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D E C I S I O N
of 18 November 2004

Case Number: T 0656/02 - 3.2.2

Application Number: 90114721.5

Publication Number: 0411591

IPC: C22C 1/04

Language of the proceedings: EN

Title of invention:
Cold accumulating material and method of manufacturing the same

Patentee:
KABUSHIKI KAISHA TOSHIBA

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56, 84

Keyword:
"Inventive step, (yes) after amendment"

Decisions cited:
-

Catchword:
-

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0656/02 - 3.2.2

D E C I S I O N
of the Technical Board of Appeal 3.2.2
of 18 November 2004

Appellant: (Proprietor of the patent)
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Decision under appeal: **Interlocutory decision of the Opposition Division of the European Patent Office posted 24 April 2002 concerning maintenance of European patent No. 0411591 in amended form.**

Composition of the Board:

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

Summary of Facts and Submissions

I. The grant of European patent No. 0 411 591 was mentioned on 16 July 1997.

II. The granted patent was opposed on the grounds that its subject matter lacked novelty and did not involve an inventive step (Article 100(a) EPC), that it did not disclose the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art (Article 100(b) EPC) and that its subject matter extended beyond the content of the application as filed (Article 100(c) EPC).

In the opposition proceedings, ***inter alia*** the following documents were referred to:

D1: Sahashi Masashi, Tokai Yoichi, Li Rui, Ogawa Masatsugu, Hashimoto Gishu: C1-11 "Development of Refrigerator Using Magnetic Regenerator Material (3) - Manufacture of Spherical Magnetic Particles", Proceedings of the 40th Autumn Conference of the Cryogenic and Super Conductivity Society, Kukuoka, November 20-22 (1988), page 63, published on 20 November 1988 (in Japanese language and accompanied by a translation into English language)

D3: Declaration by the inventor of the patent in suit (and author of D1)

III. In its letter dated 4 January 2002, the opponent withdrew from the opposition procedure.

IV. In the interlocutory decision posted on 24 April 2002, the opposition division held that, taking into account the amendments made by the patent proprietor during the opposition procedure according to the second auxiliary request, the European patent No. EP 0 411 591 and the invention to which it relates meet the requirements of the EPC.

V. An appeal against this decision was filed on 22 June 2002 by the patentee, and the appeal fee was paid on the same date. The statement of grounds was submitted on 26 August 2002.

VI. Oral proceedings before the Board were held on 18 November 2004, at the end of which the appellant requested that
- the decision under appeal be set aside and
- the patent be maintained as granted (main request),
- or on the basis of the auxiliary requests I to IV all filed on 18 October 2004.
- or on the basis of the auxiliary requests V and VI both filed on 18 November 2004.

VII. Claim 1 as granted reads:

"1. A cold accumulating material with a local maximum of its volumetric specific heat curve in an extremely low temperature range comprising:

a set of particles having a total weight, packed in a gas permeable package wherein each particle contains at least one kind of rare earth element selected from a group consisting of yttrium (Y), lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), promethium (Pm), samarium (Sm),

europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), and ytterbium (Yb), at least 70% of said total weight is comprised of particles having a particle size of 0.01 to 3 mm and at least 70% of said weight is comprised of particles which each have a major diameter to a minor diameter that is not greater than 5."

Claim 1 of the auxiliary requests I to IV differs from claim 1 as granted by the amendments of the last features as indicated in **bold** letters:

Auxiliary request I:
"1. ... and at least 70 % of said weight is comprised of particles which each have a **ratio of the** major diameter to **the** minor diameter that is not greater than **2**."

Auxiliary request II:
"1. .... and at least **90**% of said weight is comprised of particles which each have a **ratio of the** major diameter to **the** minor diameter that is not greater than **2**."

Auxiliary request III:
"1. ...and at least **80**% of said weight is comprised of particles which each have a **ratio of the** major diameter to **the** minor diameter that is not greater than **1.3**."

Auxiliary request IV:
"1. ... and at least **90**% of said weight is comprised of particles which each have a **ratio of the** major diameter to **the** minor diameter that is not greater than **2 and at least 80% of said weight is comprised of particles**

which each have a ratio of the major diameter to the minor diameter that is not greater than 1.3."

Auxiliary request V differs from claim 1 as granted by the following amendment of its last feature and additionally a new feature:
"1. ... at least 70% of said weight is comprised of particles which each have **a ratio of the** major diameter to **the** minor diameter that is not greater than 5 **and wherein no more than 30% by weight of the particles have small cracks of a length of 10 µm or greater relative to the whole of said particles, said particles being produced by a molten quenching method.**"

Auxiliary request VI reads as follows:
"1. A cold accumulating material with a local maximum of its volumetric specific heat curve in an extremely low temperature range comprising:

a set of particles having a total weight, packed in a gas permeable package wherein each particle **consists of a material selected from**

- **a magnetic compound AM_z, wherein A is one or two kinds of** rare earth elements selected from a group consisting of yttrium (Y), lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), promethium (Pm), samarium (Sm), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), and ytterbium (Yb), **and M is a metal or**

- **one or two kinds of rare earth elements selected from the group consisting of yttrium (Y), lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd),**

promethium (Pm), samarium (Sm), europium (Eu), gadolinium (Gd), terbium (Tb),dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), and ytterbium (Yb), or

- **a boride, sulfide, oxide, carbide and nitride of one or two kinds of rare earth elements selected from the group consisting of yttrium (Y), lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), promethium (Pm), samarium (Sm), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), and ytterbium (Yb),**

at least 70% of said total weight is comprised of particles having a particle size of 0.01 to 3 mm and at least 70% of said weight is comprised of particles which each have **a ratio of the** major diameter to **the** minor diameter that is not greater than 5**, and wherein no more than 30% by weight of the particles have small cracks of a length of 10 µm or greater, relative to the whole of said particles, said particles being produced by a molten quenching method.**"

VIII. The appellant argued as follows:

Claim 1 of the present invention suggested for the first time a cold accumulating material having a high mechanical strength and chemical stability as well as a high packing density and resistance to thermal shock and vibrations which were applied during the operation. These overall properties were achieved by three technical features: the composition of the material,

the shape of one discrete particle unit and the group of particles forming a set of particles.
The particle size was restricted to 0.01 and 3 mm as to guarantee an adequate fluid flow rate through the packed solid phase and to prevent the formation of micro-segregations which caused a non-uniform crystalline structure and an increased corrosion rate. The occurrence of micro-segregations and the formation of small cracks and defects of > 10 µm in the particles was effectively suppressed by using the molten metal quenching method for producing the particles. Moreover, the claimed cold accumulating material was comprised of a set of particles whereof 70% or more by weight had an aspect ratio of not greater than 5, preferably 2 or even 1.3. In so doing, the particle strength and the packing density was increased and a cold accumulating material having durability sufficiently high for practical use was obtained.

Although document D1 disclosed a magnetic particulate regenerator material consisting of $ErNi_2$, Er_3Ni or Er_3Co, this document only addressed the shape of the particles but did not give any information about a specific aspect ratio, a particle size range and distribution to adhere to when packing a set of particles in a refrigerating unit. Figures 1 and 3 of document D1 merely indicated several more or less spherical particles (out of several millions to be packed in a refrigerating unit) that were prepared by the conventional plasma-spraying method. This very limited selection of particles given in Figures 3 and 4 could, however, not be regarded as being representative for all particles produced with this method. Moreover, document D1 was also silent about the presence or

absence of small cracks or defects in the interior or on the particle surface and could not give a hint as to avoid these defects.

The cold accumulating material according to claim 1, according to the main request and all subsequent auxiliary requests was, therefore, neither taught nor made obvious by the technical teaching given in document D1.

Reasons for the Decision

1. The appeal is admissible.

2. *The prior art*

2.1 Like the patent at issue, document D1 is concerned with the mass-production of magnetic particles to be used as a regenerator material in a compact refrigerator. Particles consisting of Er_3Ni, Er_3Co or $ErNi_2$ are described as having improved regenerating abilities below the temperature of 15 K. A particle size ranging from a few microns to a few hundred microns is preferred to achieve better heat transfer characteristics. It is found that, by using a high pressure plasma jet spray gun, good spherical particles could be obtained and that the ratio of forming the source particles into spheres was improved. As an example, a group of spherical magnetic particles produced by this method is shown in Figure 3, and a single spherical particle of about 80 µm in diameter is given in Figure 4.

However, no specific information is given in D1 about the preferred powder distribution, size and aspect ratio of the particles.

2.2 The subject matter of claim 1 of all requests is, therefore, novel.

3. *Inventive step; main request and auxiliary requests I to IV*

3.1 The appellant's view is not contested that document D1 fails to specify a powder distribution of more than 70% by weight of particles satisfying the conditions stipulated in claim 1 of the patent at issue. The evaluation of the contents of document D1, however, clearly shows that the authors of D1 have aspired to produce, to the highest extent possible, a uniform and spherical powder distribution or, more specifically, spherical particles having an aspect ratio close to 1 and a size ranging from a few µm up to a few hundred µm. It is, therefore, close at hand to stipulate a proportion of 70%, 80% or even 90% of particles having the claimed size and an aspect ratio not greater than 5 or, more preferably, not greater than 2 or even 1.3 since this is exactly what is aimed at by the process known from D1 and by a skilled person putting into practice this process. Even in case that the desired powder distribution and size is not immediately achieved by plasma spraying, it could, for instance, be obtained simply by classifying the powder known from D1.

Apart from this general teaching, document D1 further comprises more detailed technical information in the exemplifying figures. The appellant itself has

determined the size and aspect ratio of 21 individual particles shown in D1, Figure 3. As set out in Exhibit I, Table R1, enclosed with the appellant's statement of the grounds of appeal, the major diameter of the particles varies between about 60 to about 200 µm, and the aspect ratio of the majority of the particles is close to about 1.1 or less, with 2.93 being the highest value of all tested samples. Contrary to the patentee's allegations and the inventor's declaration (D3), there is nothing in document D1 for concluding or implying that the mass-produced particles depicted in both Figures 3 and 4 should not be rated as being a fair representation of the powder obtained by the high pressure plasma spraying and quenching method resorted to in this document. Hence, the limiting technical features given in claim 1 of the main request and auxiliary requests I to IV amount to nothing more than what is aimed at by a person skilled in the art and by the authors of document D1.

Consequently the subject matter of claim 1 according to the main request and auxiliary requests I to IV does not involve an inventive step.

3.2 Auxiliary request V

The wording of claim 1 sets out that each particle "contains" at least one rare earth element so that it remains speculative for a skilled person what the exact composition of the particles is supposed to be so that the desired cold accumulating material is successfully achieved.

In the Board's view, claim 1 of the auxiliary request, therefore, fails to satisfy the requirements of Article 84 EPC.

3.3 Auxiliary request VI

3.3.1 Amendments

Claim 1 according to auxiliary request VI results from a combination of claims 1 and 5 as granted and the technical features given in the description on page 4, lines 35 to 45, page 5, lines 16 to 25 and lines 50 to 53, page 7, lines 16 to 21.
Dependent claims 2 to 6 correspond to claims 2 to 4, 6 and 7 as granted.

The description has been suitably adapted to the revised claims.

Hence, there are no formal objections to the present claims under Article 123(2) and (3) EPC.

3.3.2 Novelty and inventive step

The cold accumulating material stipulated in claim 1 of auxiliary request VI is defined (i) by its exact chemical composition, (ii) by the absence of cracks having a length of ≥10 µm in at least 70% by weight of the particles and (iii) in that the particles are produced by the molten quenching method which brings about the specific properties and superior overall performance of the claimed material.

As set out on page 5, lines 43 to 45 of the patent specification, defects of more than 10 µm adversely affect the mechanical strength, the cooling characteristics and the cold accumulation efficiency of the particles. Therefore, a technical effect is attributed to this restriction (feature (ii)).

With respect to feature (iii), it is discernable from the patent specification as a whole that specific technical properties of the claimed particles including surface smoothness, the absence of segregations inside each particle and superficial oxide films, the uniformity of the structure, the high yield and the improvements associated therewith (i.e. the mechanical strength, the chemical characteristics, reduced costs) are a direct consequence of the molten metal quenching method (cf. page 5, lines 50 to 53; page 4, line 55 to page 5, first line; page 5, lines 16 to 31; page 14, lines 8 to 13). The patent specification points out that these properties and improvements are not achieved with particles prepared by the "conventionally used" plasma spray method which provides for adding solid particulate material to a hot plasma jet (cf. the specification on page 3, lines 16 to 24). The Board concurs with the appellant's view that this process (and the products obtained therewith) is totally different from the molten metal quenching method which is rated in the patent under consideration as representing one of the key features of the present invention.

Specifically, the technical features (ii) and (iii) are neither described in nor suggested by the technical teaching disclosed in document D1 or any other prior

art. As set out above in more detail, document D1 is confined to the type of particles obtained by plasma spraying and does not give a hint or possibility of recognising the presence or absence of defects and cracks in or on the particle surface and problems associated therewith.

3.4 In view of the considerations above, the subject matter of claim 1 of the auxiliary request VI satisfies the requirements of the EPC.

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 the first instance with the order to grant a patent with the following documents:

   - Claims: 1 to 6 according to auxiliary request VI as filed on 18 November 2004

   - Description: pages 2 and 3 as filed with letter of 18 October 2004
     pages 4 to 14 as granted

   - Figures: 1 to 11 as granted

The Registrar: The Chairman:

V. Commare T. Kriner