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Datasheet for the decision of 10 September 2010

Case Number: T 0969/08 - 3.3.05

Application Number: 99935200.8

Publication Number: 1123908

IPC: C04B 35/38

Language of the proceedings: EN

Title of invention:
High-temperature strength and heat-resistant composite material "REFSIC"

Applicant:
INSTITUT FIZIKI TVERDOGO TELA AKADEMII NAUK SSSR

Headword:
Refractory/NAUK

Relevant legal provisions:
EPC Art. 54, 56, 84, 123(2)

Keyword:
"Clarity: yes"
"Extension beyond the content of the application as filed: no"
"Novelty: yes"
"Inventive step (yes) non obvious alternative"

Decisions cited:
-

Catchword:
-

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Case Number: T 0969/08 - 3.3.05

D E C I S I O N
of the Technical Board of Appeal 3.3.05
of 10 September 2010

Appellant: (Applicant)
INSTITUT FIZIKI TVERDOGO TELA AKADEMII NAUK SSSR
Moskovskaya obl.
p/o Chernogolovka, 142432 (SU)

Representative:
Klusmann, Peter
Hoffmann - Eitle
Patent- und Rechtsanwälte
Arabellastraße 4
D-81925 München (DE)

Decision under appeal:
Decision of the Examining Division of the European Patent Office posted 16 November 2007 refusing European application No. 99935200.8 pursuant to Article 97(1) EPC 1973.

Composition of the Board:

Chairman: G. Raths
Members: J.-M. Schwaller
H. Preglau

Summary of Facts and Submissions

I. This appeal lies from the decision of the examining division refusing European patent application No. 99 935 200.8.

II. Claims 1, 2, 3 and 5 of the main request - filed during the oral proceedings before the examining division - underlying the contested decision read as follows:

"*1. A refractory and temperature-resistant composite material comprising silicon carbide and high-melting metal silicide components, Me_5Si_3 and $MeSi_2$, wherein Me_5Si_3 is the component: W_5Si_3 and Mo_5Si_3; or $(Mo,W)_5Si_3$; or $(Mo,W)_5Si_3C$; or Mo_5Si_3C; or any combination thereof, and $MeSi_2$ is the component: $MoSi_2$ and WSi_2; or $(Mo,W)Si_2$; or a combination thereof, wherein the composite material comprises the following ratio of the components (in vol. %):*

Me_5Si_3	*from 15 to 85 vol%*
$MeSi_2$	*from 0.8 to 55 vol%*
Silicon carbide	*from 2 to 85 vol%*

and wherein the ratio of molydenum (Mo) and tungsten (W) in the total mass of the high-melting metals in the material is in the range (in wt%):

Mo	*7-80*
W	*20-93.*

2. A composite material according to claim 1, characterized in that the high-melting metal silicide components, Me_5Si_3 and $MeSi_2$, besides molybdenum and tungsten, comprise rhenium as a further metal in an amount of 0.5-20 at.%. with respect to the total content of molybdenum and tungsten.

3. A composite material according to claim 1, characterized in that 5-80% of the volume of the material which is not occupied by the high-melting metal silicide components, Me_5Si_3 and $MeSi_2$, comprises inclusions of graphite and/or carbon fibers.

5. A composite material according to claim 1, characterized in that the high-melting metal silicide components, Me_5Si_3 and $MeSi_2$, besides molybdenum and tungsten comprise at least one further element of the group consisting of tantalum, niobium, titanium, zirconium, hafnium, with the following ratio of these elements with respect to the total content of molybdenum and tungsten (in wt%): Ta: 0.1-18, Nb: 0.1-8, Ti: 0.05-10, Zr: 0.05-8, and Hf: 0.1-16."

III. The following documents were inter alia cited in the search report:

D1: WO 95/31417

D5: US 5 454 999

IV. In its first official communication of 13 May 2005 (first two lines), the examining division briefly acknowledged - without however indicating the reasons - the novelty and inventive step of independent claim 1 as originally filed.

V. Despite this early acknowledgement of the patentability, the examining division finally rejected the application because the amendments to dependent claims 2, 3 and 5

of the main request extended beyond the content of the application as originally filed.

VI. With the grounds of appeal dated 26 March 2008, the appellant filed two amended sets of claims as a main and auxiliary request, respectively.

Amended claim 1 of the main request read as follows:

"1. A refractory and temperature-resistant composite material comprising silicon carbide and molybdenum disilicide $MoSi_2$, characterized in that it further comprises W_5Si_3 and Mo_5Si_3 and/or $(Mo,W)_5Si_3$ and/or $(Mo,W)_5Si_3C$, and also WSi_2 and/or $(Mo,W)Si_2$ with the following ratio of the components (in vol.%):

(i) total of W_5Si_3, Mo_5Si_3, $(Mo,W)_5Si_3$ and $(Mo,W)_5Si_3C$: 15-85%,

(ii) silicon carbide: 2-85%,

(iii) total of tungsten and molybdenum disilicides WSi_2 and $MoSi_2$ and $(Mo,W)Si_2$: 0.8-55%,

the ratio of molybdenum and tungsten in the total mass of the high-melting metals in the material ranging within (in wt.%)

• Mo 7—80%,

• W 20—93%,

wherein rhenium may substitute 0.5-20 at.% of the molybdenum and tungsten in the material,

and wherein at least one of tantalum, niobium, titanium, zirconium, hafnium may substitute molybdenum and tungsten in the silicide phases in the following amounts with respect to the total content of molybdenum and tungsten (in wt.%): Ta, 0.1—18; Nb, 0.1-8; Ti, 0.05—10; Zr, 0.05—8; Hf, 0.1-16 and wherein inclusions

of graphite and/or carbon fibers may partially substitute silicon carbide, in an amount of 5-80 % of the volume not occupied by silicides of high-melting metals."

VII. In a communication under Rule 100(2) EPC, the board objected to above claim 1 under Article 84 EPC, as it was unclear which silicides had to be taken into consideration in the calculation of the "*volume not occupied by silicides of high-melting metals*".

VIII. With a letter dated 22 June 2010, the appellant submitted an amended set of claims as the new main request, claim 1 of which reads as follows: (differences with the main request filed with the grounds of appeal emphasised by the board):

"*1. A refractory and temperature-resistant composite material comprising silicon carbide and molybdenum disilicide $MoSi_2$, characterized in that it further comprises W_5Si_3 and Mo_5Si_3 and/or $(Mo,W)_5Si_3$ and/or $(Mo,W)_5Si_3C$,* ***and/or Mo_5Si_3C****, and also WSi_2 and/or $(Mo,W)Si_2$ with the following ratio of the components (in vol.%):*

(i) total of W_5Si_3, Mo_5Si_3, $(Mo,W)_5Si_3$ and $(Mo,W)_5Si_3C$ ***and Mo_5Si_3C****: 15-85%,*

(ii) silicon carbide: 2-85%,

(iii) total of tungsten and molybdenum disilicides WSi_2 and $MoSi_2$ and $(Mo,W)Si_2$: 0.8-55%,

the ratio of molybdenum and tungsten in the total mass of the high-melting metals in the material ranging within (in wt.%)

• Mo 7—80%,

• W 20—93%,

*wherein rhenium may **optionally** substitute 0.5-20 at.% of the molybdenum and tungsten in the material, and wherein at least one of tantalum, niobium, titanium, zirconium, hafnium may **optionally** substitute molybdenum and tungsten in the silicide phases in the following amounts with respect to the total content of molybdenum and tungsten (in wt.%): Ta, 0.1—18; Nb, 0.1-8; Ti, 0.05—10; Zr, 0.05—8; Hf, 0.1-16 and wherein inclusions of graphite and/or carbon fibers may partially substitute silicon carbide, in an amount of 5-80% of the volume not occupied by silicides of **the** high-melting metals **Mo, W, Re, Ta, Nb, Ti, Zr and Hf."***

IX. The appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the claims 1 to 6 filed as a main request with the letter dated 22 June 2010, or alternatively, on the basis of the claims 1 to 6 filed as a first auxiliary request with the letter dated 26 March 2008 respectively.

Reasons for the Decision

1. *Main request - Amendments*

The subject-matter of dependent claims 2, 3 and 5 that the examining division objected to under Article 123(2) EPC in the contested decision no longer exists as such in the set of claims at issue.

The subject-matter of the amended claims 1 to 6 of the main request dated 22 June 2010 find their support in the application as filed as follows:

Claim 1: claims 1, 2, 3, 5; page 2, last line to page 3, line 23; page 3, line 28 to page 4, line 4.

Claim 2: claim 4

Claim 3: claim 6

Claim 4: claim 7

Claim 5: claim 8

Claim 6: claim 9

These claims thus meet the requirements of Article 123(2) EPC.

2. *Main request - Clarity*

2.1 The board notes that claim 1 of this request has been amended to recite the different high-melting metals which are to be taken into consideration in the calculation of the volume "*not occupied by silicides*". This amendment overcomes the clarity objection raised by the board in its communication to the appellant.

2.2 As an obiter dictum to the contested decision, the examining division raised further objections under Article 84 EPC.

2.2.1 In particular it considered that it was unclear from the expression "*substitute therewith*" used in certain claims then on file whether the high-melting metals rhenium, tantalum, niobium, etc …, or the inclusions of graphite and/or carbon fibers were a substitute for molybdenum or tungsten or an additional component.

The board observes that it is clear from the wording now used in claim 1, namely "*rhenium may optionally substitute 0.5-20 at.% of the molybdenum and tungsten in the material*", "*at least one of tantalum, niobium, titanium, zirconium, hafnium may optionally substitute molybdenum and tungsten*", "*inclusions of graphite and/or carbon fibers may partially substitute silicon carbide*" that the above components are not additional components but substitutes for "molybdenum and tungsten" or "silicon carbide", respectively.

2.2.2 The examining division further considered that the expression "*in an amount of 5-80% of the volume not occupied by silicides of high-melting metals"* used in the claims could not be understood because it was unclear:

- which volume was considered in the above expression, in particular whether the said volume was the one of the silicon carbide;

- how the claim could be understood when silicon carbide to be partially substituted was for instance only 2 vol.% of the composition and the volume not occupied by silicides was for instance 50 vol.%.

As to this clarity objection, the appellant offered the following explanation, which also satisfies the board.

The term "*volume not occupied by silicides of high-melting metals*" meant the volume not occupied by the components (i) and (iii) defined in claim 1. In the case where the claimed composite material included only the components (i)-(iii), then the volume referred to was the volume of the silicon carbide. However, it might be that other components contribute to the total volume of the composite material; for example, pores might also be present, in which case the amount of 5-80% not occupied by silicides of high-melting metals would relate to the total volume of silicon carbide plus the total volume of the pores present.

The theoretical example referred to by the examining division did not lead to any inconsistencies. Assuming the composite included only components (i)-(iii) as set out in claim 1, then the silicon carbide volume would correspond to the volume not occupied by silicides of high-melting metals, so both would be 2% by volume. The 50% figure related to the amount of the volume not occupied by silicides of high-melting metals which was replaced; in the specific example mentioned by the examining division, this would be 1% (half of 2%). Therefore, the sample referred to by the examining division was one where 1 % by volume of the sample was SiC and 1% by volume was carbon.

2.3 For the above reasons, the wording of the claims, in particular of claim 1 at issue, is in compliance with the requirements of Article 84 EPC.

3. *Main request - Novelty*

The board can accept the examining division's conclusion that the subject-matter of claim 1 a tissue is novel. In particular it is observed that none of the cited state of the art documents discloses the addition of W_5Si_3 and Mo_5Si_3 and/or $(Mo,W)_5Si_3$ and/or $(Mo,W)_5Si_3C$ in a total amount of 15-85% in vol.% to a composite material comprising molybdenum and tungsten disilicides and silicon carbide.

Claim 1 (and claims 2 to 6, which are dependent on claim 1) thus meet the requirements of Article 54 (1) and (2) EPC.

4. *Main request - Inventive step*

4.1 Inventive step has also already been acknowledged by the examining division and the board does not see any reason to depart from this conclusion for the following reasons.

4.2 The application in suit concerns a composite material comprising silicon carbide and high-melting metals (Mo, W, Re, Ta, Nb, Ti, Zr and Hf) silicides, which material is said to be refractory and temperature resistant.

4.3 Document D1, which represents the closest state of the art and hence, the starting point for assessing inventive step, discloses composite silicide or carbide heating element compositions including improved combustion sources and refractory silicides such as tungsten and/or molybdenum disilicides. According to D1, combustion synthesised composite heating elements

including the said combustion sources could be used at temperatures up to 1900°C for long periods of time in oxidising atmosphere (D1, page 1, lines 1 to 6; claim 1).

4.4 In the light of D1, the problem underlying the application in suit can be seen in the provision of a further material having high heat resistance and high resistance to thermal shocks.

4.5 As a solution to this problem the application in suit proposes the composite material according to claim 1, characterised in particular in that it comprises W_5Si_3 and Mo_5Si_3 and/or $(Mo,W)_5Si_3$ and/or $(Mo,W)_5Si_3C$ in a total amount of 15-85% in vol.%.

4.6 In view of the Examples in the application as filed, which show that the six different materials falling within the scope of protection of claim 1 at issue are able to withstand thermal shocks and temperatures of up to 1900°C, the board is satisfied that the above-mentioned technical problem has been solved.

4.7 The next step is to assess whether the technical solution to the above problem is obvious in view of the state of the art.

The board notes that D5 concerns silicide/SiC composites such as $MoSi_2$/SiC (column 2, lines 27 to 30) and refers to the coexistence of a three phase field comprising $MoSi_2$, SiC and $C_{<1}Mo_{<5}Si_3$ (D5, column 5, lines 59 to 61). There is however no hint in D5, let alone in the other documents of the search report that W_5Si_3 and Mo_5Si_3 and/or $(Mo,W)_5Si_3$ and/or $(Mo,W)_5Si_3C$ can be added

in a total amount of 15-85% in vol.% to the composite heating element compositions disclosed in D1 with the aim of providing a further material having in particular high heat resistance and high resistance to thermal shocks. In this context, the subject-matter of claim 1 at issue cannot be regarded as being obvious to a person skilled in the art.

So, claim 1 of the main request (and claims 2 to 6 which depend on claim 1) meet the requirements of Article 56 EPC.

5. The description does not need any harmonisation with the subject-matter of the claims according to the main request, so the application is ready for grant.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to grant a patent on the basis of the set of claims 1 to 6 according to the main request filed with letter of 22 June 2010 and the description as originally filed.

The Registrar: The Chairman:

C. Vodz G. Raths