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Datasheet for the decision of 13 July 2023

Case Number: T 1891/21 - 3.3.05

Application Number: 09824871.9

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Language of the proceedings: EN

Title of invention:
POSITIVE ELECTRODE FOR LITHIUM SECONDARY BATTERY, AND LITHIUM SECONDARY BATTERY

Patent Proprietor:
GS Yuasa International Ltd.

Opponent:
SAFT

Headword:
Yuasa/positive electrode

Relevant legal provisions:
EPC Art. 100(b), 54, 56

Keyword:
Grounds for opposition - insufficiency of disclosure (no)
Novelty - main request (yes)
Inventive step - main request (no) - auxiliary request (yes)

Decisions cited:
G 0001/03, T 2001/12, T 0482/09, G 0002/21, T 0608/07,
T 0045/10

Catchword:

Beschwerdekammern
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Case Number: T 1891/21 - 3.3.05

D E C I S I O N
of Technical Board of Appeal 3.3.05
of 13 July 2023

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Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted on 16 August 2021 rejecting the opposition filed against European patent No. 2357693 pursuant to Article 101(2) EPC.**

Composition of the Board:

Chairman E. Bendl
Members: S. Besselmann
P. Guntz

Summary of Facts and Submissions

I. The appeal in this case lies from the opposition division's decision to reject the opposition against European patent EP 2 357 693 B1.
The patent in suit concerns a positive electrode for a lithium secondary battery, and a lithium secondary battery.

II. The following documents are of relevance here:

D1 US 2006/0216601 A1

D2 JP 2006 252895 A

D11 WO 2009/057834 A1

D13 "Phospho-olivines as Positive-Electrode Materials for Rechargeable Lithium Batteries", A.K. Padhi et al., J. Electrochem. Soc., Vol. 144, No. 4, April 1997, pp. 1188-94

D14 "Electrochemical study on Mn^{2+}-substitution in $LiFePO_4$ olivine compound", IMLB 2006, Abstract #0193

D16 FY 2008 "Annual Progress Report for Energy Storage Research and Development", D. Howell, U.S. Department of Energy, January 2009

D17 "$LiMnPO_4$ Cathodes for High Performance Li-Ion", I. Exnar et al., 216th ECS Meeting, October 4-9, 2009

D27 "$Li(Mn_{0.4}Fe_{0.6})PO_4$ cathode active material: Synthesis and electrochemical performance evaluation", Y.-J. Shin et al., Journal of Physics and Chemistry of Solids 69 (2008), 1253-6

D29 Experimental report on SEM/Image Analysis submitted by the opponent by letter of 21 April 2021

III. The opponent (appellant), in their statement of grounds of appeal, submitted *inter alia* document D33:

D33 G. Li et al., Journal of the Electrochemical Society, 149 (6), A743-747 (2002)

IV. In their reply to the appeal, the patent proprietor (respondent) defended the patent as granted (main request) and additionally filed auxiliary requests 1-22 and 24-28. An auxiliary request 23 was discussed (page 35) but not submitted. The respondent calculated additional data and provided an analysis concerning the examples of the patent in suit in their reply to the appeal (page 16 second paragraph to page 19, second paragraph, including Table 1 and Figure 1).

V. During the oral proceedings before the board on 13 July 2023, the respondent maintained their main request and auxiliary requests 6 and 24 and withdrew all the other requests.

VI. Independent claim 1 of the main request (patent as granted) relates to a positive electrode for a lithium secondary battery and reads as follows:

"*A positive electrode for a lithium secondary battery, comprising lithium manganese iron phosphate and a lithium-nickel-manganese-cobalt composite oxide; characterized in that an average particle size of particles of the lithium manganese iron phosphate is smaller than an average particle size of particles of the lithium-nickel-manganese-cobalt composite oxide.*"

VII. Claim 1 of auxiliary request 6 differs from the main request in that the following part is added at the end of the claim:

"*the number of manganese atoms contained in the lithium manganese iron phosphate is more than 50% and less than 100% relative to the total number of manganese atoms and iron atoms, and*
the number of cobalt atoms contained in the lithium-nickel-manganese-cobalt composite oxide is more than 0% and not more than 67% relative to the total number of nickel atoms, manganese atoms, and cobalt atoms."

Claims 2-8 relate to particular embodiments.

VIII. The appellant's arguments, where relevant to the present decision, can be summarised as follows.

Main request
The feature that an average particle size of the lithium manganese iron phosphate (LMFP) was smaller than an average particle size of the lithium-nickel-manganese-cobalt composite oxide (NMC)was so ill-defined that the claimed subject-matter was insufficiently disclosed.

This feature relating to the average particle sizes should not be taken into account for analysing novelty. Even if it were taken into account, it constituted a single selection from a limited number of alternatives. Novelty was thus lacking in view of D1 and D2.

Alternatively, this feature did not support an inventive step starting from D1 or D2 as the closest prior art, the technical problem being the provision of an alternative. The claimed subject-matter was obvious, having regard to the average particle sizes of known NMC (D16) and LMFP (D14 and D27).

Auxiliary request 6
The objection of insufficient disclosure was the same as for the main request.

The additional feature specifying the relative number of manganese atoms was not disclosed in D1 or D2 or D11. It could not be acknowledged that this feature solved the technical problem of improving the initial coulombic efficiency. Starting from D11 or D1 or D2, the objective technical problem was merely to provide an alternative. It would have been obvious to use LMFP with a high relative number of manganese atoms because this was known from D13 and D17. Moreover, it derived from D13 that the initial coulombic efficiency was highest for a manganese atom number of 50%.

IX. The respondent's arguments, where relevant to the present decision, can be summarised as follows.

Main request
The skilled person learned from the description how the average particle sizes had to be measured.

The feature relating to the average particle sizes distinguished the claimed subject-matter from D1 and D2.

This distinguishing feature had the technical effect of improved packing density, improved thermal stability and an increased high-rate charge-discharge characteristic, as taught in paragraph [0045] of the patent in suit. It could easily be visualised that the smaller LMFP particles surrounded the larger NMC particles. This had the consequence that the smaller LMFP particles acted as a heat sink and additionally that the lithium ion diffusion path was shorter.

It would not have been obvious to the skilled person to combine the teaching of D14, D16 or D27 with D1 or D2. The claimed subject-matter involved an inventive step.

Auxiliary request 6
Claim 1 was based on an additional inventive step, because it solved the technical problem of improving an initial coulombic efficiency. This effect could be directly derived from the patent in suit. The larger the manganese ratio of the LMFP, the greater the improvement in coulombic efficiency when mixing LMFP with NMC, which was beyond prediction.

X. The appellant (opponent) requested that the decision under appeal be set aside and that the European patent be revoked.

The respondent (patent proprietor) requested that the appeal be dismissed, i.e. that the patent be maintained on the basis of the main request or, in the alternative, on the basis of either auxiliary request 6 or auxiliary request 24 as submitted with the reply to the statement of grounds of appeal.

Reasons for the Decision

1. Article 12(6) RPBA

1.1 The appellant had submitted D33 with their statement of grounds of appeal but did not rely on it during the oral proceedings. In the written proceedings, the appellant had relied on Document D33 as a secondary document relevant to inventive step of the main request (patent as granted) and of auxiliary request 6. In its communication pursuant to Article 15(1) RPBA 2020, the board informed the appellant of its preliminary opinion that D33 should not be taken into account: it should have been filed before the opposition division because it concerned features of the claims as granted, and the amendments in auxiliary request 6 were based on the claims as granted (point 11 of the communication, in particular point 11.2).

1.2 D33 is not taken into account.

Main request - patent as granted

2. Sufficiency of disclosure, Article 100(b) EPC

2.1 The claim stipulates that an average particle size of particles of the lithium manganese iron phosphate (LMFP) is smaller than an average particle size of particles of the lithium-nickel-manganese-cobalt composite oxide (NMC).

2.2 According to the appellant, this feature relating to the average particle size was not defined and it was unknown how it had to be measured. Where the patent in suit mentioned the measuring of an average particle size, the disclosure did not relate to *comparing* LMFP with NMC. It was not even known if the average particle size of the primary or the secondary particles had to be considered. Even within a single method the uncertainty was high. For instance, the uncertainty of an image analysis using SEM was up to 25% as derived from D29. The examples in the patent in suit did not illustrate the claimed invention because the average particle sizes of about 10 µm for LMFP (paragraph [0092]) and 12.3 µm for NMC (paragraph [0099]) could be regarded as equal. The appellant concluded that *de facto* no measuring method was available, and that this case was thus similar to T 45/10. In their opinion, the feature was so ill-defined that the invention was insufficiently disclosed.

2.3 In decision T 45/10 (Reasons 4.3) it was found that, without indicating a measuring method, the expression "average particle size" was unclear, and the lack of any determination method in the patent specification resulted in insufficient disclosure.

2.4 In this case, the precise basis of comparing the average particle sizes is indeed unknown, because it is unknown what measuring methods are to be used for this purpose. The patent in suit mentions different methods for measuring an average particle size, but does not explicitly link them to comparing LMFP with NMC, nor is such a link implicit in this case. According to the patent in suit, the average particle size of secondary particles and primary particles of the LMFP can be determined by image analysis using TEM (paragraph [0031]). The average particle size of secondary particles of NMC is the D50 value measured using laser diffraction (paragraph [0041]). These measurements are performed in the examples (paragraphs [0092] and [0099]).

However, this disclosure of measuring methods is not explicitly linked to the teaching in paragraph [0045], which is the only paragraph where the relative comparison of the average particle sizes is mentioned, and it would in fact be counterintuitive to compare average particle sizes obtained using different methods, considering that image analysis methods may give different results from laser diffraction, and a number-based average may differ from a volume-based average.

2.5 The patent thus does not teach a clear basis for comparing the average particle sizes. Nevertheless, this case differs from the situation in T 45/10 in that measuring methods are mentioned, and more importantly in that it only needs to be established whether one average particle size is smaller than another; there is no need to additionally verify whether the average particle size falls within a certain range.

2.6 There can be no doubt that the skilled person would be able to carry out measurements of the average particle sizes and compare the values thus obtained, to select an average particle size of particles of the lithium manganese iron phosphate (LMFP) which is smaller than an average particle size of particles of the lithium-nickel-manganese-cobalt composite oxide (NMC). The skilled person would not have any difficulties to provide a positive electrode comprising these materials.

2.7 Whether the choice of a different pair of measuring methods would have led to the same result merely concerns the scope of protection, this being a requirement of Article 84 EPC. In this case this ambiguity merely concerns a limited part of the scope of the claim, where the respective average particle sizes are sufficiently similar or even identical. There is no indication that the ambiguity would permeate the whole claim (see T 608/07, point 2.5.2 of the Reasons).

Even in these hypothetical cases where the average particle sizes are almost identical, the skilled person would still have obtained a positive electrode, as can also be gathered from the examples in the patent in suit, where the average particle sizes, in the appellant's view, are equal.

In so far as the patent in suit mentions other desired effects such as improved high-rate charge-discharge characteristics (paragraph [0045]), they are not specified in the claim. Whether or not any desired effects not expressed in the claims under consideration are obtained is irrelevant to sufficiency of disclosure (see G 1/03 (OJ 2004, 413), Reasons 2.5.2; see also T 2001/12, Reasons 3.4). Furthermore, the skilled person trying to rework the invention could in unclear cases increase the difference in average particle size. This would also avoid ambiguities due to measuring uncertainty. In addition to these theoretical considerations there is no proof that in practical terms the claimed invention could not be carried out.

2.8 For these reasons, the ambiguity when comparing the average particle size in this case merely concerns the scope of protection, but would not have prevented the skilled person from carrying out the claimed invention. It thus does not result in insufficient disclosure.

3. Novelty

3.1 It was not contested that D1 did not explicitly disclose the average particle size of the materials described. According to the appellant, novelty was nevertheless lacking in view of D1 because the average particle size feature was unclear and thus should not be taken into account for analysing novelty.

3.2 While the indicated ambiguity of the average particle size feature needs to be taken into account when comparing the claim with the prior art, this does not necessarily mean that it should be entirely disregarded. There is no evidence that this feature would be entirely meaningless, in that, even in cases of a large difference between the average particle size of LMFP and NMC, it could not be verified whether that of LMFP is indeed smaller.

This view is in line with the case law cited by the appellant. According to Case Law of the Boards of Appeal of the EPO, 10th edn., 2022, II.C.5.5.1d citing T 482/09 (Reasons 2.1, last paragraph), the result of using an indefinite term in a claim was that it could not be taken to determine the limits to the protection sought, so that novelty and inventive step had to be assessed in the light of the prior art identified on the basis of all its technically meaningful possible interpretations.

According to the same case law, if the term had no specific meaning whatsoever, it would lose entirely its effect of delimiting the claimed subject-matter from the relevant prior art. However, this is not the case here.

3.3 The appellant argued that the LMFP and NMC materials inherently had a particle size. In the light of the above (point 3.2), this observation is insufficient to establish that the average particle size feature would be anticipated. D1 does not exemplify or disclose the actual preparation of LMFP particles. It only generally discloses that in chemical formula 4 ($Li_vFe_{1-w}M5_wPO_4$) M5 may represent manganese (paragraph [0039]). In the examples, $LiFePO_4$ (LFP as opposed to LMFP) is used but its preparation is not described. There are no details regarding the preparation or the physico-chemical properties of NMC either, though it is used in the examples. It is therefore not seen on what basis average particle sizes of LMFP and NMC or their relative comparison could be regarded as directly and unambiguously disclosed in D1, even in terms of a broad range taking deviations due to the choice of a measuring method and measuring uncertainties into account.

3.4 The appellant also argued that there were only three implicit possibilities as to how the average particle sizes compare, namely LMFP being smaller, LMFP being larger, or both being equal, and hence only a single selection from a list of alternatives was needed.

3.5 This argument is not convincing either. These possibilities are only conceptually encompassed, but not individualised. They cannot be treated as a list of alternatives. Moreover, a selection is already needed to arrive at M5 being manganese, so the appellant's approach would in fact involve a multiple selection.

3.6 There is no proof that the materials used in D1 would inevitably be those known from D16 (regarding NMC), D14 (regarding LMFP) and D27 (regarding LMFP), which according to the appellant show conventional values of the average particle sizes. D16 mentions three different commercial materials. D14 and D27 are scientific articles describing the preparation of LMFP. D1 does not refer to any of these documents.

3.7 Furthermore, the argument that the average particle size feature was anticipated because the primary particles of LMFP were inevitably smaller than the secondary particles of NMC is not convincing either.

Irrespective of whether the claim can be construed in this way, the ranges specified in the patent in suit do not support the appellant's assumption. The average particle size of primary particles of LMFP is 1 nm to 500 nm, and thus overlaps with the range for the average particle size of the secondary particles of NMC, which is 0.1 µm (100 nm) to 100 µm, see claims 5 and 6 of the patent in suit. There is no evidence that these ranges were incorrect, and that the primary particle size of LMFP must inevitably be smaller than the secondary particle size of NMC.

3.8 The objection of lack of novelty is thus not convincing.

4. Inventive step

4.1 The patent in suit relates to a positive electrode for a lithium secondary battery, and aims at providing high safety and improved initial coulombic efficiency and energy density (paragraph [0015]).

4.2 *Prior art*
D1 relates to a cathode material for a lithium secondary battery (paragraphs [0003] to [0008]) and thus to a similar purpose.

As follows from the comments regarding novelty, the distinguishing feature may be seen in that an average particle size of particles of the LMFP is smaller than an average particle size of particles of the NMC (i.e. the characterising portion of the claim), which however needs to be interpreted broadly for the reasons indicated (see considerations regarding sufficiency of disclosure, point 2. above).

4.3 *Technical problem*

According to the respondent, the technical problem was to provide a positive electrode for a lithium secondary battery which had an improved packing density and improved thermal stability, a short lithium ion diffusion path and thus improved high-rate charge-discharge characteristics, based on paragraph [0045] of the patent in suit.

4.4 Proposed solution

As the solution to this technical problem, the claimed positive electrode for a lithium secondary battery was proposed in which an average particle size of particles of the LMFP is smaller than an average particle size of particles of the NMC.

4.5 *Success of the proposed solution*

While paragraph [0045], which is the only paragraph where the difference of the average particle sizes is mentioned, does indicate the various effects (point 4.3), only the packing density is associated with this difference. The other mentioned effects in paragraph [0045] (improved thermal stability, shortened lithium ion diffusion path and improved high-rate charge-discharge characteristics) are attributed to the respective absolute particle sizes, which however are not specified in the claim. In contrast to the respondent's view, the observation that various effects are mentioned in a single paragraph does not lead to a different conclusion.

Moreover, the patent in suit provides no explanation or experiments supporting the alleged effect of the difference of the average particle sizes on the packing density. It cannot be assumed that the packing density would be governed by the difference of the average particle sizes alone. In line with the opposition division's view, the packing density may, for instance, be increased by broadening the particle size distribution (page 10 of the impugned decision, referring to D9). The claim neither specifies a minimum difference of the average particle sizes, nor is there any reference to the width of the particle size distribution.

As regards the alleged improvements in the thermal stability, the lithium ion diffusion path and the high-rate charge-discharge characteristics, no supporting evidence is available either.

The respondent argued that the smaller LMFP particles would necessarily surround the larger NMC particles, as could be visualised by a simple schematic drawing, and would therefore act as a heat sink, independently of the absolute particle size values. It was also readily apparent from the visualisation that the lithium ion diffusion path was shorter. In the appellant's view, this supported the conclusion that the technical effects of improved thermal stability and improved high-rate charge-discharge characteristics were due to the LMFP particles having a smaller average particle size.

However, the appellant's visualisation does not show that the absolute particle size ranges were irrelevant for obtaining the effects mentioned.

Additionally, it is to be borne in mind that the feature under consideration needs to be interpreted broadly, in that the claim does not specify the basis for measuring and comparing the respective average particle sizes, and different measuring methods may be used for each (see point 2. above). Hence no effect on packing density or any other property can be assumed where the average particle sizes are close to each other, and where the presence of the indicated distinguishing feature depends merely on the choice of measuring methods.

4.6 *Reformulation of the objective technical problem*
In the light of the above, the technical problem can merely be seen as providing an alternative.

4.7 *Obviousness of the proposed solution*
D1 does not describe the synthesis of the active materials. The skilled person wishing to carry out the teaching of D1 would need to turn to other documents to provide the active materials. These active materials inevitably have an average particle size.

D27 describes the synthesis of LMFP cathode active material having a small particle size of about 100 nm (abstract). Common average particle sizes of commercial NMC are larger (6 to 10.8 µm, see D16, page 73, Table III). Suitable NMC and LMFP materials meeting the average particle size requirement would thus be readily available to the skilled person.

Furthermore, D27 teaches that the small particle size contributes to the good properties of the material (abstract; page 1255, left-hand column, last full sentence). The skilled person would therefore even be motivated to provide this small particle size in the context of the teaching of D1 as well. At the same time, it is not apparent that the skilled person would be motivated to provide an even smaller average particle size for NMC (so the average particle size requirement would not be met). The respondent provided no counter-evidence as to the common average particle sizes.

4.8 In the light of the not-very-ambitious technical problem, the skilled person would thus readily provide active materials meeting the average particle size requirement, within the broad meaning of the claim.

4.9 The subject-matter of claim 1 thus lacks inventive step.

Auxiliary request 6

5. Claim 1 has been limited on the basis of claim 3 as granted, which specified that the number of manganese atoms contained in the lithium manganese iron phosphate is more than 50% and less than 100% relative to the total number of manganese atoms and iron atoms, and the number of cobalt atoms contained in the lithium-nickel-manganese-cobalt composite oxide is more than 0% and not more than 67% relative to the total number of nickel atoms, manganese atoms, and cobalt atoms.

6. Sufficiency of disclosure

6.1 The considerations are the same as for the main request (point 2.). Accordingly, the requirement of sufficiency of disclosure is met.

7. Inventive step in view of D11

7.1 The patent in suit is directed to a positive electrode for a lithium secondary battery which contains a positive active material that is capable of improving the initial coulombic efficiency while maintaining high safety (paragraph [0015]).

7.2 Prior art

D11 relates to the same general purpose of providing a cathode active material for a lithium secondary battery with high safety (paragraph [1]), and thus constitutes a suitable starting point for assessing inventive step.

It was not contested that D11 discloses a cathode active material for lithium secondary batteries comprising a core portion of NMC and a shell portion formed by dry-coating the core portion with a shell of olivine-structured lithium iron phosphate (LFP), and that before the dry-coating the average particle size of the LFP (of 1 µm or less) is smaller than the average particle size of the NMC (7 to 15 µm).

7.3 Technical problem

According to the respondent, the claimed positive electrode addressed the technical problem of improving the initial coulombic efficiency (paragraphs [0014], [0015], [0021], [0028], [0029] and [0037] of the patent in suit), and in particular of improving the increase in the actual initial coulombic efficiency in comparison with the predicted value (paragraphs [0137] and [0138]).

7.4 *Proposed solution*

As a solution to this technical problem there is proposed the positive electrode of claim 1, which contains lithium *manganese* iron phosphate (LMFP) and wherein the number of manganese atoms in the LMFP relative to the total number of manganese atoms and iron atoms is more than 50% and less than 100%.

7.5 *Success of the proposed solution*

7.5.1 The examples provided in the patent in suit illustrate the claimed solution.

The appellant was of the opinion that this was not the case because the average particle size requirement was not met. In their opinion, it was only known that by image analysis using TEM "the size of the obtained primary particles was about 100 nm" and "the secondary particle size was about 10 µm" (paragraph [0092] relating to LMFP). The value was thus uncertain, and it was not even known if it was an average particle size. It was similar to the average particle size (D50) of the NMC of 12.3 µm (paragraph [0099]).

This is not convincing. The average particle size requirement needs to be construed broadly (see considerations regarding sufficiency of disclosure, point 2.). The claim thus encompasses the examples, in which the particle size indicated for LMFP is smaller than the average particle size indicated for NMC.

7.5.2 The examples provided in the patent in suit show that the initial coulombic efficiency of NMC in combination with LMFP is higher than would have been expected on the basis of the mixing ratio and the respective initial coulombic efficiency of each of NMC and LMFP used individually (paragraph [0137]).

In Example 7, an initial coulombic efficiency of 92.3% was obtained, while the expected value was only 89% (same paragraph). In the case of Examples 1 to 5, the initial coulombic efficiency is even greater than that of each of the two components (i.e. NMC and LMFP) used alone (Table 1).

All the examples relate to LMFP materials with a high number of manganese atoms (relative to the sum of iron and manganese). According to the patent in suit, a high relative number of manganese atoms of more than 50% and less than 100% leads to improved initial coulombic efficiency (paragraphs [0028] and [0029]). Figure 1 submitted by the respondent with their reply to the grounds of appeal depicts some data of the examples in the patent in suit. It shows that the extent of the increase over the predicted value depends on the relative number of manganese atoms in the LMFP, the increase being greater at higher numbers of manganese atoms. This figure thus supports the indicated general teaching in the patent that a high proportion of manganese contributes to an improved initial coulombic efficiency. Furthermore, no counter-evidence is available.

7.5.3 The indicated technical effect of increasing the actual initial coulombic efficiency in comparison with the predicted value may be derived from the patent in suit and the underlying application as originally filed.

As follows from the above considerations, the concept of comparing the actual initial coulombic efficiency with the predicted value is described in paragraphs [0137] and [0138] of the patent in suit (paragraph [0096] of the application as originally filed). The predicted value has been indicated in respect of Example 7, and it is also mentioned that the result of Example 9 is "beyond prediction". Furthermore, all the necessary data to enable the skilled person to easily calculate the predicted values of the other examples where necessary is provided in Tables 1 and 2. The calculation is unnecessary where an increase beyond the predicted value is directly apparent from the initial coulombic efficiencies of each of the respective NMC and LMFP, used alone, being lower.

The respondent's submission in their reply to the appeal (page 16, second paragraph to page 19, second paragraph, including Table 1 and Figure 1) included the calculation of additional values of some of these other examples. This submission thus merely made explicit what the skilled person would have derived from the patent in suit. This also applies to the "rate of increase" which the respondent defined as the ratio of the measured value of the initial coulombic efficiency over the predicted value. This rate is a numerical expression reflecting the taught comparison of the measured initial coulombic efficiency with the predicted value.

There is thus no doubt that the requirement formulated in G 2/21 (Headnote II) is met, and the patent proprietor may rely on this technical effect for inventive step (ibid.).

7.5.4 The appellant argued that the comparison with the predicted value was not a relevant indicator, because the predicted value was of no technical interest. They also observed that the initial coulombic efficiency of Example 7 (50:50 mixture of LMFP and NMC) was lower than that of the same NMC material taken alone

(Comparative Example 6); this also applied to Example 9 in view of Comparative Example 1 (Tables 1 and 2 of the patent in suit). They furthermore held that the law of mixture was irrelevant with regard to D11, where no mixture, but core-shell particles, were prepared.

This is not convincing. While it is desirable that the actual initial coulombic efficiency be even increased compared with using each of NMC and LMFP alone (Table 1, paragraph [0134]), it is to be borne in mind that the general purpose of combining NMC with LMFP is to obtain high battery safety (paragraphs [0007]-[0010], [0014], [0015], [0021]). Using NMC alone, as in the indicated comparative examples, is described as providing lower battery safety (paragraphs [0137] and [0138]). It is therefore justified to compare the actual initial coulombic efficiency with the value predicted on the basis of the separate contributions of the individual components. This allows any combined effect caused by the interaction of the NMC and the LMFP to be identified.

7.5.5 There is no reason either why this concept of comparing the actual value with the predicted value could not be applied when the core-shell particles of D11 were the closest prior art. The basis of the comparison, namely the sum of the individual contributions, remains the same irrespective of whether the actual material is a mixture or, for instance, a core-shell particle as known from D11. While the claim does not specify how the two materials, NMC and LMFP, are combined, and thus does not exclude a core-shell particle, there is no evidence that the desired effect would not be obtainable for a core-shell particle.

7.5.6 It was also debated whether the technical problem was only solved in the case of an LMFP:NMC mass ratio within the range of 10:90 to 70:30. However, according to the respondent's calculation, the correctness of which was not contested, the initial coulombic efficiency of Example 6 - relating to an LMFP:NMC mass ratio of 80:20 - was also greater than the predicted value. The teaching in the patent in suit that the initial coulombic efficiency is higher in the case of a mass ratio within the indicated range than in the case of other ratios (paragraphs [0043] and [0135]) is thus to be construed as a preferred feature, aimed at a further improvement, but is not essential for solving the problem posed. The examples illustrate a broad range of mass ratios (from 10:90 to 80:20, Table 1). There is no evidence that the technical problem under consideration would only be solved within a limited range of mass ratios.

7.5.7 In the light of the above, it is to be accepted that the technical problem of improving the initial coulombic efficiency, as evidenced by an increase compared with the predicted value, is solved.

7.6 Non-obviousness of the proposed solution

The skilled person could not have expected that LMFP materials having a high relative number of manganese atoms of 50% would lead to a combined effect in interaction with NMC, namely an increase in the initial coulombic efficiency beyond the individual contributions.

7.6.1 D11 is silent as to the possible presence of manganese in the olivine-structured lithium iron phosphate (LFP); there is no mention of lithium *manganese* iron phosphate (LMFP).

According to D11, the olivine-structured LFP layer increases the resistance to overcharge (paragraphs [16], [17], [51]). It restricts the contact of the active material core with an electrolyte liquid when overcharging (paragraph [51]). D11 teaches that the active material core (the NMC core) is coated with the olivine-structured lithium iron oxide phosphate having "the largest resistance increasing rate according to overcharge" (paragraph [51]). The skilled person thus learns from D11 to use specifically LFP, and there is no indication in D11 that it could be replaced with alternative materials for this same purpose.

For this reason alone it is doubtful whether the skilled person would have considered this material to be replaceable by LMFP, which likewise has an olivine structure but in which more than 50% of the iron is replaced by manganese.

7.6.2 Furthermore, while LMFP materials having a high relative number of manganese atoms of 50% and more are known (D13, D17), and are even described as having beneficial properties, there is no teaching in the prior art that these materials would be particularly suitable for combining with NMC.

D13 depicts charge-discharge curves of LMFP having different relative numbers of manganese atoms (x=0.25, x=0.5, x=0.75, see Figure 10). The appellant calculated the initial coulombic efficiency on the basis of these curves, comparing the respective initial charging and the first discharge cycle. They found that it was best at x=0.5.

D17 does not specifically address the effect of manganese on initial coulombic efficiency. It relates to an LMFP material having a relative number of manganese atoms of 80% and describes several beneficial properties. D17 indicates that the LMFP particles are an excellent cathode material for lithium-ion batteries, have a stable reversible capacity of 165 $mAhg^{-1}$, excellent cycling, very fast rate capabilities, and excellent thermal stability, and are the least surface-reactive cathodes in lithium-ion electrolyte (slide 16). It is also indicated that lithium manganese phosphate has a better potential than lithium iron phosphate (slide 4). According to the appellant, this disclosure implied that LMFP had a better energy density than LFP.

However, these results in D13 and D17 concern the properties of LMFP used alone, not its interaction with NMC. They do not enable the skilled person to predict any increase in the initial coulombic efficiency due to the interaction of LMFP with NMC.

7.7 In conclusion, the increase in the initial coulombic efficiency compared with the predicted value is to be regarded as surprising. The skilled person, starting from D11 and faced with the technical problem of improving the initial coulombic efficiency as evidenced by an increase compared with the predicted value (point 7.5.7), would not have been prompted by the cited prior art to replace the LFP with LMFP having a high relative number of manganese atoms of greater than 50% and less than 100%.

7.8 Therefore an inventive step in view of D11 is to be acknowledged.

8. Inventive step in view of D1

8.1 The appellant raised an additional objection of lack of inventive step starting from D1.

8.2 Reference is made to the comments regarding the main request (point 4.).

As regards lithium iron phosphate, D1 more specifically discloses the chemical formula $Li_vFe_{1-w}M5_wPO_4$ (paragraph [0037]), where M5 can be manganese and its relative proportion can be up to 10 atom% ($w \leq 0.1$) (paragraph [0039]).

The subject-matter of claim 1 thus additionally differs from D1 in the relative proportion of manganese atoms.

8.3 The technical problem associated with this difference is the same as considered in relation to D11, namely to improve the initial coulombic efficiency, as evidenced by an increase compared with the predicted value (point 7.5.7).

8.4 For the same reasons as set out in view of D11 (point 7.), the skilled person faced with this technical problem would not have been prompted by the teachings of D13 or D17 to increase the manganese proportion to a value of more than 50% relative to the total number of manganese atoms and iron atoms. They would not have combined the teachings of D13 or D17 with D1, all the more so since D1 teaches only a small proportion of manganese of up to 10% ($w \leq 0.1$, as indicated), while the examples point to even lower proportions because they only relate to LFP (0%, no manganese or other M5 element present).

9. Document D2

9.1 The appellant cited D2 in their written submission as being relevant similarly to D1 (statement of grounds of appeal, point 5.3 in conjunction with the appellant's submission of 26 September 2022, point 3.6.2.1).

9.2 D2 is only available in the Japanese-language original: no translation was filed. The novelty objection in view of D2 relied on the examining division's opinion in the examination phase (statement of grounds of appeal, point 3.1) and may thus be understood to be based on the acknowledgement in paragraph [0002] of the patent in suit. The appellant did not submit any arguments as to why D2 should be more relevant than D1, nor is this apparent from D2, as far as it can be understood. Thus, starting from D2, no different conclusion would be reached. There was consequently no need additionally to deal with D2.

10. In the light of the above (points 7. to 9.), the subject-matter of claim 1 of auxiliary request 6 is novel and involves an inventive step.

Claims 2 to 8 directly or indirectly depend on claim 1 and thus meet the requirements of novelty and inventive step for the same reasons.

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 auxiliary request 6 as submitted with the reply to the grounds of appeal and a description to be adapted.

The Registrar: The Chairman:

C. Vodz E. Bendl

Decision electronically authenticated