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D E C I S I O N
of 5 May 2004

Case Number: T 0631/00 - 3.4.2

Application Number: 93306087.3

Publication Number: 0582448

IPC: H01M 4/52, H01M 4/48

Language of the proceedings: EN

Title of invention:
Non-aqueous battery

Patentee:
SANYO ELECTRIC CO., LTD.

Opponent:
H.C. STARCK GmbH & Co. KG

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Problem/solution approach not applied by first instance"
"New documents filed by both parties"
"Fresh case - remittal"

Decisions cited:
-

Catchword:
-

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Case Number: T 0631/00 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 5 May 2004

Appellant: (Opponent)	H.C. STARCK GmbH & Co. KG Im Schleeke 78-91 D-38642 Goslar (DE)
Representative:	Peters, Frank M., Dr. Bayer AG Bayer Chemicals Law and Patents Patents and Licensing D-51368 Leverkusen (DE)
Respondent: (Proprietor of the patent)	SANYO ELECTRIC CO., LTD. 18, 2-chome, Keihan-Hondori Moriguchi-shi, Osaka (JP)
Representative:	Allman, Peter John MARKS & CLERK Sussex House, 83-85 Mosley Street Manchester M2 3LG (GB)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted 16 May 2000 rejecting the opposition filed against European patent No. 0582448 pursuant to Article 102(2) EPC.**

Composition of the Board:

Chairman: A. G. Klein
Members: A. G. M. Maaswinkel
C. Rennie-Smith

Summary of Facts and Submissions

I. The appellants (opponents) lodged an appeal, received on 21 June 2000, against the decision of the opposition division, dispatched on 16 May 2000, to reject the opposition against the European patent No. 0 582 448. The fee for the appeal was paid on 21 June 2000. The statement setting out the grounds of appeal was received the same day.

II. Opposition had been filed against the patent as a whole on the basis of Article 100(a) EPC, and in particular on the grounds that the subject-matter of the patent was not patentable within the terms of Articles 52(1) and 56 EPC because it did not involve an inventive step. The further objection under Article 100(b) EPC raised during opposition was not maintained in the grounds of appeal. To support their objections the opponents referred to the following documents:

(D1) J. Solid State Chemistry, vol.96 (1992), pages 123 to 131, G. Dutta et al., "Chemical Synthesis and Properties of $Li_{1-\delta-x}Ni_{1+\delta}O_2$ and $Li[Ni_2]O_4$"

(D2) J. Phys.: Condens. Matter, vol. 3 (1991), pages 4721 to 4730, K. Hirota et al., "Magnetic properties of the S=1/2 antiferromagnetic triangular lattice $LiNiO_2$"

(D3) Mat. Res. Bull., vol. 20 (1985), pages 1137 to 1146, M.G.S.R. Thomas et al., "Synthesis and structural characterization of the normal spinel $Li[Ni_2]O_4$"

(D4) EP-B-0 017 400

(D5) EP-B-0 468 942.

III. During the appeal procedure the following further documents were filed by the parties:

(D6) Journal of Power Sources, vol. 32 (1990), pages 373 to 379, T. Nohma et al.: "Lithium-containing manganese dioxide (composite dimensional manganese dioxide: CDMO) as positive material for a lithium secondary battery"

(D7) "The Rietveld Method", R.A. Young ed., Oxford University Press (1993), Chapter "Introduction to the Rietveld Method", pages 1 to 13

(D8) JP-A-5-290845

(D8a) English Abstract of JP-A-5-290845 (Japanese Patent Office)

(D8b) English translation of JP-A-5-290845

(D9) "The 33rd Battery Symposium in Japan", 16 to 18 September 1992, Tokyo, paper 1A07, T. Ohzuku et al.: "An Approach to Secondary Nonaqueous Lithium Cell (I) Synthesis and Characterization of $LiNiO_2$"

(D10) Journal of the Ceramic Society of Japan, Int. Edition, vol. 100, pages 358 to 361, T. Ohzuku et al.: "Synthesis and Characterization of $LiMeO_2$ (ME=Ni, Ni/Co and Co) for 4-Volts Secondary Nonaqueous Lithium Cells"

(D11) Mat. Res. Bull., vol.25 (1990), pages 623 to 630, J. Morales et al.: "Cation distribution and chemical deintercalation of $Li_{1-x}Ni_{1+x}O_2$"

(D13) US-A-4 980 080

(D14) Solid State Ionics, vol. 80 (1995), pages 261 to 269, H. Arai et al.: "Characterization and cathode performance of $Li_{1-x}Ni_{1+x}O_2$ prepared with the excess lithium method".

Documents D6 and D7 were filed by the respondents with their letter dated 15 January 2001 in order to rebut the appellants' position in the Grounds of Appeal that X-ray peak intensity and ratio can be readily calculated from scattering cross-sections of individual atoms and the position of the atoms in the crystal lattice, and that the claimed peak ratio also implies that the $LiNiO_2$-crystal must be pure for ensuring a high discharge capacity.

Document D13, acknowledged in the patent specification to disclose a conventional $LiNiO_2$-battery, was referred to by the appellants in their letter dated 14 March 2001. According to this letter Example I and column 3, lines 1 to 5 of document D13 disclose a material composition of $LiNiO_2$ with peak intensity ratio of 0.73, and in this passage it is explained that the observed lithium deficiency of 6.35% versus the calculated value of 7.11% may be corrected by using in the preparation of the material a slight excess of lithine at the outset.

Documents D8, D8a, D8b, D9, D10 and D11 were filed with the appellants' letter dated 31 October 2001. According to this letter, patent application D8 had been filed by the same applicant with partly the same inventors as the patent in suit. Since, furthermore, one of the examples of D8 was within the scope of claim 1, the opposed patent was not entitled to the Japanese priority. In consequence, document D9 became a prepublished document and anticipated the subject-matter of claim 1. Finally, documents D10 and D11 were also highly relevant.

Document D14 was cited by the respondents in their letter of 5 April 2004. This document had been published three years after the priority date of the patent in suit and, according to the respondents, was the first publication addressing the relationship between discharge capacity and both X-ray diffraction analysis and crystal structure and the role of Ni atoms located at Li sites on the discharge capacity of $LiNiO_2$, which was a clear indication that prior to the filing of the patent in suit this relationship had not been known.

IV. On 5 May 2004 oral proceedings were held.

V. At the oral proceedings the appellants requested that the decision under appeal be set aside and that the patent be revoked.

VI. The respondents requested as main request, that the appeal be dismissed and that the patent be maintained as granted and, as auxiliary request, that the case be remitted to the first instance for further prosecution.

VII. At the oral proceedings the Chairman announced that, considering the possible relevance of the considerable number of documents filed by both parties only at the appeal phase, the Board would only hear the parties' arguments based on the documents on which the decision under appeal had been based (documents D1 to D5) and that, if necessary, the Board intended to remit the case to the first instance for consideration of documents D6 to D14.

VIII. The wording of independent claim 1 reads as follows:

"A non-aqueous battery, comprising:
a positive electrode having a composition comprised of a nickel-lithium oxide and cobalt;
a negative electrode having a material which occludes and releases lithium reversibly; and
a non-aqueous electrolyte; wherein
the nickel-lithium oxide essentially represented by a formula LixNiOy, where $0 < x \leq 1.3$ and $1.8 < y < 2.2$, said formula having a ratio of I_{104} to I_{003} ranging from 0.35 to 0.68 when measured by an X-ray powder diffraction method using CuK_α as a target, said I_{104} representing a peak intensity of reflection of X-rays at an angle about $2\theta = 44$ degrees with respect to a lattice plane (104) in the composition, and said I_{003} representing another peak intensity of reflection of X-rays at an angle about $2\theta = 19$ degrees with respect to a lattice plane (003) in the composition; and
said cobalt is selected from a group consisting of an impurity and a composite oxide and is present in an amount less than 0.06 mole per 1 mole of nickel."

Claims 2 to 4 are dependent claims.

IX. The arguments of the appellants may be summarised as follows.

Document D3 discloses in Figure 1 an electrochemical cell comprising a positive electrode, having a composition comprised of nickel-lithium oxide, a negative electrode (which necessarily occludes and releases lithium), and a non-aqueous electrolyte ($LiBF_4$ in propylene carbonate). By galvanostatic charging the cathode material, $Li_{1-x}Ni_{11x}O_2$ is delithiated (to x=0.5) and the simulated X-ray diffraction pattern for this material is shown in Figure 7b. As can be seen from this Figure, the ratio of I_{104}/I_{003} equals 0.677 which is within the range defined in claim 1. On page 1145, at the end of the 2nd paragraph, it is disclosed that there is a very close correspondence between the simulated and the observed spectrum (in Figure 6). Therefore the delithiated ($Li_{0.5}NiO_2$) electrochemical cell in document D3 comprises all the features of claim 1 and thereby anticipates its subject-matter. In consequence, since claim 1 does not define any additional features its subject-matter does not involve an inventive step.

Furthermore, the subject-matter of claim 1 is obvious in the light of either of documents D4 or D5 in combination with the disclosure of document D2. Document D5 discloses an electrochemical cell comprising a nonaqueous electrolyte (see page 4, lines 49 to 56). On page 7, lines 3 to 6, it is disclosed that, for maximizing the cell capacity, the value x in Li_x should be between 0.8 and 1.0, i.e. the material should be close to stoichiometric. The skilled

person would find the solution of this problem, a $LiNiO_2$-material with stoichiometric composition, in document D2 which on page 4723 lists three compositions for $LiNiO_2$ of which the sample marked KH114 is closest to stoichiometric ($Li_{0.944}Ni_{1.056}O_2$). According to Table 1 on this page the observed X-ray intensities for the angles 44° and 19° have a ratio of 0.534. Therefore, by selecting the material according to sample KH114 in order to obtain a stoichiometric composition, the skilled person would arrive at the subject-matter of claim 1 without taking an inventive step.

For the same reason the subject-matter of this claim is obvious over the teaching of document D4 which discloses an electrochemical cell with a $Li_xNi_yO_2$ electrode. In Example 2 in column 4, lines 49 to 51, it is disclosed that in order to reduce the overvoltage it is recommended to start from a more stoichiometric material (than the material $Li_{0.85}Ni_{1.15}O_2$ in this Example). The skilled person would find a more stoichiometric material in the sample KH114 in document D2.

X. The arguments of the respondents may be summarised as follows.

The objections of the appellants based on document D3 are unfounded. Firstly, an objection pertaining to lack of novelty would constitute a fresh ground of opposition which, according to established Case Law, may not be introduced on appeal without the agreement of the patent proprietor. In the present case the patentee does not agree.

Secondly, with respect to inventive step, it should be noted that the electrochemical cell shown in Figure 1 of document D3 is not a battery as such. It is not clear how the appellant finds a value of 0.67 for the ratio of intensities in the simulated spectrum shown in Figure 7b because, according to the proprietor's own calculation, the value is between 0.7 and 0.73. Furthermore, the spectrum in Figure 7b is not a measured spectrum and the passage on page 1145 relied on by the appellant, apparently to provide a link between the simulated spectrum and the measured spectrum in Figure 6, is equivocal because it is not disclosed how "close" the correspondence is.

As to the other cited prior art documents D4 and D5, these disclose prior art non-aqueous electrochemical secondary batteries. The subject-matter of claim 1 differs from these prior art batteries in the selection of a Li_xNiO_y composition for the positive electrode having a particular ratio of two X-ray reflection peaks. This solves the technical problem of producing a battery having an improved discharge capacity. None of documents D1 to D5 provides a pointer towards this solution. The appellants have argued that the skilled person, by following the suggestion in D4 and D5 to select a stoichiometric composition for the $LiNiO_2$ material, would arrive at this solution, but this is incorrect, since a stoichiometric composition does not necessarily result in the claimed intensity peak ratio. This is, for instance, illustrated in document D1, Figure 2, where the intensity ratio of the 104- and 003-lines for the stoichiometric composition $Li_{0.96}Ni_{1.04}O_2$ is approximately 0.9. Also, it is quite clear from the patent that, apart from the mixture

ratio of the components (LiOH, $Ni(OH)_2$), the further process conditions (temperature, partial oxygen pressure) are equally decisive for the ratio of x-ray line intensities and the discharge performance as can be seen in the Examples in Table 1. Therefore there is no clear relationship between the amount of lithium present in the compound and the resultant discharge capacity nor between the stoichiometry and the discharge capacity.

Reasons for the Decision

1. The appeal is admissible.

2. *Grounds of appeal*

In the grounds of appeal only the reasoning of the opposition division pertaining to Articles 52(1) and 56 EPC was challenged by the appellants. With respect to the appellants' argument during the appeal proceedings that document D3 anticipates the subject-mater of claim 1, since the novelty of the claims was not an issue in the opposition proceedings (*see the Decision under Appeal, page 2, under "Article 100(a) EPC"*) this fresh ground of opposition may not be introduced into the appeal proceedings without the agreement of the patent proprietor (*see G 10/91*). Since the proprietor did not consent, the only issue to be considered in the appeal is therefore the question of inventive step.

3. *Inventive step - documents D1 to D5*

3.1.1 Closest prior art

In addressing the issue of inventive step, neither the decision nor the grounds of appeal follow the problem-solution approach. Although this approach is not mandatory, its correct application facilitates the objective assessment of inventive step and avoids *ex-post facto* analysis of the prior art (see Guidelines, Chapter C IV, 9.8; see also Case Law of the Boards of Appeal, 4th edition, 2001, Chapter I.D.2). In the decision, in discussing inventive step (page 2 of the Reasons, under "Article 100(a) EPC"), the first sentence reads "*D2 and D3 are the most relevant citations to the question of the intensity ratio I_{104}/I_{003} of the X-ray peaks of the lithiated nickel oxide*" and the arguments in the remainder of page 2 and page 3 are developed starting from these documents. In the grounds of appeal, the appellants cite this passage, state their agreement with it, and then draw a quite different conclusion.

3.1.2 At the oral proceedings before the Board, the appellants considered both document D3 (*their lack of novelty objection apart*) and either of documents D4 or D5 as the closest prior art.

3.1.3 Addressing these documents in sequence, document D2 relates to the measurement of magnetic properties (susceptibility) of a particular $LiNiO_2$-lattice. Document D3 addresses synthesis of the normal spinel $Li[Ni_2]O_4$ and a comparison with the trigonal $Li_{0.5}NiO_2$ phase. Although in Figure 1 of D3 an electrochemical

cell is shown, the purpose of this cell is to deintercalate the lithium-compound by galvanostatic charging in order to obtain the lithium compound in the desired metastable normal spinel phase. Therefore neither document D2 nor D3 relates to optimising a non-aqueous battery within the meaning of the patent in suit, because cyclic charging/ discharging behaviour, and in particular the question of optimising the discharge capacity, is not addressed. Hence these documents do not meet the criterion normally used to identify the closest prior art, namely a prior art document disclosing subject-matter conceived for the same purpose or aiming at the same objective as the claimed invention and having the most relevant technical features in common (Case Law already mentioned, Chapter I.D.3.1).

3.1.4 Document D4 discloses an electrochemical cell and ion conductors for these, wherein one type of ion conductor is $Li_xNi_yO_2$, and with the emphasis on reversibility and overvoltage. Document D5 equally relates to secondary electrochemical cells with a lithium-compound for a cathode material. Therefore either of these documents may be considered as a suitable starting point for applying the problem-solution approach.

3.2.1 The subject-matter of claim 1 differs from the electrochemical cell in document D4, Example 2 (column 4, lines 30 to 51) in that the nickel-lithium compound defined in claim 1 has a range of the intensity ratio of the I_{104} and I_{003} lines from 0.35 to 0.68. The subject-matter of claim 1 differs from the cell in document D5 by the same feature.

3.2.2 According to the patent specification (see page 2, "Summary of the Invention") this selection of the material provides improved battery characteristics such as an improved discharge capacity.

3.2.3 The objective problem solved in claim 1 can therefore be seen as improving the battery characteristics.

3.2.4 In the opinion of the appellants the skilled person, starting from the teaching of document D4, Example 2, would follow the suggestion in lines 49 to 51 of column 4 to prepare the $Li_xNi_yO_2$ material in a more stoichiometric composition than the actual composition of this Example ($Li_{0.85}Ni_{1.15}O_2$) in order to reduce the overvoltage. In looking for such a stoichiometric composition he would find in document D2 several samples of $LiNiO_2$ from which he would select the composition closest to stoichiometric, namely the sample labelled KH114 on page 1723, having the composition $Li_{0.944}Ni_{1.056}O_2$. Since this composition has the observed intensity of the 104-line of 534 versus an intensity value 1000 of the 003-line (Table 1), therefore resulting in an intensity ratio of 0.534, he would, by selecting this sample KH114 of the best stoichiometric composition, arrive at the subject-matter of claim 1 without an inventive step being involved.

3.2.5 The Board agrees with the position of the appellants that document D4, Example 2, suggests that a more stoichiometric $Li_xNi_yO_2$ material may be advantageous because of a lower overvoltage. However it does not share the view that the skilled person *would* have considered document D2 at all, because this document

does not relate to electrochemical batteries but to magnetic properties of the S=1/2 antiferromagnetic triangular lattice $LiNiO_2$. Furthermore, in following the suggestion in document D4 the skilled person would try to maintain the process parameters close to those of Example 2 (starting materials $LiOH.H_2O$ and Ni powder; heating under O_2-atmosphere at 750°C for 12 hours) and would only modify any steps to the extent necessary to obtain a more stoichiometric material (for instance, using an excess of LiOH as recommended in document D5, page 4, lines 26 and 27). In contrast, the samples KH112, KH113 and KH114 in document D2 are prepared under rather different process conditions (different starting materials, different temperature and heating duration), whence the skilled person, even if he would have become aware of this document's existence, would not have considered following a completely different preparation process since the electrochemical properties of the material prepared in this way are not disclosed in document D2 and are not readily predictable.

3.2.6 For similar reasons the Board considers a combination of the teachings of documents D5 and D2 as not obvious. The appellants referred to page 7, lines 3 to 6 of document D5 to a value of x in Li_x between about 0.8 and about 1.0 from which they concluded that this is a clear incentive to prepare a stoichiometric composition. In the Board's opinion, there is no reason why the skilled person would have consulted document D2, because document D5 itself teaches how to prepare a stoichiometric composition, namely by using about a 10% to about a 25% excess of LiOH (page 4, lines 26 to 27).

3.2.7 Therefore neither a combination of the teachings of documents D4 and D2, nor such a combination of the teachings of D5 and D2 would appear obvious.

3.3.1 As to document D3, the appellants have made reference to Figure 7b from which an intensity ratio of 0.677 of the diffracted X-ray lines at angles 44° and 19° could be determined. Although this is a simulated spectrum, by reference to page 1145, 2nd paragraph, it would be clear that the simulated spectrum closely corresponds to the measured one (in Figure 6). From this it would follow that the electrochemical cell in Figure 1 of D3 had all technical features of the battery defined in claim 1.

3.3.2 The Board finds these arguments of the appellants unpersuasive for the following reason. According to the cited paragraph on page 1145, the *measured* spectral data in Figure 6 were of insufficient quality to allow a quantitative structural analysis (because of the non-linear response of the photographic film and intense reflections). Therefore, if only because of the poor quality of the data, a reliable determination of the X-ray intensities from this data is impossible. Furthermore, the lowest curve recorded in Figure 6 is for the $Li_{0.5}NiO_2$-material at a temperature of 180°C, at which temperature the diffracted X-ray spectrum may well differ from the spectrum recorded at the temperature for usual conditions of the electrochemical cell (i.e. at room temperature). It appears that the simulation resulting in the spectra in Figure 7 was carried out in an attempt to compare the respective spectral *patterns*, i.e. the occurrence and position of the X-ray peaks for the spinel and the trigonal phase

in order to enable an identification of the phases in the measured spectra in Figure 6. Hence, in the opinion of the Board, no conclusion may be drawn from the simulation data in Figure 7 of document D3 on the ratio of the X-ray diffraction intensities of an actual $LiNiO_2$-sample.

3.3.3 It is therefore concluded that document D3 does not anticipate the subject-matter of claim 1. Furthermore, since the teaching of this document relates to a different purpose than that of the patent in suit, the document is not relevant for the discussion of inventive step.

3.4 The remaining document D1 cited during the opposition proceedings does not relate to non-aqueous batteries and its subject-matter is not relevant to the question of patentability.

3.5 In conclusion, none of the documents presented in the opposition proceedings (D1 to D5), alone or taken in combination, challenge the patentability of the subject-matter of claim 1. The opposition division's rejection of the opposition, inasmuch it was based on documents D1 to D5, was therefore justified.

4. *Documents D6 to D14*

4.1 However, of the documents filed by both parties in the appeal, at least document D13 appears to disclose more relevant prior art than documents D1 to D5, because it not only discloses a secondary battery material (as do documents D4 and D5), but in addition gives in its Table II X-ray diffraction intensities for the lattice

planes 104 and 003 of the LiNiO2-material and a ratio close to that claimed in claim 1. Surprisingly, although this document had been acknowledged in the patent application as originally filed, it was not previously referred to by the parties or by the examining and opposition divisions.

4.2 The Board does not wish to go into the merits of all the documents filed during the appeal proceedings but, since at least the relevance of document D13 has not yet been assessed and it appears to be greater than that of the documents considered so far, it finds that the basis of the proceedings has so substantially changed in the appeal as to present the Board with a completely fresh case.

4.3 Therefore the Board follows the accepted practice to remit the case to the first instance in accordance with Article 111(2) EPC for assessing the relevance of documents D6 to D14 to the question of inventive step of the claimed subject-matter, following the problem and solution approach.

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 for further prosecution.

The Registrar: The Chairman:

P. Martorana A. Klein