

Internal distribution code:

- (A) [-] Publication in OJ
(B) [-] To Chairmen and Members
(C) [-] To Chairmen
(D) [X] No distribution

**Datasheet for the decision
of 12 November 2015**

Case Number: T 1319/13 - 3.3.05

Application Number: 01123894.6

Publication Number: 1195827

IPC: H01M4/58, H01M10/0565

Language of the proceedings: EN

Title of invention:

Method for producing cathode active material and method for producing a non-aqueous electrolyte cell

Patent Proprietor:

Sony Corporation

Opponent:

The Dow Chemical Company

Headword:

Cathode material/SONY

Relevant legal provisions:

EPC Art. 114(2), 54(1), 54(3), 56, 83
RPBA Art. 13(1), 13(3)

Keyword:

Late-submitted material - correct exercise of discretion (no)
Late-filed request - admitted (yes)
Sufficiency of disclosure (yes)
Disclaimer - auxiliary request 2 (accepted)
Novelty - auxiliary request 2 (yes)
Inventive step - auxiliary request 2 (yes)

Decisions cited:

G 0002/10, T 0440/04, T 2130/11

Catchword:



**Beschwerdekammern
Boards of Appeal
Chambres de recours**

European Patent Office
D-80298 MUNICH
GERMANY
Tel. +49 (0) 89 2399-0
Fax +49 (0) 89 2399-4465

Case Number: T 1319/13 - 3.3.05

D E C I S I O N
of Technical Board of Appeal 3.3.05
of 12 November 2015

Appellant: The Dow Chemical Company
(Opponent) 2030 Dow Center, Abbott Road
Midland, Michigan 48640 (US)

Representative: Beck Greener
Fulwood House
12 Fulwood Place
London WC1V 6HR (GB)

Respondent: Sony Corporation
(Patent Proprietor) 7-35, Kitashinagawa 6-chome
Shinagawa-ku
Tokyo (JP)

Representative: Müller Hoffmann & Partner
Patentanwälte mbB
St.-Martin-Strasse 58
81541 München (DE)

Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted on 12 February 2013 rejecting the opposition filed against European patent No. 1195827 pursuant to Article 101(2) EPC.**

Composition of the Board:

Chairman G. Glod
Members: J.-M. Schwaller
C. Vallet

Summary of Facts and Submissions

- I. The present appeal lies from the decision of the opposition division to reject the opposition against European patent No. 1 195 827, independent claims 1 and 5 of which read as follows:

"1. A method for preparing a cathode active material comprising:

a mixing step of mixing starting materials for synthesis of a compound represented by a general formula $Li_xFe_{1-y}M_yPO_4$, where M is at least one selected from a group consisting of Mn, Cr, Co, Cu, Ni, V, Mo, Ti, Zn, Al, Ga, Mg, B and Nb, with $0.05 \leq x \leq 1.2$ and $0 \leq y \leq 0.8$;

a milling step of milling a mixture obtained in said mixing step;

a compressing step of compressing the mixture milled in said milling step to a preset density; and

a sintering step of sintering the mixture compressed in said compressing step, wherein a carbon material is added in any of the above steps previous to said sintering step, and wherein the preset density of said mixture in said compressing step is not less than 1.71 g/cm^3 and not larger than 2.45 g/cm^3 ."

"5. A method for preparing a cathode active material comprising:

a mixing step of mixing starting materials for synthesis of a compound represented by a general formula $Li_xFe_{1-y}M_yPO_4$, where M is at least one selected from a group consisting of Mn, Cr, Co, Cu, Ni, V, Mo, Ti, Zn, Al, Ga, Mg, B and Nb, with $0.05 \leq x \leq 1.2$ and $0 \leq y \leq 0.8$;

a milling step of milling a mixture obtained in said mixing step; and

a sintering step of sintering the mixture milled in said milling step, wherein a carbon material is added in any of the above steps, and wherein, after said milling step, a tap density of the starting materials for synthesis is not less than 0.4 g/cc and not larger than 2.0 g/cc."

II. The following documents cited in the opposition proceedings are relevant for the present decision:

D1: JP 9 134725 and its English translation

D6: JP 9 171827 and its English translation

D8: EP 1 193 785 A2

D9: EP 1 193 784 A2

D10: EP 1 184 920 A2

R3: WO 01/53198 A1

R12: Experimental report - December 2012.

III. According to the contested decision, the late-filed documents R3 and R12 were not admitted into the proceedings. R3 was *prima facie* not more relevant than the other documents already in the proceedings. R12 was inconsistent because, on the one hand, on page 5 it was stated that tap densities greater than about 1.3 were not achievable but, on the other hand, tap densities of 1.31, 1.32, 1.34 and 1.35 were reported in the table on page 3.

The requirements of Article 83 EPC were met, as the patent disclosed at least one way to carry out the

invention and a method to measure the parameter "tap density".

The claimed subject-matter was novel over D8, since it disclosed neither the compression step required in claim 1, nor the milling step to a tap density as defined in claim 5.

As to inventive step, starting from D1 as the closest state of the art, the problem underlying the invention was seen in the provision of an improved cathode active material. As none of the known prior-art documents disclosed that working within the density ranges defined in claims 1 and 5 led to improved cathode active material, the claimed subject-matter involved an inventive step.

The same conclusions arose starting from document D6 as the closest state of the art.

- IV. With its grounds of appeal dated 19 June 2013, the opponent ("the appellant") requested the board to admit documents R3 and R12 into the appeal proceedings. Further it objected to under Articles 100(a) and (b) EPC claims 1 and 5 as granted. In particular, it argued with respect to claim 5 that a tap density greater than 1.35 g/cm^3 could not be achieved by merely milling a mixture of lithium phosphate, iron phosphate and carbon black. In its view, claim 5 furthermore lacked novelty over D8, example 1 and the subject-matter of claims 1 and 5 lacked inventive step over D1.
- V. By letter dated 23 October 2013, the patentee ("the respondent") filed three sets of amended claims as auxiliary requests 1 to 3.

VI. At the oral proceedings, which took place on 12 November 2015, the novelty of the main request over document D8 was discussed, especially in the light of document R12 which was admitted into the proceedings. In replacement of the auxiliary requests then on file, the respondent filed two new auxiliary requests 1 and 2, which were admitted into the proceedings.

Auxiliary request 1 differs from the main request by the introduction in claims 1 and 5 of the disclaimer "*excluding the compound LiFePO_4* ".

Auxiliary request 2 differs from the main request by the introduction in claims 1 and 5 of the disclaimer "*excluding the compound Li_xFePO_4 where $0 < x \leq 1$* ", and by the deletion of claims 6 and 8.

VII. Closing the debate, the chairman established the appellant's request to be that the decision under appeal be set aside and that the patent be revoked.

The respondent requested that the appeal be dismissed or, alternatively, that the patent be maintained on the basis of auxiliary requests 1 or 2 filed during the oral proceedings.

Reasons for the Decision

1. Admissibility of documents R3 and R12

1.1 Documents R3 and R12 were filed late (one month before the oral proceedings), and the opposition division exercised its discretion pursuant to Article 114(2) EPC by deciding to disregard them.

Since the appellant requested that these documents be admitted into the appeal proceedings, the board has to assess whether the opposition division correctly exercised its discretionary power.

Document R3 discloses (page 7, line 22 to page 9, line 6) a method for preparing compounds falling under the formula defined in claim 1 as granted, the method comprising a step of mixing the starting materials with carbon (page 8, lines 9 to 11), a dry-milling step (page 9, lines 3 to 5), a compressing step ("the mixed powders are pressed into pellets", page 9, lines 5 and 6) and a sintering step (page 8, line 30 to page 9, line 1). Although R3 does not disclose any density, it appears to be *prima facie* more relevant than documents D1, D6 or D8, at least for the preparation of compounds including the elements Mn, Cr, Co, Cu, Ni, V, Ti, Zn or Mg, which are not mentioned in D1, D6 or D8. R3 is therefore admitted under Article 114(2) EPC into the appeal proceedings.

With respect to R12, the board is of the opinion that the alleged "inconsistency" ("about 1.30 g/cc" vs. "1.35 g/cc" for the upper limit of the tap density achievable by milling) is not reason enough to decline to admit this experimental report into the proceedings, because the two values are so close to one another that the experimental results cannot simply be ignored for this reason alone. Furthermore, both values are so far away from the disputed upper limit value of 2.0 g/cc, of the range defined in claim 5 as granted that one cannot disregard the teaching of R12 that under certain conditions tap densities greater than 1.35 g/cc are hardly achievable by milling a mixture of lithium phosphate, iron phosphate and carbon black. It follows

that document R12 is admitted under Article 114(2) EPC into the appeal proceedings.

2. Main request - Novelty

2.1 It is noted that document D8 is a prior-art document under Article 54(3) EPC.

2.2 D8, paragraph [0015], discloses a method for preparing a cathode active material including **mixing, milling and sintering** a starting material for synthesis of a compound of general formula Li_xFePO_4 , with $0 < x \leq 1$, **adding a carbon material** to the resulting mass at an optional time point in the course of the mixing, milling and sintering steps, and employing Li_3PO_4 , $\text{Fe}_3(\text{PO}_4)_2$ or hydrates $\text{Fe}_3(\text{PO}_4)_2 \cdot n\text{H}_2\text{O}$ thereof, n being the number of hydrates, as the starting material for synthesis of the Li_xFePO_4 .

D8, paragraph [0037], further discloses that the materials for synthesis of the above compound are milled to such an extent that the powder density is **at least 2.2 g/cm³**, so as to achieve a cathode active material having a higher charging ratio and a high capacity.

D8, paragraph [0038], also discloses that if the powder density of the LiFePO_4 carbon composite material is less than 2.2 g/cm³, the LiFePO_4 carbon composite material is **not compressed sufficiently**.

This means that there is a direct and unambiguous disclosure that a compressing step to a density of at least 2.2 g/cm³ is necessary, in particular after the milling step, to obtain the Li_xFePO_4 carbon composite

material according to D8.

2.3 Since the value 2.2 g/cm^3 disclosed in D8 falls within the range "*not less than 1.71 g/cm^3 and not larger than 2.45 g/cm^3* " defined in claim 1 of the patent, it follows that D8 directly and unambiguously anticipates the **subject-matter of claim 1 as granted**, which, therefore, does not meet the requirements of Article 54(1) and (3) EPC.

2.4 For the board, the subject-matter of claim 5 as granted also lacks novelty, in particular over D8, example 1, which discloses the preparation of a LiFePO_4 carbon composite material under operating conditions which are extremely similar to those used in Example B-2 of the patent.

In fact, example 1 of D8 makes use of a different carbon starting material as example B-2 of the patent, and the amount of carbon used in the preparation of said LiFePO_4 carbon composite is not indicated. Further, example 1 of D8 does not indicate the tap density of the milled mixture of starting materials.

Notwithstanding these missing or different features, the board has no doubt that the LiFePO_4 carbon material produced in example 1 of D8 anticipates the material defined in claim 5 at issue. The initial discharge capacity of the cell is directly linked to the tap density (see Table B-1 of the patent). The initial discharge capacity of the cell manufactured with said LiFePO_4 carbon material is higher (161 mAh/g) than the one (153.3 mAh/g) of the cell manufactured with the cathode active material of example B-2 of the patent, which means that the material produced in example 1 of D8 inevitably has a tap density after milling, falling

within the range defined in claim 1 at issue, which therefore lacks novelty under Article 54(1) and (3) EPC.

2.5 As a consequence, the main request is not allowed.

3. Admissibility of auxiliary requests 1 and 2

Although these requests were only filed during the oral proceedings before the board, they are admitted into the proceedings.

At the oral proceedings, the respondent was confronted for the first time with the argument that D8 disclosed a compression step, so the filing of these requests at this advanced stage of the proceedings is to be seen as a legitimate attempt to overcome the novelty objection based on this particular argument with respect to claim 1. The amendment to claim 1 of auxiliary request 1 is a disclaimer that was already proposed as an amendment to independent claim 5 in an auxiliary request (then auxiliary request 2) filed with the reply to the grounds of appeal and had also been presented in an auxiliary request filed before the opposition division. The disclaimer introduced in auxiliary request 2 is broader in scope and has the same goal as the disclaimer of auxiliary request 1. Both requests are clearly converging. Since amendment by disclaimer was already known to the appellant from the proceedings before the opposition division, the amendment proposed in auxiliary request 2 cannot take him by surprise.

Furthermore, the appellant argued that the amendment would lead to new problems, without however what these might be. The board considers that the amendments made do not lead to increased complexity and that it can

deal with them without adjourning the oral proceedings. The same can be expected of the appellant.

Therefore, the board does not see any reason for not exercising its discretion under Articles 13(1) and 13(3) RPBA to admit these requests into the proceedings.

4. Auxiliary request 1 - Novelty

Claim 1 of this request differs from claim 1 of the main request by the introduction of the disclaimer "*excluding the compound LiFePO_4* ".

The above disclaimer does not establish novelty over D8, since this document discloses the preparation not only of the single compound disclaimed from claim 1 at issue, but also of the family of compounds defined by the general formula Li_xFePO_4 (with $0 < x \leq 1$) (see D8, paragraph 28). So even if claim 1 at issue aims at disclaiming the compound with $x = 1$, the compounds with $0 < x < 1$ still fall under the wording of the general formula $\text{Li}_x\text{Fe}_{1-y}\text{M}_y\text{PO}_4$ defined in claim 1.

In the appellant's view, the disclaimer should be allowed because D8 disclosed only the preparation of the specific compound LiFePO_4 .

The board cannot accept this argument, because it is established case law that the technical disclosure in a document must be considered as a whole, as it is not confined to the examples disclaimed (T 440/04, Reasons 4.3.2). In the present case, the disclosure of D8 being not limited to the single compound disclaimed from the claimed subject-matter, the reasons in point 1.1. to 1.2 apply *mutatis mutandis* for the preparation of the compounds Li_xFePO_4 , with $0 < x < 1$, which are

covered by claim 1 at issue, and which thus lacks novelty under Article 54(1) and (3) EPC.

5. Auxiliary request 2

5.1 Amendments

Claims 1 and 5 of auxiliary request 2 differ from the main request by the introduction of the disclaimer "*excluding the compound Li_xFePO_4 where $0 < x \leq 1$* ".

The exclusion from the compounds to be prepared of Li_xFePO_4 where $0 < x \leq 1$ amounts to the deletion of a disclosed embodiment (see claim 7) from a generic class, and therefore results in the limitation of the cathode active materials. This means that examples A-1 to A-7, B-1 to B-4 and B-19 no longer fall under the scope of the claims, while the rest of the examples still do. Therefore, the subject-matter remaining in the claim is considered directly and unambiguously disclosed to the skilled person in the application as filed. In accordance with G 2/10, Reasons 4.5.4, this disclaimer is allowable. The case is similar to that underlying T 2130/11 (reasons 4.4).

5.2 Disclosure of the invention

The objection of the appellant only related to claim 5.

5.2.1 It is established case law that the requirements for sufficiency of disclosure are met if the claimed invention could be performed at the filing date of the application by a person skilled in the art in the whole area claimed without undue burden, using common general knowledge and having regard to further information given in the patent in suit. When the definition of the

claimed invention moreover includes one or more parameters - in the present case the tap density - the skilled person should also be able to check whether the parameter is complied with while the invention is being carried out.

- 5.2.2 In the case at issue, the measurement of the tap density is described at paragraphs [0192] to [0200], so the latter criterion is met.

The patent specification furthermore discloses (paragraphs [0190] to [0201] and [0222] to [0238]) ample details and examples regarding the production of the carbon composite material defined in claim 5. So there is no doubt that the invention could be performed at the filing date of the application by a person skilled in the art in the whole area claimed without undue burden.

Regarding the appellant's argument that tap density greater than 1.35 g/cm^3 could not be achieved by merely milling a mixture of lithium phosphate, iron phosphate and carbon black, the board notes that this assertion was based on the results of the tests carried out in R12. However, these tests no longer concern compounds according to claim 5, and the tap density measurements in the patent have been carried out using a different apparatus - the Ishiyama volume tester illustrated in Figures 4 to 6 - and a different testing procedure (paragraphs [0192] to [0199]) from the one used in R12. Therefore, the appellant's argument is not relevant for the scope of claim 5 and cannot be accepted.

- 5.2.3 The burden of proof is upon the opponent (here the appellant) to show that a skilled person was unable to carry out the invention. In the present case the

appellant did not identify any information gap for the subject-matter of claim 5. The board has thus no reason to doubt that the invention as defined in claim 5 meets the requirements of Article 83 EPC.

5.3 Novelty

5.3.1 D8 is no longer relevant, since it only relates to a compound represented by the general formula Li_xFePO_4 where $0 < x \leq 1$, which is now excluded from the scope of claims 1 and 5. The same applies to D9 and D10 that were also cited in the appellant's novelty objections.

5.3.2 Document R3 (claims 1, 2 and 3) discloses a method of making a lithium mixed-metal compound by mixing particles of starting materials comprising:

- a compound of one or more metals selected from the group consisting of Fe, Co, Ni, Mn, Cu, V, Sn, Ti, Cr,
- a lithium compound selected from the group consisting of lithium carbonate, lithium phosphate, lithium oxide, lithium vanadate, and
- carbon in an amount sufficient to reduce the oxidation state of at least one metal ion of said starting materials without full reduction to an elemental state; and

heating said starting materials in a non-oxidising atmosphere at a temperature sufficient to form a reaction product comprising lithium and said reduced metal.

According to claims 5 and 6, the starting materials can include a second metal selected from the group consisting of Mg, Ca, Zn, Sr, Pb, Cd, Sn, Ba, Be, and mixtures thereof.

In example I (page 33), LiFePO_4 is produced by ball-milling 1 mol FePO_4 , 0.5 mol Li_2CO_3 , 0.5 mol carbon (+ 100% excess carbon), pelletising the powder mixture, and heating the pellet to 750°C in flowing argon for 8 hours.

In examples IV and V (pages 36 and 37) $\text{LiFe}_{0.9}\text{MgO}_{0.1}\text{PO}_4$ is produced in a similar way, without however any indication of ball-milling.

For the board, the fact that the ball-milled mixture is pelletised implicitly involves a compressing step.

- 5.3.3 R3 however does not indicate the pressure applied to the milled powder mixture, nor does it indicate the density of the mixture after the compressing step. It follows that claim 1 of this request is novel over R3.
- 5.3.4 R3 does not directly and unambiguously disclose the subject-matter of claim 5 of this request either, because there is no evidence that the tap density obtained after milling would inevitably fall within the claimed range of densities. The appellant's argument - based on R12 - that the tap density would inevitably fall within the range defined in claim 5 at issue is not convincing, because R12 only relates to LiFePO_4 that is no longer part of claim 5, and since none of the examples of R3 have been reproduced in R12, it cannot be directly and unambiguously concluded that the examples of R3 were novelty-destroying.
- 5.3.5 Hence, the invention as defined in the subject-matter claimed in this request lacks novelty meets the requirements of Article 54(1) and (3) EPC.

5.4 Inventive step

Applying the problem-solution approach, the board came to the conclusion that the subject-matter of claims 1 to 8 of this request involves an inventive step for the following reasons:

- 5.4.1 The invention as defined in the claims relates to methods for preparing a lithium-, iron- and phosphate-containing cathode material.
- 5.4.2 The closest state of the art to this invention is document D1, which the appellant held at the oral proceedings to be the best starting point for assessing the inventive step of the claimed subject-matter. R3 incidentally cannot be taken as the closest state of the art, because it is prior art under Article 54(3) EPC.

D1 (claim 1) discloses a non-aqueous electrolyte battery containing an alkali metal-containing iron complex oxide of the general formula A_yFeXO_4 (with A being an alkali metal, X an element of groups VI-VII of the periodic table, and $0 < y < 2$) as the cathode active substance. D1 further discloses (page 4, lines 4 and 5) that X can be V, P, As, Sb, Bi or the like. D1 also discloses the possibility of adding carbon powder in the preparation process.

In its preferred embodiments (examples and Table 1), D1 discloses the preparation of $LiFePO_4$, $LiFeVO_4$ and $LiFeV_{0.5}P_{0.5}O_4$. These compounds are prepared by mixing the starting materials (lithium carbonate, iron oxalate and/or ammonium vanadate and diammonium phosphate) - without any addition of carbon powder - and sintering for several days at $800^\circ C$ in a nitrogen stream.

D1 does not disclose a single one of the compounds defined in the set of claims at issue, nor does it disclose any milling or compressing step, or any density after compression or tap density after milling.

5.4.3 The contested patent (paragraph [0013] states that the problem underlying the invention is to provide a method for the preparation of a cathode material having high cell capacity.

5.4.4 As a solution to this problem, the patent proposes two alternatives:

- claim 1, which proposes a method for preparing a lithium iron phosphate compound characterised in particular in that the compound contains a further metal selected from the group consisting of Mn, Cr, Co, Cu, Ni, V, Mo, Ti, Zn, Al, Ga, Mg, B and Nb, and in that the preparation process includes a step of milling the starting materials and a step of compressing the thus obtained mixture to a preset density of from 1.71 g/cm^3 to 2.45 g/cm^3 ;
- claim 5, which proposes a method for preparing a lithium iron phosphate compound characterised in particular in that the compound contains a further metal selected from the group consisting of Mn, Cr, Co, Cu, Ni, V, Mo, Ti, Zn, Al, Ga, Mg, B and Nb, and in that the preparation process includes a step of milling the starting materials up to a tap density of from 0.4 g/cc to 2.0 g/cc.

5.4.5 As to whether the problem identified in point 5.4.3 above has indeed be solved by either of the above alternative solutions, the board observes that there is no evidence for any improvement over the cathode

material of D1, and so it is necessary to reformulate the problem as the provision of an alternative cathode material.

- 5.4.6 As to the obviousness of the solutions proposed, the board notes that none of the documents in the proceedings discloses the preparation of a lithium iron phosphate cathode material containing a metal selected from the group consisting of Mn, Cr, Co, Cu, Ni, V, Mo, Ti, Zn, Al, Ga, Mg, B and Nb.

The appellant's argument that D1 disclosed such a compound with the additional metal being vanadium fails, because vanadium replaces the phosphor atom, not the iron atom as required by the formula in claims 1 and 5 at issue.

For that reason already, the solution proposed in independent claims 1 and 5 can be considered not obvious, since none of the prior-art documents discloses the possibility of using a lithium iron phosphate according to the formula defined in claims 1 or 5 as an alternative cathode material.

Moreover, none of the prior-art documents suggests milling and compressing the starting materials defined in claim 1 to a preset density of from 1.71 g/cm^3 to 2.45 g/cm^3 or, alternatively, milling the starting materials defined in claim 5 up to a tap density of from 0.4 g/cc to 2.0 g/cc , with the aim of producing an active lithium iron phosphate cathode material.

It follows that, having regard to the state of the art, the subject-matter of independent claims 1 and 5 at issue is not obvious to a person skilled in the art,

and therefore involves an inventive step within the meaning of Article 56 EPC.

Dependent claims 2 to 4 and 6 to 8 derive their patentability from claims 1 and 5, on which they depend, and so likewise meet the requirements of Article 56 EPC.

6. As the appellant has not succeeded in establishing that the set of claims of this request does not meet the requirements of the EPC, the patent is to be maintained in amended form.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the opposition division with the order to maintain the patent on the basis of claims 1 to 8 of auxiliary request 2 filed during the oral proceedings, and a description and drawings to be adapted.

The Registrar:

The Chairman:



C. Vodz

G. Glod

Decision electronically authenticated