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**Datasheet for the decision  
of 2 March 2010**

**Case Number:** T 0952/08 - 3.2.08

**Application Number:** 04253497.4

**Publication Number:** 1486575

**IPC:** C22C 1/04

**Language of the proceedings:** EN

**Title of invention:**

Method for preparing metallic superalloy articles without melting

**Applicant:**

GENERAL ELECTRIC COMPANY

**Opponent:**

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**Headword:**

-

**Relevant legal provisions:**

EPC Art. 54, 56

EPC R. 115, 118

**Relevant legal provisions (EPC 1973):**

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**Keyword:**

"Novelty (no)"

"Inventive step (no)"

**Decisions cited:**

-

**Catchword:**

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Case Number: T 0952/08 - 3.2.08

**D E C I S I O N**  
of the Technical Board of Appeal 3.2.08  
of 2 March 2010

**Appellant:**

GENERAL ELECTRIC COMPANY  
1 River Road  
Schenectady  
NY 12345 (US)

**Representative:**

Bedford Grant Richard  
GE  
London Patent Operation  
15 John Adam Street  
London WC2N 6LU (GB)

**Decision under appeal:**

Decision of the Examining Division of the  
European Patent Office posted 20 December 2007  
refusing European patent application  
No. 04253497.4 pursuant to Article 97(1) EPC.

**Composition of the Board:**

**Chairman:** T. Kriner  
**Members:** R. Ries  
E. Dufrasne

## Summary of Facts and Submissions

- I. European patent application No. 04 253 497.4 (published as EP-A-1 486 575 concerns a method for preparing superalloy articles without melting.
- II. With its decision posted on 20 December 2007, the examining division refused the application. The grounds for this decision were contained in the communications dated 5 August 2005 and 21 September 2006 wherein the applicant (appellant) was informed that the subject matter of claim 1 then on file lacked novelty (Article 54 EPC). The examining division took inter alia the following documents into account:
- D1: Eoghan T. M. Doyle and Animesh Jha: "Carbothermic Reduction of Nickel-Cobalt-Chromium Oxide Mixture for the Production of NiCoCr Superalloy Powder", EPD Congress 1992, Edited by J.P. Hager, The Minerals, Metals and Materials Society, 1991, pages 745 to 758;
- D2: US-A-4 894 086;
- D3: EP-A-1 449 928 (Article 54(3) EPC document);
- D5: EP-A-1 433 555 (Article 54(3) EPC document).
- III. The appellant filed a notice of appeal which was received at the European Patent Office on 25 February 2008 and paid the appeal fee on the same date. Enclosed with the statement setting out the grounds of appeal received on 29 April 2008, the appellant submitted a revised set of claims 1 to 10.

IV. In the communication accompanying the summons to oral proceedings, the Board issued a preliminary opinion, setting out its view on the issue of novelty and inventive step of the subject matter of claim 1 vis-à-vis the technical teaching given in the documents D1 to D3 and D5, respectively.

V. Enclosed with its response, dated 2 February 2010, to the official communication, the appellant submitted six auxiliary requests in addition to the main request filed on 29 April 2008.

By a further letter dated 25 February 2010, the appellant informed the Board that it would not attend the oral proceedings scheduled for 2 March 2010 and requested the Board of Appeal to issue a written decision on the basis of the present requests.

VI. Oral proceedings took place on 2 March 2010 in accordance with Rule 118 EPC. As announced by letter of 25 February 2010 and although having been duly summoned the appellant did not attend the oral proceedings. Therefore, according to Rule 115(2) EPC and Article 15(3) of the Rules of Procedure of the Boards of Appeal (2008), the proceedings were continued without the appellant.

In the written proceedings, the appellant requested that the decision under appeal be set aside and a patent be granted on the basis of

- claims 1 to 10 of the main request submitted on 29 April 2008, or, in the alternative,

- claims 1 to 9 of the first to sixth auxiliary requests, respectively, all submitted on 2 February 2010.

VII. Claim 1 of the main request reads as follows:

"A method for preparing an article (20) of a base metal alloyed with an alloying element, comprising the steps of

providing a chemically reducible nonmetallic base-metal precursor compound of a nickel-base metal;

providing a chemically reducible nonmetallic alloying-element precursor compound of an alloying element, wherein the alloying element is thermophysically melt incompatible with the base metal and selected from the group consisting of: bismuth, cadmium, calcium, magnesium, chromium, gallium, hafnium, indium, lanthanum, niobium, rhenium, tantalum, tungsten, molybdenum, erbium, europium, gadolinium, nitrogen, neodymium, yttrium, silver, barium, lithium, lead, thallium, arsenic, gold, beryllium, germanium, scandium, silicon, zirconium, zinc or cerium; thereafter

mixing the base-metal precursor compound and the alloying-element precursor compound to form a compound mixture; thereafter

chemically reducing the compound mixture to produce a metallic superalloy without melting the metallic superalloy so that the gross shape of the metallic superalloy material remains unchanged; and thereafter

consolidating the metallic superalloy to produce a consolidated metallic article (20), without melting the metallic superalloy and without melting the consolidated metallic article (20) so that the gross

shape of the metallic article (20) also remains unchanged."

Claim 1 of the first to sixth auxiliary requests, respectively, differs from claim 1 of the main request by the addition of the following features (bold letters added by the Board):

First auxiliary request:

" A method ...cerium, **wherein the alloying element has a vapor pressure of greater than 10 times a vapor pressure of the base metal in a melt of the base metal, both measured at a melt temperature;** thereafter...unchanged."

Second auxiliary request:

"A method ...cerium, **wherein the alloying element has a melting point different from that of the base metal by more than 400°C;** thereafter...unchanged."

Third auxiliary request:

"A method ...cerium, **wherein the alloying element has a density difference with the base metal of greater than 0.5 gram per cubic centimeter;** thereafter...unchanged."

Fourth auxiliary request:

"A method ...cerium, **wherein the alloying element, if melted, chemically reacts with the base metal in a liquid phase to form chemical compounds including the base metal and the alloying element;** thereafter...unchanged."

Fifth auxiliary request:

"A method ...cerium, **wherein the alloying element, if melted, exhibits a miscibility gap with the base metal in the liquid phase;** thereafter...unchanged."

Sixth auxiliary request:

"A method ...cerium, **wherein the alloying element, if melted, chemically reacts with the crucible material or the melting atmosphere;** thereafter...unchanged."

VIII. The appellant's arguments in the written proceedings can be summarized as follows:

Document D1 did not clearly and unambiguously relate to a process which was wholly without melting, as defined in claim 1 of all requests. In that respect document D1 stated on page 751, lines 18 to 20 that "... the alloy phase produced was in the form of frozen droplets of metallic liquid." Even if the known alloy was consolidated in a manner without melting, the reduction process itself could still not be considered to be performed without a melting step.

In addition, D1 failed to consider the thermophysical melt incompatibility of alloying elements by suggesting on page 756 that a molten metallic liquid phase be used. Clearly, the use of such a molten phase would result in the separation of any thermophysically melt incompatible elements such that no alloy would be formed between them.

Document D2 related to the production of alloys with fine dispersions of oxide powders therein without any indication that the base and alloying materials were themselves thermophysically melt incompatible. In this

process the second phase precipitates were added as metals or alloy, respectively, because they cannot be reduced by a reduction reaction (see D2, column 1, lines 46 to 50). In the example given in D2, column 2, lines 48 to 55, aluminium, titanium and chromium were added as metals because they could not be reduced by the known method. Accordingly, D2 failed to disclose a process that was wholly without melting and in which a chemical reduction of a compound mixture of non-metallic base metal compounds and non-metallic alloying element precursor compounds occurs to produce a metallic superalloy in which the alloying element was thermophysically melt incompatible with the base metal.

Regarding the citations D3 and D5, which were to be considered only with respect to novelty, none of these documents related to the use of thermo-physically melt incompatible elements.

Accordingly, none of the cited documents clearly and unambiguously taught or suggested the method of claim 1 of the main request or any of the first to sixth auxiliary requests, respectively, in which an alloying element was thermophysically melt incompatible with the nickel-base material.

### **Reasons for the Decision**

1. The appeal is admissible.



2. *Main request*

2.1 Claim 1 of the primary request relates to a method for producing a superalloy article, without melting at any stage of the process, comprising the steps of

(a) providing a non-metallic precursor compound of a nickel-base metal;

(b) providing a non-metallic precursor compound of an alloying element selected from the group set out in claim 1;

(c) mixing the non-metallic precursor compounds;

(d) chemically reducing the mixture of non-metallic precursor compounds to produce a metallic superalloy material so that the gross shape of the metallic superalloy material remains unchanged; and

(e) consolidating the superalloy material to a metallic article so that the gross shape of the metallic superalloy article remains unchanged.

2.2 Document D3, representing prior art pursuant to Article 54(3) EPC, relates to Ni-base superalloys which are produced by chemically reducing, without melting, a mixture of non-metallic precursor compounds of Ni and of alloying elements including elements listed in present claim 1. Particular reference is made to the flow-chart depicted in D3, Figure 2 and paragraphs [0007] to [0010]; [0014]; [0015]; [0018]; [0027]; [0028]; [0030]; [0039], in particular lines 38 to 45; and [0041].

Exemplifying compositions of Ni-base superalloys well known in the art as trademarks, which are producible with the known process are given in D3, paragraph [0028] and include Alloys 718, 706, 720, Waspaloy, GTD222,

Rene<sup>TM</sup>220, Rene<sup>TM</sup>88 (also mentioned on page 13, lines 18 to 27 of the application as originally filed) and MERL76. Contrary to the appellant's position, all these Ni-base superalloys comprise alloying elements which are "thermophysically melt-incompatible" with the Ni-base material within the meaning given in the present application.

The mixture of non-metallic precursor compounds (claimed steps (a) to (c)) is chemically reduced without melting the superalloy material. There may be some minor amount of localized melting as low-melting-point elements do melt and are diffusively alloyed with the higher-melting-point elements which do not melt. Even in such cases the gross shape of the material remains unchanged (see document D3, paragraph [0030]). This statement complies with the explanations given in the originally filed application on page 18, second full paragraph). Hence the process of D3 also anticipates step (d) of the claimed process.

The different types of solid-state consolidation after the reduction step described in document D3, paragraphs [0015] to [0018] are the same as those used in the claimed process and described on page 8, last line to page 9 line 6 of the application as originally filed. As in the claimed process, there is no melting of the metallic form in the known process so that the gross shape of the metallic superalloy material remains unchanged. Thus step (e) of the claimed process is also anticipated by the process described in document D3.

Apart from the statement that D3 did not relate to producing an alloy material in which an alloying

element is thermophysically melt incompatible with the Ni-base material, which does not apply, the appellant did not present any specific arguments with respect to the novelty of the claimed process vis-à-vis the technical disclosure of document D3.

Consequently, the process set out in claim 1 of the main request lacks novelty vis-à-vis the technical disclosure of document D3.

- 2.3 Likewise, document D1 is concerned with the production of a superalloy powder by chemically reducing non-metallic precursor compounds of (a) a nickel base metal and (b) an alloying element to form the final alloy. To this end, the oxides NiO, CoO, Cr<sub>2</sub>O<sub>3</sub> and, at least in part, Nb<sub>2</sub>O<sub>5</sub> are provided in a mixture (c) which is pressed to a cylindrical shape (pellets) and chemically reduced (d) at temperatures between 1523 K and 1623 K (see D1, Table I, page 750; and Experimental, page 748). Crushing the reduced pellets to form a powder for identifying new phases described in D1 page 750, last paragraph is understood as meaning that the gross shape of the pellet consisting of the metallic superalloy material remained unchanged.

The observation reported in D1 on page 757, second paragraph that a liquid NiCrCo alloy has formed above 1523 K implies that for the examples given in Table 1 of D1 and treated at 1523 K, a solid-state reduction without melting of the mixture has taken place.

The appellant's argument based on D1, page 751, second paragraph according to which the visual examination of the reaction product revealed that the alloy phase

produced was in the form of frozen droplets of metallic liquid is not disputed.

It is however noted that the formation of droplets in D1 is to be read in the light of the explanation of the term "without melting" and "no melting" given in the application as originally filed on page 18, second full paragraph: *In the claimed solid state reduction process, "there may be, for example, some minor amount of localized melting as low melting elements actually melt and are diffusionaly alloyed with the higher alloying elements which do not melt. Even in such cases the gross shape of the material remains unchanged."*

Many of the alloying elements listed in claim 1 exhibit a low melting point (e.g. Be:  $T_m = 271^\circ\text{C}$ ; Cd:  $T_m = 320^\circ\text{C}$ ; Ga:  $T_m = 29,7^\circ\text{C}$ ; Li:  $T_m = 180,5^\circ\text{C}$ ). After their reduction, these low-melting metals are expected to form a liquid which alloys with the high-melting base metal Ni ( $T_m = 1453^\circ\text{C}$ ). It is noted in this context that also in the process known from document D1, the reduced pellets maintained their form and were not grossly melted so that they liquefied and lost their shape. Hence also in that respect the claimed process cannot be unambiguously distinguished from the solid state reduction process carried out at 1523 K or lower according to D1.

The metal powder produced by the process of D1 is regarded as being suitable for compacting by powder forging and extrusion processes for shape forming, corresponding to step (e) of the claimed process (see D1, page 746, third full paragraph to the last paragraph). Consequently, in the light of the technical teaching given in document D1, consolidating the

reduced metal powder e.g. by HIPing, sintering, extrusion etc without melting amounts to nothing more than conventional practice and therefore would not involve an inventive step.

In view of these considerations the process set out in claim 1 of the main request fails to comprise technical features which involve an inventive step vis-à-vis the process disclosed in document D1.

### 3. *Auxiliary requests*

3.1 As to claim 1 of the first to third auxiliary requests, the nominal composition of the exemplifying alloy 718 given in D3, column 8, lines 1 to 8 comprises, amongst other components such as Nb, Ta, Mo Ti, Al and Fe, specifically chromium which exhibits an evaporation rate of greater than 10 times that of the Ni-base metal at melt temperature (see in this context page 15, lines 7 to 12 of the application as filed).

3.2 Niobium and tantalum in alloy 718 exhibit a melting point different from nickel of more than 400°C (see the application as filed, page 15, lines 21 to 27).

3.3 Moreover, the density difference between Mo, W, Ta and Nb and the base metal Ni is greater than 0.5 g/cm<sup>3</sup> (see the application as filed, page 16, lines 7, 8).

3.4 The composition of alloy MERL<sup>TM</sup> 76 (trademark of Pratt&Whitney) referred to in D3, column 8, line 20 as another example of Ni-base superalloys generally comprises 0.5% Hf as one element that may react with the crucible material or the melting atmosphere, and

thus represents one of the elements set out in claim 1 of the sixth auxiliary request (see the corresponding part in the application as filed page 17, second paragraph).

It is also to be noted that many of the alloying elements of Alloy 718 such as Cr, Mo, Nb, Ta etc are prone to react with the melting atmosphere and need special countermeasures to prevent such a reaction.

Hence, the subject-matter of claim 1 of the first to third and sixth auxiliary requests lacks novelty with respect to the technical disclosure of document D3.

- 3.5 The process defined in claim 1 of the fourth auxiliary request specifies that, if melted, the alloying element obtained from the chemically reducible non-metallic alloying-element precursor compound chemically reacts with the nickel-base metal to form chemical compounds including Ni and the alloying element. As further set out in the corresponding part in the description page 16, second full paragraph of the application as filed, this chemical reactivity leads in conventional melting practice inter alia to the formation of intermetallic compounds including the base metal Ni and the alloying element.

The composition of the Ni-base alloy Rene<sup>TM</sup>88, referred to as an exemplifying alloy in the application as filed on page 13, lines 18 to 32 and also in document D3, column 8, line 20 comprises specific proportions of Mo and W, respectively which are known to the metallurgist to form intermetallic compounds with Ni in the solid state. Thus the process shown in document D3 of

preparing a final article having a composition comparable to superalloy Rene<sup>TM</sup>88 anticipates all the technical features of the process set out in claim 1 of the fourth auxiliary request.

Hence the subject-matter of claim 1 of the fourth auxiliary request lacks novelty over the technical disclosure of document D3.

The process set out in the fifth auxiliary request is concerned with the production of superalloys comprising alloying elements which are immiscible with the Ni-base metal in the liquid state. The person skilled in metallurgy is well aware of the fact that for certain metals it is impossible to form an alloy with other metals in a melt. One reason could reside in the fact that the metals to be alloyed exhibit a huge difference in their melting point or that they are actually immiscible in the liquid state. The only feasible method of producing a structural part comprising immiscible components is the powder-metallurgical (PM) route. Hence, if the problem of immiscibility of an alloying element with Ni arose, it could be successfully solved by using the (PM) process disclosed in document D1. Like the features set out in claim 1 of the fifth auxiliary request, document D1 describes a (PM) process for producing a superalloy powder that is suitable for powder forging and extrusion processes for shape forming. As previously shown, the other process steps essentially comply with those of the process claim 1 of the fifth auxiliary request (see D1, page 746, last paragraph).

The subject-matter of claim 1 of the fifth auxiliary request therefore lacks an inventive step in view of the technical teaching of document D1 in combination with the general technical knowledge of the person skilled in the art.

4. In view of the considerations above, the claims of the main request and also of the first to sixth auxiliary requests are not allowable.
5. The appellant, by not attending the oral proceedings which it had requested itself, waived the possibility of commenting on the above mentioned objections in a dialogue for which the Board had been prepared.

## **Order**

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

The appeal is dismissed.

The Registrar:

The Chairman:

V. Commare

T. Kriner