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D E C I S I O N
of 4 February 1997

Case Number: T 0907/93 - 3.4.2

Application Number: 85304139.0

Publication Number: 0165047

IPC: H01M 4/96, C04B 35/52

Language of the proceedings: EN

Title of invention:
Pseudo graphite electrode material obtained by pyrolysis

Patentee:
MITSUBISHI CHEMICAL CORPORATION

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56

Keyword:
"Novelty (yes - after amendment)"
"Inventive step (yes - after amendment)"

Decisions cited:
-

Catchword:
-

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Case Number: T 0907/93 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 4 February 1997

Appellant: MITSUBISHI CHEMICAL CORPORATION
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JP-Chiyoda Ku, Tokyo-to (JP)

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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 4 June 1993 refusing European patent application No. 85 304 139.0 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: E. Turrini
Members: R. Zottmann
L. C. Mancini

Summary of Facts and Submissions

I. European patent application No. 85 304 139.0 with the publication No. 0 165 047 was refused by decision of the Examining Division.

The reason given for the refusal was that claim 1 lacks novelty with respect to prior art disclosed in

D2: JP-A-58-93176 (English translation submitted by a Third Party)

II. The Appellant (Applicant) lodged an appeal against said decision.

III. Said Third Party filed observations under Article 115 EPC in connection with the grounds of appeal contesting novelty of the claims filed with said grounds of appeal.

IV. In communications pursuant to Article 110(2) EPC and a consultation by telephone, the Board of Appeal expressed its preliminary opinion that and why the application did not meet the provisions of the EPC.

To meet these objections, the Appellant reformulated the claims and amended the description.

V. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the following documents:

Description: page 1 as filed with the letter dated 6 June 1990;

page 2 as originally filed, whereby in line 5 "of the present" is replaced by "specified in present";

pages 3 to 8 and 10 to 26 as filed with the letter dated 23 September 1996;

page 9 as filed with the letter dated 5 December 1996;

Claims: 1 to 3 as filed with the letter dated 23 September 1996;

Drawings: sheets 1/4 to 4/4 as originally filed.

VI. Claim 1 reads as follows:

"1. A secondary battery wherein at least one electrode is made of an electrode material which is a carbonaceous material having a pseudo-graphite structure and being obtained by pyrolyzing any of the following compounds:

(a) naphthalene, perylene, terrylene, pyrene, chrysene, or coronene;

(b) a tetracarboxylic acid dianhydride of naphthalene, perylene, terrylene, pyrene, chrysene, or coronene;

(c) a corresponding tetracarboxylic acid substituted compound of naphthalene, perylene, terrylene, pyrene, chrysene, or coronene

said carbonaceous material having such a pseudo-graphite structure that the spacing of (002) planes, d002, is 3.37 to 3.70 x 10^{-10}m and

the crystallite size in the direction of c axis, LC, is 7.0 to 220 x 10^{-10}m, as determined by X-ray wide-angle diffractiometry, and the line width ΔHpp between peaks in the first differential of the absorption spectrum of electron spin resonance is at lest 10 gauss and the distance a_a, which is twice the spacing of the (110) planes, d110, is at least 2.37 x 10^{-10}m (2.37 Å) and not more than 2.47 x 10^{-10}m (2.47 Å), and the crystallite size in the direction of a axis, La, is from 10 to 80 x 10^{-10}m (10 to 80 Å),

the electrode being submerged in an electrolyte selected from alkali metal salts, alkaline earth metal salts and tetraalkylammonium salts, so that the electrode material is doped with negative ions or positive ions, thereby making the same an electrode for a secondary battery."

Claims 2 and 3 are dependent on claim 1.

VII. Arguments supporting the request to grant a patent on the basis of the claims now restricted to a particular group of compounds were not submitted by the Appellant. However, mainly the following arguments (put forward in the letter dated 29 September 1994) seem to support also said request:

The starting materials for the carbonaceous material of the present invention have only C atoms as ring-forming atoms resulting in an ability to assume a pseudo-graphite structure even at a relatively low temperature through generation of radicals and a high conductivity. On the other hand, the starting materials polyacrylonitrile (PAN) and polybenzoxazole used in D2 contain N atoms in the ring which are quite thermally stable, and denitrogenation does not proceed unless the calcination temperature is high. As a consequence, the

structure of the electrodes is less uniform. The electrode material of the invention is excellent with respect to those of D2 as regards the amount of metal ions entrapped, and the entering and emerging properties of the metal ions.

Reasons for the Decision

1. The appeal is admissible.

2. The Board is satisfied that the present application documents do not contain subject-matter extending beyond the content of the application as originally filed (requirements of Article 123(2) EPC).

3. *Novelty*

Document D2 discloses a secondary battery wherein at least one electrode is made of a carbonaceous pyrolyzed polymer. As examples of the starting material are mentioned:

PAN, polyvinyl chloride, polyvinylidene chloride, polyoxydiphenylene, polyimides, polyamide, polysemicarbazide, polybenzoxyazinone (see page 3, second paragraph) and pitch (see page 7, first paragraph). Polybenzoxazolimide and PAN are used as starting materials for the examples.

Said compounds have a chain structure, are heterocyclic and/or contain other rings than six-membered aromatic rings. Pitch contains also heterocyclic compounds and other substances. In opposition to this, the starting material according to claim 1 is selected from

particular condensed hydrocarbons containing only six-membered aromatic rings and particular derivatives of these compounds.

The other documents on file are less relevant and do not disclose such starting materials either. Mostly PAN is used for this purpose.

Therefore, the subject-matter of claim 1 is novel with respect to the prior art documents on file in the sense of Article 54 EPC.

4. *Inventive step*

The nearest prior art with respect to the subject-matter of claim 1 is disclosed in D2.

The main difference between the battery of claim 1 and that of D2 consists in the use of a particular group of compounds as starting material for the electrode, namely particular polycyclic hydrocarbons containing only condensed six-membered aromatic rings and particularly suitable derivatives of these compounds (see also section 3, above).

Apparently, the electrode material, obtained by pyrolyzing such a starting material, is improved with respect to the materials of D2 as regards the amount of metal ions entrapped, and the entering and emerging properties of the metal ions. It seems that these advantages result from the more uniform structure of the electrode material.

The problem underlying the subject-matter of claim 1 is, therefore, to improve the battery of D2 to obtain improved ion entrapping and emerging properties of the electrodes.

Neither D2 nor the other documents on file hint at the use of the particular starting materials of claim 1 of the application-in-suit. Apart from pitch, the materials of the prior art are quite different from those materials of claim 1. Though D2 mentions use of pitch as a possible starting material, containing - apart from heterocyclic compounds and other substances like soot - also hydrocarbons with condensed aromatic six-membered rings, the skilled person would not envisage use of such material alone, since in D2 a lot of heterocyclic compounds are characterized as suitable starting materials. Moreover, in all examples heterocyclic compounds are used as starting materials.

Therefore, the subject-matter of claim 1 involves also an inventive step as defined in Article 56 EPC with respect to the prior art documents on file.

5. The dependent claims concern particular embodiments of the subject-matter of claim 1 and are, therefore, likewise novel and inventive.

6. It is evident that "12" at the end of line 15 on page 9 of the description should be replaced by "7", as already done with the corresponding figure in the middle of said line.

7. In the result, the Board of Appeal takes the view that the claims comply with the requirements of the EPC. This applies also to the other documents of the application.

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 following application documents as agreed by the Board of Appeal:

Description: page 1 as filed with the letter dated 6 June 1990;

page 2 as originally filed, whereby in line 5 "of the present" is replaced by "specified in present";

pages 3 to 8 and 10 to 26 as filed with the letter dated 23 September 1996;

page 9 as filed with the letter dated 5 December 1996, whereby "12" at the end of line 15 is replaced by "7";

Claims: 1 to 3 as filed with the letter dated 23 September 1996;

Drawings: sheets 1/4 to 4/4 as originally filed.

The Registrar: The Chairman:

P. Martorana E. Turrini