to be solved is to improve control over the spatial distribution of such incompatible elements in the article.

The solution according to the method of claim 1 is to locate the incompatible alloying elements in the surface coating of substrate particles. There is no hint of this feature either in D4 or in D1. In addition,

if the process of D1 is followed, it inevitably involves melting of the particles during the final consolidation step.

The claimed method thus has an inventive step.

(b) Starting from D1

The appellant submits that, although D1 discloses particles having a nickel-base substrate, this is a coating powder which is not a superalloy and does not have the microstructure as required by claim 1. In addition, the coating powder is applied using high temperature techniques such as plasma deposition which do not avoid melting, a feature that is essential to the method of claim 1.

Starting from D1, the problem to be solved is how to improve the strength of the resulting article and/or allow for a wider range of alloying elements to be incorporated into the material.

Given the strong emphasis in D1 for applying the coating powders by high temperature techniques, there is no reason for the skilled person to take D4 into account and modify the method of D1 so that melting is avoided. Consequently, the claimed method is not obvious starting from the disclosure of D1.

Reasons for the Decision

1. The appeal is admissible.

2. Admissibility of D5

The appellant referred to D5 during the oral proceedings before the Board, as evidence that the nickel alloy referred to in D1 is not a superalloy having a continuous matrix phase with a face-centred-cubic (fcc) crystal structure strengthened by both solid solution strengthening and the presence of discrete phases distributed throughout the matrix, as is defined in claim 1.

The appellant said that D5 was an extract from "Corrosion and Protection of Superalloys", Donachie & Donachie, "Superalloys, a Technical Guide", 2002. However, no evidence was provided to support the origin of the document.

In addition, D5 does not provide a detailed description of the microstructure of the material; it merely states that "the composition and microstructure of the coating alloy depends on postdeposition treatment and service exposure".

Given that D5 was filed late in the proceedings, is of unknown origin and does not seem to be relevant, it is not admitted into the proceedings.

3. Inventive Step (Article 56 EPC)

Starting from D4

- 3.1 Document D4 discloses a method for making an article from an alloy containing thermophysically melt

incompatible alloying elements without any melting taking place.

The alloy is formed from a base alloy, such as a nickel-base alloy (paragraph [0008]), and is used to make components for gas turbine engines (paragraph [0021]). It is thus clear that the method of D4 concerns superalloys, ie alloys typically having an fcc crystal structure and which exhibit strength at high temperatures as a result of solid solution strengthening and the presence of fine discrete phases in the alloy.

The alloys of D4 also contain thermophysically melt incompatible elements, such as those which have a significantly higher evaporation rate compared with the base metal (paragraph [0028]), those with a melting point that is too high or too low (paragraph [0029]), those that have a density difference of more than 0.5 g/cm^3 (paragraph [0030]), those that form deleterious phases (paragraph [0031]) and those that lead to the formation of a miscibility gap (paragraph [0032]).

According to the method of D4, the material is prepared by providing a non-metallic precursor compound. This is chemically reduced to an alloy in the form of powder particles (paragraph [0048]), which are then consolidated into an article (paragraph [0050]). Both of these steps take place without melting.

- 3.2 As submitted by the appellant, the claimed method differs from that of D4 in that it results in particles of a substrate metal coated with a metallic material

comprising the thermophysically melt incompatible element. The method of D4, on the other hand, leads to a powder in which the thermophysically melt incompatible element is uniformly distributed.

- 3.3 Starting from D4, the appellant formulated the problem to be solved as being how to have a better control over the distribution of thermophysically melt incompatible alloying elements in the material (see also paragraph [0022] of the application).

However, the subject-matter of claim 1 is formulated more broadly, merely requiring the formation of coated particles, which are then processed to form the article. Although the thermophysically melt incompatible elements are present in the surface layer of the substrate particles, there is no requirement that the claimed method leads to them being located in particular regions of the bulk material or article.

In addition, the particles of D4 could also be used in combination with other types of powders to locate the thermophysically melt incompatible elements at specific regions within the article.

Consequently, the objective problem is to provide an alternative way of incorporating thermophysically melt incompatible elements into the alloy (see paragraphs [0005] and [0006] of the application).

- 3.4 The use of coated particles is well known in the art of powder metallurgy, and D1 describes coating platinum group metals onto particle substrates made of a nickel-based NiCoCrAlYTaN alloy (NI-482, Praxair). Such powders

are used for coating components of turbine engines (section 4.2 on page 138).

- 3.4.1 According to the specific example on page 139 of D1, the nickel-based particles are coated with ruthenium but, given the overall disclosure of D1, the process could clearly apply to a coating of any of the platinum group metals. Platinum is identified in paragraph [0033] of the application as being a thermophysically melt incompatible element, and the examining division calculated that, in the cited example, ruthenium would have a melting point more than 400°C higher than the nickel alloy substrate, which corresponds to definition (ii) of a thermophysically melt incompatible element given in claim 1. Hence D1 discloses deposition of thermophysically melt incompatible materials onto substrate metal particles.
- 3.4.2 D1 teaches (section 4.2) that corrosion and oxidation properties of bond coats based on nickel-based powders are improved by doping the alloy with elements such as Hf, Ir, Pd, Pt, Re, Ru, Ta and Zr, but there are difficulties regarding the mixing process when alloying such elements with high melting temperatures.
- 3.4.3 The solution given in D1 is to coat commercially available nickel-based powders with, for example, ruthenium. According to the process described in the last paragraph of the right-hand column on page 139, the substrate powders are contacted with a non-metallic precursor compound (eg $\text{Ru}(\text{C}_2\text{H}_5)_2$) which is reduced to the metal by hydrogen. Since contacting and coating the substrate powder with the precursor compound takes

place simultaneously, there is no melting of the powder particle substrate.

- 3.5 Starting from D4, and bearing the teaching of D1 in mind, the skilled person would realise that the step of providing an alloying compound (step 42 of Figure 2) can be also be carried out by coating the base-metal compound particles. In doing so, the method as defined in claim 1 is achieved, hence this method lacks an inventive step.

Starting from D1

- 3.6 Alternatively, there is a lack of inventive step if D1 is considered as the starting point for the claimed method.
- 3.7 As set out above, D1 discloses the coating of nickel-based particles with a thermophysically melt incompatible material, namely a platinum group metal.
- 3.8 The appellant submits that D1 fails to disclose powder substrates formed of a superalloy nickel-based alloy, having the structure defined in the claim.
- 3.8.1 Superalloy is a general term for alloys having good mechanical and corrosion properties at high temperatures, and this applies to the nickel-based alloy referred to in D1. However, D1 does not discuss the nature of the substrate alloy in detail. The examining division argued (point 5.1 on page 6 of the decision) that when powders are formed from by plasma spraying the NiCoCrAlYTa alloy, it is inevitable that the alloy is strengthened by both solid solution

strengthening and by discrete phases such as precipitates. The Board agrees with this view, as the alloy contains cobalt, chromium and aluminium, which are well known in the art for the solid solution hardening of nickel alloys. Likewise, chromium, aluminium, yttrium and tantalum are well known for forming precipitates or intermetallics such as a γ' phase, ie discrete phases having a different composition to that of the matrix metal.

- 3.8.2 The examining division recognised novelty of the claimed method on the basis that D1 does not disclose that the substrate alloy has a fcc crystal structure. However, nickel alloys generally have an fcc matrix. The appellant submitted D5 as evidence that the structure of the nickel alloy of D1 does not correspond to the requirements of claim 1. However, as stated in Point 2 above, D5 does not give a clear description of the metallurgical structure of the alloy of D1 that would rebut the presumption that the nickel alloy of D1 meets the requirements of claim 1.
- 3.9 According to D1, the powder particles are used as a bond coat for thermal barrier coatings on turbine components, and are applied by plasma deposition or high velocity oxy-fuel deposition (first paragraph of 4.2 on page 138). Hence, as argued by the appellant, D1 does not disclose a method in which melting is avoided.
- 3.10 Starting from D1, the objective problem to be solved is to improve the incorporation of thermophysically melt incompatible elements into the alloy, and hence improve the strength of the resulting article.

3.11 D4 teaches that this can be achieved by avoiding melting throughout the process, including the step of consolidating the powder particles. Adapting the method of D1 so that melting is avoided, or to use the powders described in D1 in a consolidation step that avoids melting, is therefore obvious in light of D4 for a skilled person wishing to improve the distribution of platinum group metals in the alloy, and hence the method of claim 1 lacks an inventive step.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

D. Hampe

U. Krause