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

**Case Number:** T 0432/07 - 3.3.05

**Application Number:** 99402595.5

**Publication Number:** 1035601

**IPC:** H01M 4/50

**Language of the proceedings:** EN

**Title of invention:**

Positive active material for rechargeable lithium battery and  
method of preparing same

**Applicant:**

Samsung Display Devices Co., Ltd.

**Headword:**

Coated electrode material/SAMSUNG

**Relevant legal provisions:**

EPC Art. 54(1)(2), 56, 123(2)

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

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**Keyword:**

"Novelty (yes)"

"Inventive step (yes) - non obvious alternative"

**Decisions cited:**

-

**Catchword:**

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Case Number: T 0432/07 - 3.3.05

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.05  
of 2 August 2010

**Appellant:** Samsung Display Devices Co., Ltd.  
575, Sin-Dong,  
Paldal-Ku  
Suwon-Si,  
Kyungki-do (KR)

**Representative:** Moncheny, Michel  
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**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office posted 23 October 2006  
refusing European application No. 99402595.5  
pursuant to Article 97(1) EPC 1973.

**Composition of the Board:**

**Chairman:** G. Raths  
**Members:** J.-M. Schwaller  
S. Hoffmann

## Summary of Facts and Submissions

- I. This appeal lies from the decision of the examining division refusing European patent application No. 99 402 595.5 with an amended claim 1 according to the main request reading as follows:

*"1. A positive active material for rechargeable lithium batteries, the positive active material comprising:*

*an active material component processed from a manganese-based compound, the manganese-based compound being selected from the group consisting of  $Li_xMnO_2$ ,  $Li_xMnF_2$ ,  $Li_xMnS_2$ ,  $Li_xMnO_{2-z}F_z$ ,  $Li_xMnO_{2-z}S_z$ ,  $Li_xMn_{1-y}M_yO_2$ ,  $Li_xMn_{1-y}M_yF_2$ ,  $Li_xMn_{1-y}M_yS_2$ ,  $Li_xMn_{1-y}M_yO_{2-z}F_z$ ,  $Li_xMn_{1-y}M_yO_{2-z}S_z$ ,  $Li_xMn_2O_4$ ,  $Li_xMn_2F_4$ ,  $Li_xMn_2S_4$ ,  $Li_xMn_2O_{4-z}F_z$ ,  $Li_xMn_2O_{4-z}S_z$ ,  $Li_xMn_{2-y}M_yO_4$ ,  $Li_xMn_{2-y}M_yF_4$ ,  $Li_xMn_{2-y}M_yS_4$ ,  $Li_xMn_{2-y}M_yO_{4-z}F_z$ , and  $Li_xMn_{2-y}M_yO_{4-z}S_z$  where  $0 < x \leq 1.5$ ,  $0.05 \leq y \leq 0.3$ ,  $z \leq 1.0$  and  $M$  is selected from the group consisting of Al, Co, Cr, Mg, Fe, La, Sr and Ce; and*

*a vanadium pentoxide ( $V_2O_5$ ) coated on the active material component, wherein the vanadium pentoxide has a thickness ranged from 1 to 100 nm."*

- II. In the contested decision, the examining division held the subject-matter of above claim 1 to lack novelty in the light of document

D2: Patent Abstracts of Japan, vol. 011, no. 338 (E-553) & JP 62-119 867 and its translation in English.

- III. During the examination procedure, reference was also made to document:

D1: Patent Abstracts of Japan, vol. 1998, no. 03 &  
JP 09-293 508.

- IV. With the grounds of appeal dated 28 February 2007, the appellant contested the above decision and filed three further sets of claims as 1<sup>st</sup>, 2<sup>nd</sup> and 3<sup>rd</sup> auxiliary requests, respectively.
- V. In a communication, the board informed the appellant that it accepted the examining division's arguments and that claim 1 of the main request then on file appeared to lack novelty over the disclosure in document D2.
- VI. With its letter dated 12 May 2010, the appellant declared that it was not maintaining the main request and that the first auxiliary request was to become the new main request, with claim 1 reading as follows:

*"1. A positive active material for rechargeable lithium batteries, the positive active material comprising:  
an active material component processed from a manganese-based compound, the manganese-based compound being selected from the group consisting of  $\text{Li}_x\text{MnO}_2$ ,  $\text{Li}_x\text{MnF}_2$ ,  $\text{Li}_x\text{MnS}_2$ ,  $\text{Li}_x\text{MnO}_{2-z}\text{F}_z$ ,  $\text{Li}_x\text{MnO}_{2-z}\text{S}_z$ ,  $\text{Li}_x\text{Mn}_{1-y}\text{M}_y\text{O}_2$ ,  $\text{Li}_x\text{Mn}_{1-y}\text{M}_y\text{F}_2$ ,  $\text{Li}_x\text{Mn}_{1-y}\text{M}_y\text{S}_2$ ,  $\text{Li}_x\text{Mn}_{1-y}\text{M}_y\text{O}_{2-z}\text{F}_z$ ,  $\text{Li}_x\text{Mn}_{1-y}\text{M}_y\text{O}_{2-z}\text{S}_z$ ,  $\text{Li}_x\text{Mn}_2\text{O}_4$ ,  $\text{Li}_x\text{Mn}_2\text{F}_4$ ,  $\text{Li}_x\text{Mn}_2\text{S}_4$ ,  $\text{Li}_x\text{Mn}_2\text{O}_{4-z}\text{F}_z$ ,  $\text{Li}_x\text{Mn}_2\text{O}_{4-z}\text{S}_z$ ,  $\text{Li}_x\text{Mn}_{2-y}\text{M}_y\text{O}_4$ ,  $\text{Li}_x\text{Mn}_{2-y}\text{M}_y\text{F}_4$ ,  $\text{Li}_x\text{Mn}_{2-y}\text{M}_y\text{S}_4$ ,  $\text{Li}_x\text{Mn}_{2-y}\text{M}_y\text{O}_{4-z}\text{F}_z$ , and  $\text{Li}_x\text{Mn}_{2-y}\text{M}_y\text{O}_{4-z}\text{S}_z$  where  $0 < x \leq 1.5$ ,  $0.05 \leq y \leq 0.3$ ,  $z \leq 1.0$  and M is selected from the group consisting of Al, Co, Cr, Mg, Fe, La, Sr and Ce; and  
a vanadium pentoxide ( $\text{V}_2\text{O}_5$ ) coated on the active material component, wherein the vanadium pentoxide has a thickness ranged from 1 to 100 nm, and wherein the*

positive active material for rechargeable lithium batteries is prepared by a method comprising the steps of:

obtaining a powder from a source material, the source material being selected from the group consisting of  $Li_xMnO_2$ ,  $Li_xMnF_2$ ,  $Li_xMnS_2$ ,  $Li_xMnO_{2-z}F_z$ ,  $Li_xMnO_{2-z}S_z$ ,  $Li_xMn_{1-y}M_yO_2$ ,  $Li_xMn_{1-y}M_yF_2$ ,  $Li_xMn_{1-y}M_yS_2$ ,  $Li_xMn_{1-y}M_yO_{2-z}F_z$ ,  $Li_xMn_{1-y}M_yO_{2-z}S_z$ ,  $Li_xMn_2O_4$ ,  $Li_xMn_2F_4$ ,  $Li_xMn_2S_4$ ,  $Li_xMn_2O_{4-z}F_z$ ,  $Li_xMn_2O_{4-z}S_z$ ,  $Li_xMn_{2-y}M_yO_4$ ,  $Li_xMn_{2-y}M_yF_4$ ,  $Li_xMn_{2-y}M_yS_4$ ,  $Li_xMn_{2-y}M_yO_{4-z}F_z$ , and  $Li_xMn_{2-y}M_yO_{4-z}S_z$  where  $0 < x \leq 1.5$ ,  $0.05 \leq y \leq 0.3$ ,  $z \leq 1.0$  and M is selected from the group consisting of Al, Co, Cr, Mg, Fe, La, Sr and Ce;

coating the powder with a vanadium pentoxide aqueous solution or a vanadium pentoxide alcoholic solution, wherein the vanadium pentoxide solution or the vanadium pentoxide alcoholic solution contains a 0.1 to 30 weight percent of vanadium component, to make an vanadium pentoxide solution-coated powder; and

heat-treating the vanadium pentoxide solution-coated powder to prepare a vanadium-coated active material."

- VII. The appellant requested that the decision under appeal be set aside and that the case be granted on the basis of the set of claims submitted as the main request with the letter of 12 May 2010.

## Reasons for the Decision

### 1. *Main request - Amendments*

Amended claim 1 of this request results from the combination of claims 1, 2, 3 and 4 as originally filed.

Amended claim 2 corresponds to claim 5 as originally filed.

Amended claim 3 has its origin in the sentence bridging pages 4 and 5 of the application as filed.

So, the claims of this request have a basis in the application as filed and they meet the requirements of Article 123(2) EPC.

### 2. *Main request - Novelty*

- 2.1 Document D2 discloses a method for preparing a positive active material for a battery including a negative electrode and a positive active material, which uses a light metal as an active material, and an organic electrolyte, the method comprising the steps of forming a vanadium pentoxide thin film on the surface of the positive active material by impregnating the material with a vanadium pentoxide aqueous solution, drying the solution, and performing heat treatment (page 1, lines 5 to 10 of the translation).

In the lithium-manganese dioxide-based battery exemplified in D2 (page 3, line 19 to page 4, line 17 of the translation), the positive active material is an electrolytic manganese dioxide.

2.2 The examining division argued that in the battery system according to D2, the vanadium layer had necessarily to be permeable to lithium ions, and so lithium would diffuse into the core of manganese dioxide during operation of the battery. The formation of a  $\text{Li}_x\text{MnO}_2$  "compound" is thus inevitable in D2, and therefore the subject-matter of claim 1 of the main request then on file was no longer novel.

2.3 The board observes that the amended claim 1 of the main request now at issue requires that the active material component to be coated by vanadium pentoxide be a powder from a source material consisting of a lithium-manganese compound having a specific formula. The word "compound" used in claim 1 and the wording used for said claim - drafted in "product-by-process" terms - make clear that the core of the coated powder consists of a material with a specifically defined and substantially homogeneous composition. So, a manganese dioxide such as would be obtained during operation of the battery disclosed in document D2, i.e. a manganese exhibiting a lithium diffusion layer with a gradient of concentration of lithium, is implicitly excluded.

Claim 1 (and claims 2 and 3, which depend on claim 1) therefore meet the requirements of Article 54(1) and (2) EPC.

3. *Main request - Inventive step*

3.1 The present application concerns a manganese-based positive active material for rechargeable lithium batteries (claim 1 and page 1, first lines).

3.2 As D2 relates also to active materials for a positive battery electrode, the board does not see any reason to depart from the opinion of both the examining division and the appellant that this document is to be taken as the closest state of the art.

3.3 The next step in assessing inventive step is to determine the problem to be solved in the light of D2. According to the application in suit, it is an object of the present invention to provide a manganese-based positive material for rechargeable lithium batteries which exhibits a good cycle life characteristic at high temperatures (page 2, lines 3 to 5)

The application as filed states that "*it is **presumed** (emphasis added by the board) that the good cycling life characteristic of the cell is resulted because the vanadium pentoxide layer coated on the surface of the manganese-based active material component prevents elution of manganese*" (page 8, line 21 seq.). The board notes, however, that no evidence for the elution of manganese has been provided. So, this problem cannot be taken into consideration in the assessment of the problem to be solved.

But Figures 1 to 3 of the application give evidence that rechargeable lithium batteries manufactured with the positive active material claimed have **good cycle life characteristics**. No direct comparison between the materials of D2 and those claimed has been provided, however, so an improvement vis-à-vis D2 cannot be recognised in the claimed subject-matter.



- 3.4 Under these circumstances, the problem to be solved in view of D2 has to be reformulated in less ambitious terms, namely as the provision of an **alternative** positive electrode material for rechargeable lithium batteries having good cycling life characteristics at high temperatures.
- 3.5 As a solution to this problem, the application proposes the material according to claim 1 at issue, characterised in particular by the provision of a powder from a source material selected from the group of compounds defined in claim 1, said powder being coated with a vanadium pentoxide according to the process defined in the said claim.
- 3.6 It is now necessary to assess whether the problem underlying the invention has been solved. Examples 1 to 4 provide evidence that positive electrodes can be prepared with different source powders ( $\text{LiMnO}_4$  and  $\text{Li}_x\text{Mn}_{2-y}\text{Al}_y\text{O}_{4-z}\text{F}_z$ ) and different heat treatments ( $200^\circ\text{C}$  and  $600^\circ\text{C}$ ). As shown in Figure 3, a cell using the positive electrode from Example 1 exhibits a far better cycle life characteristic than the same cell without a vanadium pentoxide coating on the active material of the positive electrode (page 8, lines 17 to 21). The application (page 9, lines 1 to 3) further indicates that the cells fabricated with the electrodes according to Examples 2 to 4 also exhibit the desired performance characteristic similar to that of Example 1.

Nor does the board see any reason to expect that the other unspecified lithium-manganese-based compounds would not exhibit a good cycle life characteristic. After all, like the compounds  $\text{LiMnO}_4$  and  $\text{Li}_x\text{Mn}_{2-y}\text{Al}_y\text{O}_{4-z}\text{F}_z$

specified in Examples 1 to 4, they all include lithium and manganese in the starting source material before coating with the vanadium pentoxide, contrary to the active material of document D2.

The board is therefore satisfied that the problem is solved over the whole scope of claim 1.

4. It remains to be decided whether the proposed solution to the above problem is obvious or not in view of the known state of the art.

4.1 In this connection, the board notes that the two documents disclose different positive active materials, namely a lithium-**nickel** based composite oxide coated with vanadium pentoxide (D1), and a **manganese oxide** coated with vanadium pentoxide (D2).

4.2 Since none of these documents discloses or suggests that lithium-manganese-based compounds - such as those listed in claim 1 at issue - would be suitable as positive electrode material for a rechargeable lithium battery exhibiting good cycle life characteristics, the skilled person faced with the problem indicated in item 3.4 above is not prompted to arrive at the subject-matter of claim 1 at issue in the light of D1 and/or D2.

For these reasons, it is concluded that, having regard to the state of the art, the subject-matter of claim 1 of this request (and of the claims 2 and 3, which both depend on claim 1) is not obvious to a person skilled in the art. Therefore, the requirements of Article 56 EPC are fulfilled.

**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 following documents:
  - Claims 1 to 3 according to the main request filed with letter of 12 May 2010;
  
  - Pages 1 to 9 of the description filed with letter of 12 May 2010;
  
  - Pages 1/4 to 4/4 of Figures as originally filed.

The Registrar:

The Chairman:

C. Vodz

G. Raths