Internal distribution code:

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

Datasheet for the decision of 26 September 2013

Case Number: T 2367/10 - 3.3.05

Application Number: 99960299.8

Publication Number: 1137598

IPC: C01G51/00, C01G1/00, H01M4/52

Language of the proceedings: EN

Title of invention:
LAYERED LITHIUM METAL OXIDES FREE OF LOCALISED CUBIC SPINEL-LIKE STRUCTURAL PHASES AND METHOD OF MAKING SAME

Patent Proprietor:
UMICORE

Opponent:
H.C. Starck GmbH

Headword:
LiCoM oxides/Umicore

Relevant legal provisions:
EPC Art. 54(1), 54(2), 56, 83, 123(2)
EPC R. 80

Keyword:
Sufficiency of disclosure - enabling disclosure (yes)
Novelty - main request (no)
Amendments - allowable (yes)
Inventive step - auxiliary request (yes) - reformulation of the problem to an alternative - solution not obvious

Decisions cited:

Catchword:

Europäisches Patentamt
European Patent Office
Office européen des brevets

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 2367/10 - 3.3.05

D E C I S I O N
of Technical Board of Appeal 3.3.05
of 26 September 2013

Appellant: (Patent Proprietor)	UMICORE Broekstraat 31 1000 Brussels (BE)
Representative:	Meissner, Bolte & Partner Anwaltssozietät GbR Widenmayerstrasse 47 80538 München (DE)
Respondent: (Opponent)	H.C. Starck GmbH Im Schleeke 78-91 D-38642 Goslar (DE)
Representative:	Woods, Geoffrey Corlett J A Kemp 14 South Square Gray's Inn London WC1R 5JJ (GB)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted on 29 September 2010 revoking European patent No. 1137598 pursuant to Article 101(3)(b) EPC.**

Composition of the Board:

Chairman: H. Engl
Members: J.-M. Schwaller
D. Prietzel-Funk

Summary of Facts and Submissions

I. The present appeal lies from the decision of the opposition division revoking European patent No. 1 137 598 on the grounds that the subject-matter of the claims 1 then on file (main and auxiliary requests) lacked novelty under Article 54(1)(2) EPC.

Claim 1 of the main request read as follows:

"*1. A compound having the formula $Li_\alpha M_\beta A_\gamma O_2$, wherein M is one or more transition metals, A is one or more dopants having an average oxidation state N such that $+2.5 \leq N \leq +3.5$, $0.90 \leq \alpha \leq 1.10$ and $\beta + \gamma = 1$, said compound having a substantially single phase, hexagonal layered crystal structure and being substantially free of localized cubic spinel-like structural phases, wherein, in the powder x-ray diffraction pattern, there are no diffraction peaks at a smaller scattering angle than the diffraction peak corresponding to Miller indices (003).*"

Claim 1 of the auxiliary request read as follows:

"*1. A compound having the formula $Li_\alpha M_\beta A_\gamma O_2$, wherein M is one or more transition metals, A is one or more dopants having an average oxidation state N such that $+2.5 \leq N \leq +3.5$, $0.90 \leq \alpha \leq 1.10$ and $\beta + \gamma = 1$, said compound having a substantially single phase, hexagonal layered crystal structure and being substantially free of localized cubic spinel-like structural phases, wherein, in the powder x-ray diffraction pattern, there are no diffraction peaks at a smaller scattering angle than the diffraction peak corresponding to Miller indices (003), wherein the ratio of the integrated intensity of the diffraction peak corresponding to*

Miller indices (110) to the integrated intensity of the diffraction peak corresponding to Miller indices (108) using powder x-ray diffraction is greater than or equal to 0.7, and wherein the ratio of the integrated intensity of the diffraction peak corresponding to Miller indices (102) to the integrated intensity of the diffraction peak corresponding to Miller indices (006) using powder x-ray diffraction is greater than or equal to 1.0."

II. Among the documents cited in the first instance proceedings, the following are still of relevance to the present decision:

D1: JP 7-112929 B2 and its English translation
D2: EP 0 646 546 A1
D5: J.N. Reimers et al., Solid State Ionics, 61 (1993), pages 335 to 344
D6: JP 7-114915 A and its English translation
D7: EP 0 918 041 A1

III. In its decision, the opposition division held claim 1 of the main request to lack novelty over documents D2, D5, D6 and D7, which individually disclosed mixed lithium transition metal oxides falling under the formula $Li_{\alpha}M_{\beta}A_{\gamma}O_2$ as defined above. The single phase hexagonal layered crystal structure and the absence of a localised cubic spinel-like phase were demonstrated by the absence, in the powder x-ray diffraction (XRD) pattern, of diffraction peaks at a smaller scattering angle than the diffraction peak corresponding to Miller indices (003).

The subject-matter of claim 1 of the auxiliary request was held to lack novelty in view of its disclosure of a mixed lithium transition metal oxide having an XRD

pattern identical to the ones shown in the opposed patent.

IV. With its statement of grounds of appeal dated 26 January 2011, the appellant (patent proprietor) filed an amended set of claims as its sole request.

V. The observations of the respondent (opponent) were received by letter dated 31 May 2011. It contested in particular the allowability of the amendments under Articles 123(2) and Rule 80 EPC as well as novelty and inventive step of the claimed subject-matter. It also argued that the invention was not disclosed in a manner sufficiently clear and complete for it to be carried out by the skilled person (Article 83 EPC).

VI. Under cover of a letter dated 29 February 2012, the appellant submitted four sets of amended claims as main and first to third auxiliary requests.

VII. A further submission of the appellant, dated 2 July 2012, contained additional arguments and an experimental study with comparative data.

VIII. At the oral proceedings before the board, which took place on 26 September 2013, the appellant submitted corrected versions of the main and auxiliary requests submitted earlier (by letter dated 29 February 2012).

Claim 1 of the main request reads as follows (differences with respect to the main request underlying the contested decision emphasised by the board):

"*1. A compound having the formula* $Li_{\alpha}M_{\beta}A_{\gamma}O_2$*, wherein M is one or more transition metals, A is one or more*

dopants having an average oxidation state N such that +2.5 ≤ N ≤ +3.5, 0.90 ≤ α ≤ 1.10 and β + γ = 1, said compound having a substantially single phase, hexagonal layered crystal structure and being substantially free of localized cubic spinel-like structural phases, wherein, in the powder x-ray diffraction pattern, there are no diffraction peaks at a smaller scattering angle than the diffraction peak corresponding to Miller indices (003), ***said compound obtained by uniformly cooling the compound from a temperature of at least about 600°C at a rate of between 8°C/min and 140°C/ min.***"

Independent claims 1 and 9 of the first auxiliary request read as follows (differences with respect to claim 1 of the main request emphasised by the board):

"*1. A compound having the formula $Li_\alpha M_\beta A_\gamma O_2$, wherein M is* ***Co****, A is one or more dopants* ***selected from Ti, Zr, Mg, Ca, Sr, Ba, Al, Ga, Si, Ge, Sn and combinations thereof*** *having an average oxidation state N such that +2.5 ≤ N ≤ +3.5, 0.90 ≤ α ≤ 1.10,* ***γ > 0*** *and β + γ = 1, said compound having a substantially single phase, hexagonal layered crystal structure and being substantially free of localized cubic spinel-like structural phases, wherein, the powder x-ray diffraction pattern, there are no diffraction peaks at a smaller scattering angle than the diffraction peak corresponding to Miller indices (003), said compound obtained by uniformly cooling the compound from a temperature of at least about 600°C at a rate of between 8°C/min and 140°C/min.*"

"*9. A method of preparing a compound according to claim 1, the method comprising the steps of providing a lithium metal oxide having the formula $Li_\alpha M_\beta A_\gamma O_2$,*

wherein M is Co, A is one or more dopants selected from Ti, Zr, Mg, Ca, Sr, Ba, Al, Ga, Si, Ge, Sn and combinations thereof having an average oxidation state N such that +2.5 ≤ N ≤ +3.5, 0.90 ≤ α ≤ 1.10, γ > 0 and β + γ = 1, at a temperature of at least about 600°C; and uniformly cooling the compound at a rate of between 8°C/min and 140°C/min."

IX. The respondent contested the novelty of the subject-matter of claim 1 of the main request having regard to the disclosure of documents D6 and D7.

Further, it raised objections against the claims of the auxiliary request under Articles 54, 56, 83 and 123(2) EPC and under Rule 80 EPC. Example C-58 of D6 destroyed the novelty of claim 1. Document D6 was identified by the respondent as representing the starting point for assessing inventive step of this claim.

X. At the end of the oral proceedings, the chairman established the parties' requests as follows:

The appellant requested that the decision under appeal be set aside and the patent be maintained in amended form on the basis of the claims of the main request, or in the alternative on the basis of the claims in accordance with the first, second or third auxiliary request, filed during the oral proceedings before the board of appeal.

The respondent requested that the appeal be dismissed.

Reasons for the Decision

1. Sufficiency of disclosure of the invention

1.1 According to Article 83 EPC and its counterpart in Article 100(b) EPC, an invention must be disclosed in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art. According to the case law of the boards of appeal, for demonstrating an insufficiency of disclosure, gaps in information and/or lack of guidance have to be ascertained.

1.2 In the case at issue, a detailed description of the preparation of two different lithium metal oxide compounds falling under the terms of claim 1 of the main request on file is provided by way of Examples 1 and 2 of the patent in suit. The compounds of these examples do not contain a dopant - as required by the claims of the auxiliary request; however, as shown by the experimental report submitted with letter of 2 July 2002, the preparation of such doped compounds can be easily realised by following the process steps defined in claims 1 and 9 of the first auxiliary request.

1.3 The respondent argued that certain process steps were missing and that the invention was therefore insufficiently disclosed. The board cannot accept this argument without further proof, the burden of which lies with the respondent. In the present case the respondent neither put forward any evidence that the claimed process would not result in the formation of the claimed compounds, nor that the compounds thus obtained would not possess the improved cycle performance as claimed in the patent. Under these circumstances, with the respondent having been unable to identify any gaps of information for the performance

of the claimed invention, the board concludes that the requirements of Article 83 EPC are satisfied for all requests on file.

2. Main request - Amendments

In the board's view, the subject-matter of claim 1 of this request finds its basis in the passage from page 7, line 17 to page 9, line 8 of the application as filed, which discloses "*a method of preparing compounds having a substantially single phase, hexagonal layered crystal structure and being substantially free of localized cubic spinel-like structural phases. In accordance with this method, a lithium metal oxide is provided having the formula* $Li_{\alpha}M_{\beta}A_{\gamma}O_2$*, wherein M is one or more transition metals, A is one or more dopants having an average oxidation state N such that* $+2.5 \leq N \leq +3.5$, $0.90 \leq \alpha \leq 1.10$ *and* $\beta + \gamma = 1$*, at a temperature of at least about 600°C, and preferably of greater than 800°C.*" [...]. *Once the lithium metal oxide is at its final preparation temperature or after previously synthesized lithium metal oxide has been reheated, the lithium metal oxide is cooled at a rate of greater than 8°C/min and preferably lower than 140°C/min. [...] It has been discovered that cooling at a rate of less than 8°C/min results in the formation of localized cubic spinel-like structural phases on the surface of the crystal or within the crystal and thus decreased electrochemical performance.*"

For the board, there is no doubt from this passage that the product obtained by the above process directly and unambiguously corresponds to the one of formula $Li_{\alpha}M_{\beta}A_{\gamma}O_2$ defined in claim 1 at issue. The board thus cannot accept the respondent's argument that the amendments in claim 1 of the main request were based on

an unallowable combination of features pertaining to different products, namely to the final and the intermediate products defined in claims 1 and 13 as originally filed, respectively.

The requirements of Article 123(2) EPC are thus met.

3. Main request - Novelty

3.1 D6 (see English translation; paragraphs [0005], [0006], [0012] and [0013]) concerns the production of compounds of formula $Li_xM_{y1}N_{y2}O_2$ wherein M is Co or Ni, N is Ni, V, Mn, Ti or Cu, y^1 is 0.6 to 1.0, y^2 is 0 to 0.4 and $y^1+y^2=1$. Said compounds find use in the positive electrode of a rechargeable lithium battery. They are formed by a first baking step at a temperature of 450°C to 800°C for 3 to 100 h followed by a second baking step for 0.5 to 50 h at a temperature of 50°C to 600°C higher than the first baking temperature, followed by cooling at a rate of 0.1 to 25°C/min after baking.

Examples C-12, C-18 and R-2 exemplify the preparation of $LiCoO_2$ at different baking temperatures comprised between 600°C and 950°C and at the respective cooling rates of 24°C/min, 12°C/min and 50°C/min.

3.2 The respondent argued that concerning the above three $LiCoO_2$ samples, neither the absence of a cubic spinel-like structural phase nor any XRD data indicating the absence of peaks at a smaller scattering angle than the diffraction peak corresponding to Miller indices (003) in the powder XRD pattern was disclosed in D6.

In the board's view, however, the absence of a disclosure in D6 of these features does not mean that the compounds did in fact contain a cubic spinel-like

structural phase; it simply indicates that no XRD powder diffraction analysis has been carried out and so no conclusion can be drawn from this absence.

3.3 The board is nevertheless convinced that the $LiCoO_2$ oxides prepared in examples C-12, C-18 and R-2 anticipate the subject-matter of claim 1 at issue, for the following reasons.

3.4 As explained in paragraphs [0012] and [0032] to [0034] of the patent in suit, for obtaining these oxides in a substantially single phase, hexagonal layered crystal structure substantially free of localised cubic spinel-like structural phases it is essential and sufficient to synthesise or heat the lithium metal oxide at a temperature of at least about 600°C and cool it at a rate of greater than 8 C/min. The appellant further stated in this respect (see grounds of appeal, point 2.3) that given the fact that the cubic spinel-like phases were energetically favoured, there was no way to avoid them in the production process, unless the controlled rapid cooling as suggested in the patent in suit was employed, thereby preventing phase transformation to these energetically favoured cubic phases. This statement is corroborated by the content of paragraph [0034] of the patent in suit which discloses that cooling at a rate of less than 8°C/min results in the formation of localised cubic spinel-like structural phases on the surface of the crystal or within the crystal and thus decreases electrochemical performance.

The lithium metal oxides of examples C-12, C-18 and R-2 were furthermore synthesised by the sequence of process steps described in the patent in suit as leading to the claimed lithium metal oxides.

For the board, it follows from the above considerations that compounds falling under the terms of claim 1 of this request were obtained in accordance with said examples of D6.

The subject-matter of claim 1 of the main request therefore lacks novelty (Article 54(1)(2) EPC).

4. First auxiliary request - Amendments

4.1 Claim 1 of this request is distinguished from claim 1 of the main request in that (emphasis added by the board)

- M is **Co**,
- A is one or more dopants selected from **Ti**, **Zr**, **Mg**, **Ca**, **Sr**, **Ba**, **Al**, **Ga**, **Si**, **Ge**, **Sn and combinations thereof**,
- γ **> 0**.

The board cannot accept the respondent's argument that the above combination of features resulted from a multiple selection because cobalt is the preferred metal M (see first two lines of page 7 and the examples). Furthermore the list of dopants as defined in present claim 1 is directly and unambiguously disclosed at page 6, lines 8 to 10. No selection is made from among the elements of this list of dopants. Obviously, when a dopant is present, then γ must be greater than 0.

The amendments to claim 1 are therefore directly and unambiguously derivable from the application as originally filed.

Amended claim 2 finds its basis in independent claim 2 of the application as filed. The respondent argued that the amendment in claim 2 was not occasioned by a ground for opposition and thus was not allowable under Rule 80 EPC. In the board's view, the present amendment which consists in the transformation of an independent claim into a dependent one, was appropriate to meet the conciseness and clarity requirements of Article 84 EPC, following the amendments to claim 1 (which were not objected to under Rule 80 EPC). Since claims being amended during an opposition procedure have to meet all the requirements of the EPC, the amendment to claim 2 was thus indirectly occasioned by a ground of opposition. It is therefore allowable under Rule 80 EPC.

4.2 The other claims have a basis as follows in the application as filed:

- claims 3 to 6: in claims 4 to 7 as filed, respectively;
- claims 7 and 8: in claims 11 and 12 as filed, respectively;
- claim 9: in claim 12; page 7, lines 17 to 25 and page 8, line 31 to page 9, line 5 as filed;
- claims 10 to 14: in claims 14, 16, 17, 19 and 20 as filed.

4.3 It is also evident that the amendments do not extend the scope of protection conferred by the claims with respect to the granted claims.

4.4 The requirements of Article 123(2) and (3) are thus met.

5. First auxiliary request - Novelty

In the board's view, none of the available documents discloses the combination of features defined in claim 1 of the first auxiliary request.

In particular, the board cannot share the respondent's argument that D6 anticipated the subject-matter of claim 1 at issue. It is true that document D6 discloses the claimed controlled cooling and the use of dopants, in particular of Ti. D6 however fails to disclose the claimed average oxidation state N of the dopant A within + 2.5 and + 3.5. In particular in example C-58, which discloses a metal mixed oxide of the formula $Li_{0.97}Co_{0.95}Ti_{0.05}$, the oxidation state of the dopant A (= Ti) is +4 and thus outside the range mentioned in claim 1.

From the above considerations, it follows that the subject-matter of claim 1 of this request, and by the same token that of claims 2 to 14 which depend on claim 1, is novel and meets the requirements of Article 54(1) (2) EPC.

6. First auxiliary request - Inventive step

By applying the problem-solution approach, the board arrives at the following conclusions.

6.1 The invention concerns particular doped lithium cobalt oxides for use as a positive electrode material for lithium and lithium-ion secondary batteries, and to a method of making such oxides (paragraph [0001] of the contested patent).

6.2 Lithium cobalt oxides are already known from document D6 which according to the respondent (and not contested

by the appellant) represented the closest state of the art to the claimed subject-matter, and which thus is to be taken as starting point for the assessment of the inventive step of claim 1 (see details of the disclosure of document D6 under point 3.1 above).

6.3 The technical problem underlying the contested patent is defined in paragraph [0007] of the patent in suit as consisting in the provision of lithium cobalt oxides having a good stability and a more consistent electrochemical performance than prior art compounds, and maintaining their structure during cycling.

6.4 As a solution to this problem, the invention proposes the lithium cobalt oxide compound according to claim 1 at issue, which is in particular characterised in that it contains one or more dopants selected from Ti, Zr, Mg, Ca, Sr, Ba, Al, Ga, Si, Ge, Sn and combinations thereof having an average oxidation state N such that $+2.5 \leq N \leq +3.5$.

6.5 As to the question whether the above-defined problem has been solved, the board observes that there is no evidence on file for an improvement over the lithium cobalt oxides known from document D6. The technical problem must therefore be reformulated as the provision of alternative lithium cobalt oxide compounds.

The experimental evidence submitted with appellant's letter dated 7 June 2012 show that such alternative compounds - with the dopant A being e.g. $Mg_{0.0025}Ti_{0.0025}$ and N being 3 - can be provided. These compounds furthermore have high cycling performance and high electrochemical performance. The appellant furthermore explained that an oxidation state between +2.5 and +3.5 provided high stability to the compounds because of

their reduced capacity to distort the hexagonal crystalline structure. These results remained uncontested and the board considers that the above-defined, less ambitious, problem has been successfully solved.

6.6 It remains to be decided whether the proposed solution is obvious in view of the other documents of the cited prior art.

The board observes that none of the known state of the art documents discloses the necessity of maintaining the oxidation state of the dopant metal between +2.5 and +3.5 with the purpose of obtaining doped lithium cobalt oxides with high cycling performance, high stability and high electrochemical performance.

The respondent argued that the solution was obvious in view of document D7 which disclosed the use of a dopant metal having an oxidation state of +3, namely Al^{+3}.

The board cannot accept this argument. Document D7 discloses a complex oxide to be used as cathode active material for rechargeable batteries, said oxide being represented by the formula $Li_yNi_{1-x}Co_{x1}M_{x2}O_2$ (with M representing at least one element selected from the group of Al, Fe, Mn and B; $0.9 \leq y \leq 1.3$; $0 < x \leq 0.5$, $0 < x_1 < 0.5$, $x_1 + x_2 = x$, [...]). It follows that notwithstanding that D7 discloses the use of Al as a dopant, it does so in a different chemical context. A combination of this teaching with that of document D6 would still not lead to the subject-matter of claim 1 at issue, because in D7 the presence of Ni **and** Co is mandatory and the **amount of Ni is higher than that of Co**, while D6 requires the presence of either Ni **or** Co.

It follows that claim 1, and by the same token claims 2 to 14, which include all the features of claim 1, meet the requirements of Article 56 EPC.

7. The claims of the first auxiliary request thus meet the requirements of the EPC.

8. As the first auxiliary request is allowable, there is no need to consider the subordinate requests.

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 first instance with the order to maintain the patent on the basis of the claims according to auxiliary request 1, submitted during the oral proceedings before the board of appeal, and a description and figures to be adapted.

The Registrar: The Chairman:

C. Vodz H. Engl

Decision electronically authenticated