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# Datasheet for the decision of 12 May 2015

Case Number: T 0168/11 - 3.3.05

05732741.3 Application Number:

Publication Number: 1829140

IPC: H01M4/36

Language of the proceedings: ΕN

Title of invention:

HIGH DISCHARGE CAPACITY LITHIUM BATTERY

Patent Proprietor:

EVEREADY BATTERY COMPANY, INC.

Opponent:

The Gillette Company

Headword:

Lithium iron sulfide battery/EVEREADY BATTERY COMPANY INC.

#### Relevant legal provisions:

EPC Art. 56, 114(2)

## Keyword:

Inventive step (main request, auxiliary requests 1 and 2): no - obvious alternative

Inventive step (auxiliary request 3): yes

Refusal of a document by first instance (discretion correctly exercised)

Late filed documents (not admitted)

# Decisions cited:

G 0007/93

# Catchword:



# Beschwerdekammern Boards of Appeal Chambres de recours

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Case Number: T 0168/11 - 3.3.05

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

Appellant: The Gillette Company (Opponent) One Gillette Park - 3E Boston, MA 02127 (US)

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Decision under appeal: Interlocutory decision of the Opposition

Division of the European Patent Office posted on 17 November 2010 concerning maintenance of the European Patent No. 1829140 in amended form.

#### Composition of the Board:

D. Prietzel-Funk

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# Summary of Facts and Submissions

- I. European patent EP-B1-1 829 140 was granted with 32 claims (Bulletin 2008/24). An opposition was lodged against the granted patent, based on the grounds of opposition according to Articles 100(a) and (b) EPC.
- II. The documents cited included the following:
  - D2: A. Webber, "Improvements in Energizer's L91
    Li-FeS<sub>2</sub> AA Cells", Proc. of the 41<sup>st</sup>
    Power Source Conference, 14-17 June 2004,
    Philadelphia, Pa., USA, paper 2.3
  - D2a: 41<sup>st</sup> Power Source Conference, 14-17 June 2004, Philadelphia, Pa., USA, "List of Participants, Exhibition Program, and Sessions Listing"
  - D4: C. Iwakura et al., "Preparation of Iron
    Disulfide and its use for Lithium batteries",
    Electrochim. Acta vol. 28, no. 1,
    pages 269 to 275, 1993
  - D5: Yang Shao-Horn et al., "Nano-FeS<sub>2</sub> for Commercial Li/FeS2 Primary Batteries", J. Electrochem. Soc. 149 (11) A 1499 to A1502 (2002)
  - D7: L.A. Montoro and J.M. Rosolen, "Gelatine/DMSO:

    A new approach to enhancing the performance of a
    pyrite electrode in a lithium battery", Solid
    State Ionics 159 (2003), pages 233 to 240
  - D8: E. Strauss et al., "To the electrochemistry of pyrite in Li/solid composite-polymer-electrolyte battery", J. Power Sources 115 (2003), pages 323 to 331

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- D10: Yang Shao-Horn and Quinn Horn, "Chemical, structural and electrochemical comparison of natural and synthetic  $FeS_2$  pyrite in lithium cells", Electrochim. Acta 46 (2001), pages 2613 to 2621 (cited in D5)
- III. Documents D8 and D9 were not admitted by the opposition division as being late filed and not relevant.
- IV. The opposition division rejected the ground of opposition under Article 100(b) EPC, directed only against the subject-matter of claim 29 of the main request then on file, as unfounded.

Concerning the ground of opposition under Article  $100\,(a)$  EPC, the opposition division decided that the subject-matter of the claims 1 to 32, as amended during the oral proceedings, was novel. The distinguishing claim feature was the average particle size range of the iron disulfide particles of 1 to 19  $\mu$ m which was not disclosed in documents D1, D4, D5 or D7. D7 mentioned a 10  $\mu$ m size  $FeS_2$  cathode powder, but it was not clear whether this value referred to the average particle size. This 10  $\mu$ m particulate material, after sieving through a 400 mesh (38  $\mu$ m) sieve, was further processed by flotation and drying, processes which could affect the particle size. Therefore, D7 was not considered to be novelty-destroying.

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The opposition division considered D7 to represent the closest prior art. The problem to be solved consisted in improving the cell discharge characteristics at normal and at lower temperatures. The solution consisted in selecting pyrite powder of 1 to 19  $\mu m$  average particle size as a cathode active material. The opposition division accepted the success of the solution in view of the effect demonstrated by example 7, Tables 5a and 5b and example 10, Table 7, in comparison with a FeS2 control sample of 22  $\mu m$  average particle size.

As to obviousness, none of the available documents in the opposition division's view suggested to use pyrite having the claimed particle size to achieve the desired increase in cell performance. In fact, no prior art document even addressed the problem of depressed temperature discharge of this type of cell.

Therefore, the opposition division decided to maintain the patent on the basis of claims 1 to 32 of the patentee's main request. This is the decision under appeal.

V. The notice of appeal of the opponent (henceforth: the appellant) was received with letter dated 19 January 2011. The statement of grounds of appeal was accompanied by four new documents:

D11: US-A-6 203 947

D12: US-A-4 902 589

D13: US-B2-7 687 189 (30 March 2010)

D14: US-A-2004/0121234.

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- VI. The patentee (respondent) replied by letter dated 26 September 2011. It filed new sets of claims as auxiliary requests 1 and 2.
- VII. Oral proceedings took place on 12 May 2015. The respondent filed a new sets of claims as a <u>main request</u> and further an <u>auxiliary request</u>. The previously filed auxiliary requests 1 and 2 were withdrawn. In both newly filed requests, claim 29 and the claims depending on claim 29 were deleted.
- VIII. The independent claims thereof read:

the positive electrode."

## Main request:

- "1. An electrochemical battery cell comprising: a housing;
- a negative electrode comprising lithium;
- a positive electrode comprising an active material, said active material comprising greater than 49 weight percent of iron disulfide, said iron disulfide having an average particle size of 1 to 19  $\mu$ m; an electrolyte mixture comprising at least one salt dissolved in a nonaqueous electrolyte disposed within
- the housing; and a separator disposed between the negative electrode and
- "17. An electrochemical battery cell comprising: a housing;
- a negative electrode comprising lithium; a positive electrode comprising an active material, said active material comprising greater than 49 weight percent of iron disulfide; an electrolyte mixture comprising at least one salt dissolved in a nonaqueous electrolyte disposed within the housing; and a separator disposed

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between the negative electrode and the positive electrode, wherein

- a) the electrolyte has a conductivity greater than
- 2.5 mS/cm and the iron disulfide has an average particle size of 1 to 19  $\mu$ m; or
- b) the separator has a thickness of 1 to 25  $\mu m$  and the iron disulfide has an average particle size of 1 to 19  $\mu m$  ."
- "26. A process for preparing a cathode, comprising the steps of

forming a slurry comprising a wetting agent and iron disulfide articles having an average particle size greater than 20  $\mu\text{m}_{\star}$ 

milling the slurry utilizing a media mill comprising grinding media to reduce the particle size of the iron disulfide particles to 1 to 19  $\mu$ m average particle size,

applying the milled cathode slurry to a cathode substrate to form a cathode, and drying the cathode."

## Auxiliary request:

- "1. An electrochemical battery cell comprising:
- a housing;
- a negative electrode comprising lithium;
- a positive electrode comprising an active material, said active material comprising greater than 49 weight percent of iron disulfide, said iron disulfide having an average particle size of  $\bf 2$  to 19  $\mu m$  and wherein the iron disulfide is natural;

an electrolyte mixture comprising at least one salt dissolved in a nonaqueous electrolyte disposed within the housing;

and a separator disposed between the negative electrode

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and the positive electrode."

- "16. An electrochemical battery cell comprising: a housing;
- a negative electrode comprising lithium; a positive electrode comprising an active material, said active material comprising greater than 49 weight percent of iron disulfide; an electrolyte mixture comprising at least one salt dissolved in a nonaqueous electrolyte disposed within the housing; and a separator disposed between the negative electrode and the positive electrode, wherein
- a) the electrolyte has a conductivity greater than 2.5 mS/cm and the iron disulfide has an average particle size of 2 to 19  $\mu$ m and wherein the iron disulfide is natural; or
- b) the separator has a thickness of 1 to 25  $\mu m$  and the iron disulfide has an average particle size of 1 to 19  $\mu m$  ."
- "25. A process for preparing a cathode, comprising the steps of

forming a slurry comprising a wetting agent and iron disulfide articles having an average particle size greater than 20  $\mu\text{m}\textsc{,}$ 

milling the slurry utilizing a media mill comprising grinding media to reduce the particle size of the iron disulfide particles to  $\bf 2$  to 19  $\mu m$  average particle size and wherein the iron disulfide is natural,

applying the milled cathode slurry to a cathode substrate to form a cathode, and

drying the cathode."

Claims 2 to 15, 17 to 24, and 26 and 27 represent particular embodiments of claims 1, 16 and 25 on which they depend.

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# IX. The <u>appellant</u> essentially argued as follows:

The opposition division had incorrectly and unfairly exercised its discretion in not admitting documents D8 and D9. The board should admit them due to their relevance and due to the fact that they were filed in response to amended claims.

## Novelty:

D7 related to Lithium batteries wherein the positive electrode was formed of a mixture of gelatine-pyrite, DMSO-pyrite or pristine pyrite (78%), PVDF (5%), PEO (7%), carbon black (10%) and acetone. This mixture was spread on an Al foil using a doctor blade and the solvent evaporated. The active material, pyrite, had been prepared by ball-milling natural pyrite crystals, followed by sieving, to yield particles of "~10 µm".

Without doubt "particle ~10 µm" meant particles of average size of about 10 µm, as this was the only sensible interpretation. The step of sieving through a 37 µm mesh sieve was not in contradiction with a particle size of 10 µm, because the opposed patent itself referred to sieving through a 63 µm sieve in order to obtain 22 µm particles. There was also no indication in D7 that subsequent flotation and drying changed the average particle size so as to take the sample outside the range of claim 1. These process steps were carried out purely for purification of the iron sulphide.

Therefore, D7 anticipated the subject-matter of claim 1.

Document D8 disclosed pyrite electrodes for Li batteries. According to chapter 3 (page 325), 1 to 10 um particles of pyrite could be distinguished on the surface of the composite cathode, whereas nano-size pyrite particles were found on a RFE (sputtered) cathode (see SEM images Figure 2). Because the pyrite was homogeneously distributed throughout the electrodes, measuring the particle size at the surface also revealed the particle size elsewhere in the electrode. The pyrite particles in the cathode formed by the doctor blade technique could not possibly have had a particle size greater than 10µm, because that was the thickness of the cathode itself (legend to Figure 5 on page 328). For falling outside the claimed range, more than half of the pyrite particles would have to be of a size of less than 1 um, which was in contradiction to the actual disclosure of D8.

Therefore, D8 also anticipated the subject-matter of claim 1.

Document D11: Example 2 disclosed a Li/CPE/FeS $_2$  cell with a 7 µm thick composite cathode of 100% iron disulfide formed by ball milling pristine pyrite for 48 hours (column 7, lines 21 to 24). This cell had all the features of claim 1 of the opposed patent.

D12: The preferred average particle size of the active material ( $inter\ alia$  iron sulfide) in a Li battery was 1 to 30  $\mu m$  (see column 7).

## Inventive step:

The advantage of using smaller particle size pyrite was well known to the skilled person (for instance from D4 and D9). D5 taught that there existed a lower level of

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about 1  $\mu m$  particle size below which no further advantage was seen.

D2 could be taken as the closest prior art. It disclosed jelly-roll Li/FeS $_2$  batteries with a cathode of pyrite coated onto Al foil, a polyolefin microporous separator (20  $\mu$ m thickness), and a Li foil anode. Starting from D2, it would have been obvious to select a mean particle size of 1 to 19  $\mu$ m for the reasons given by documents D4, D5 and D9.

In another line of argument, starting from D5 as the closest prior art, the only difference was that the particle size of the  $FeS_2$  was selected to be 1  $\mu m$  rather than 0.5  $\mu m$ . Because there were no date relating to the lower end of the claimed particle size range, there was no evidence that any problem was solved by this minor modification. In fact, it was highly unlikely that a cell comprising  $FeS_2$  having a particle size of 1  $\mu m$  had a higher discharge capacity than a cell as disclosed in D5 with an average particle size of the  $FeS_2$  of 0.5  $\mu m$ . The problem to be solved by the patent under appeal was thus merely the provision of an alternative  $Li/FeS_2$  cell. This was obvious in view of D5 itself using both  $FeS_2$  particles greater and smaller than 1  $\mu m$ .

In still another line of argument, D11 was considered to represent the closest prior art.

X. The respondent essentially argued as follows:

#### Late-filed documents:

Documents D8, D9, D11, D12, D13 and D14 should not be admitted as they were late filed and irrelevant. The

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version of claims under consideration in the first instance decision was substantially the same as the claims as granted, i.e. the documents could have been filed earlier. If any of these documents was admitted, the respondent requested that the case be remitted to the opposition division for further prosecution.

As a precaution, the respondent put forward arguments as to why none of the new documents D11 to D14 was pertinent for novelty and/or inventive step.

## Novelty:

D8: It was unclear what was meant by "~10  $\mu$ m size particles" and whether "size" referred to diameter, circumference, length, width or some other measure. Said "size" was determined by reference to SEM images, a method different from the one prescribed in the opposed patent. D8 failed to describe or quantify the particles that were not on the surface of the cathode. There was also no disclosure of the lower limit or particle size. The appellant's arguments based on the thickness of the electrodes were also assuming several facts not yet proven. Finally, D8 included a grinding step which made it likely that a substantial number of particles in the electrode was smaller than 1 µm. In D8, the pyrite was said to be distributed homogeneously, but this did not suggest that the particles themselves in the distribution were the same.

D7: This document disclosed a pyrite powder having a micrometer size, but did not specify it further (for example its average particle size). The 400 mesh sieve used in the sieving process was considerably larger, which cast doubt on the size of the particles sieved. Finally, flotation and drying were expected to

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influence the particle size in an unknown manner, for instance by reactions on the pyrite surface. Therefore, the opposition division correctly concluded that D7 was not novelty-destroying.

#### Inventive step:

The object of the opposed patent was the provision of an electrochemical cell being improved in discharge density at low temperature (for instance, -20 °C). The appellant had started, in various approaches, from D2 and D5 as the closest prior art, and from D11 as regarded the subject-matter of process claim 26.

However, none of these documents attempted, directly or indirectly, to solve the problem of providing an improved cell performance at low temperatures. Therefore, these documents did not qualify as the closest prior art.

Admittedly, D4 contained a broad and generic teaching that synthetic pyrite may be preferred for use over natural pyrite. However, the effect of the average particle size was not discussed in D4, and no specific maximum limits of size were disclosed. When combining D4 with D5, it was apparent that the preferred particle size range allegedly implied by this reference was actually above the range stated in the claims of the opposed patent.

The appellant cited D5 as the closest prior art for the proposition that "there was a lower limit to particle size, beyond which no further advantage is obtained". However, D5 in fact taught two average particle sizes, one for the "natural sample" and one for the "synthetic sample", both of which were above the range stated in

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the claims of the opposed patent. D5 gave no incentive for going below 20  $\mu m$  average particle size.

## XI. Requests

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

The respondent requested that the patent be maintained in amended form on the basis of the claims according to the main request, filed during oral proceedings, or in the alternative on the basis of the auxiliary request, filed during oral proceedings.

## Reasons for the Decision

- 1. Admissibility of late-filed documents
- 1.1 Documents D12 to D14

D12 to 14, filed with the statement of grounds of appeal, are not admitted into the procedure. These documents are *prima facie* not more relevant than documents D1 to D7.

#### 1.2 Documents D9 and D11

According to the appellant, document D11, column 7, lines 16 to 35, example 2, disclosed a Li/CPE/FeS<sub>2</sub> cell having a 7 µm thick composite cathode comprising a 7 µm pyrite powder. Said powder was formed by ball milling pristine pyrite powder for 48 hours. According to the appellant, the cell disclosed in example 2 anticipated the novelty of the subject-matter of claim 1 of the opposed patent.

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D11 literally discloses that "For the preparation of ultra-tin [sic] cathodes, 7  $\mu$ m pristine pyrite powder was used. This powder was formed by ball milling pristine pyrite for 48 hours." The board considers this passage to be ambiguous, because the term "pristine" is used in connection with the starting material of FeS<sub>2</sub> and in connection with the milled powder. It is therefore unclear what the particle size of the pyrite after milling was. Furthermore, it is not stated whether the particle size is a (volume) average particle size.

There is no further mentioning in D11 of particle sizes in connection with  $\text{Li/FeS}_2$  cells.

In summary, for the board, D11 thus adds nothing relevant to the other documents. It is therefore not admitted at this stage of the procedure.

D9 is a theoretical (mathematical) paper on the influence of the particle size distribution (PSD) on discharge capacity of intercalation electrodes. Pyrite electrodes are not mentioned, nor is there a disclosure or suggestion towards using an active material within the claimed range of average particle sizes. Therefore, D9 is not admitted.

#### 1.3 Document **D8**

In certain situations, the EPC allows an examining or opposition division discretion in giving or withholding its consent to, for instance, accept requests or admit documents. This discretion has to be exercised responsibly and the relevant reasons must be given (see G 7/93 (OJ 1994, 775). A board should overrule a first

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instance decision of discretion only if it came to the conclusion that the division had not exercised its discretion in accordance with the right principles, or unreasonably, or that it had exceeded its proper limit of discretion.

The appellant did not submit any reason why the discretion of the opposition division not to admit D8 was not correctly exercised. Thus the decision not to admit D8 stands and will not be overruled by the Board.

## 2. Admission of the late filed main request

The problem whether to admit of the late filed main request may have included some slight arguments in favour or against. Since the request fails for other reasons, the answer on this point does not have any impact on the result.

#### 3. Novelty regarding both requests on file

D7 relates to lithium batteries having a positive electrode formed from a mixture of gelatine-pyrite, DMSO-pyrite or pristine pyrite (78%), PVDF (5%), PEO (7%), carbon black (10%) and acetone. The active material, pyrite, was prepared by ball-milling natural pyrite crystals (from Mineropar) for 24 hrs, followed by sieving (400 mesh), to yield pyrite particles of "~10 µm" which were purified by flotation in tetrabromomethane and acetone and dried under vacuum (see page 234, Section "2. Experimental"). This mixture was applied onto an Al foil using a doctor blade, and the solvent evaporated. The electrodes were then subjected to cyclic voltagrammetry (see page 235, Section "3. Results and discussion", Figure 1).

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In the appellant's view, D7 anticipated the subject-matter of claim 1 of all requests. However, the board disagrees.

- Firstly, the term "~10  $\mu m$  size particles" is unclear as such and certainly cannot be interpreted as "average particle size of 10  $\mu m$ ".
- Secondly, the particle size was determined by reference to SEM images, a method different from the one prescribed in the opposed patent (see paragraph [0060] of the specification). In such circumstances, it would have been incumbent on the appellant to provide evidence showing that the two methods indeed yield identical results.
- Thirdly, in D7 the pyrite was distributed homogeneously, but this does not suggest that the particles themselves in the distribution were the same.
- Lastly, while there is no disclosure in D7 that the process steps of flotation and drying change the average particle size so as to take the sample outside the range of claim 1, this cannot be ruled out.

In conclusion, D7 fails to disclose, clearly and unambiguously, the claimed subject-matter.

The requirements of Article 54 EPC are therefore met.

#### 4. Inventive step

#### 4.1 Invention

The patent in suit concerns lithium ion batteries, in particular of the FR6 (or AA) type, comprising iron

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disulfide ( $FeS_2$ ) as an active material in the positive electrode.

## Main request

# 4.2 Closest prior art

D5 is considered to represent the closest prior art.

D5 is concerned with improvements in the discharge properties of Li/FeS2 AA primary batteries. It is reported from previous studies that the performance of Li/FeS2 system at high current demands could be improved by reducing the crystal sizes from 10 to 1 µm. It was expected that a further improvement could be achieved by going to nano-size pyrite powder (see page A1499, left hand column, lines 1 to 15). Accordingly, in the D5 paper the rate capability of lithium cells containing nano-FeS2 pyrite samples was compared to that of micrometer-sized FeS2 in lithium cells (see abstract). The nano-FeS2 sample (obtained from US Nanocorp) had an average particle size of 0.5 µm and each particle consisted of nano-FeS2 crystals on the order of 50 nm. Using this active material, coin cell Li/FeS2 packages were constructed containing an FeS2 electrode consisting of 49%  $FeS_2$ , an electrolyte consisting of 1M LiCF<sub>3</sub>SO<sub>3</sub> in a mixed organic solvent (dioxolane/dimethoxyethane/dimethylisoxazole), and two pieces of Celgard 2500 separators per cell (see Figure 2; pages A1499, right hand column, section "Experimental"; page A1500, left hand column).

The rate capability of batteries containing the nanosize  $FeS_2$  was found to be superior to that of the Li/  $FeS_2$  batteries containing natural iron sulfide with an

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average <u>crystal</u> size of 10 µm (about 30 µm particle size) (supplier Chemetall). However, no improvement was found relative to the synthetic FeS<sub>2</sub> (supplier Alfa AESAR) electrodes having an average <u>crystal</u> size of 1 µm. The authors argue that the rate capability of commercial Li/FeS<sub>2</sub> batteries may be limited by the ion conductivity of the electrolyte when FeS<sub>2</sub> samples have crystal sizes smaller than 1 µm (see Figures 3a, 3b and 5; page A1501, right hand column).

Therefore, D5 may be said to have the same purpose and effect as the opposed patent as regards improving discharge capacity and runtime of the Li batteries. However, D5 does not report low temperature behaviour of such cells.

#### 4.3 Problem

According to the description, the object of the opposed patent is to improve the discharge capacity (see paragraphs [0004] and [0013]), discharge time (paragraph [0063]) and low temperature cell performance (paragraph [0108]).

## 4.4 Solution

As a solution to this problem, the patent proposes an electrochemical lithium/iron disulfide cell in accordance with claim 1 of the main request, characterized in that the iron disulfide has an average particle size of 1 to 19  $\mu m$ .

#### 4.5 Success of the solution

The opposed patent contains experimental evidence showing that the discharge time of a cylindrical FR6

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type lithium cell containing jet-milled  $FeS_2$  (average particle size 4.9 µm) is longer (better) than that of a comparative lithium cell containing coarse  $FeS_2$  (average particle size 75 µm) or of a control sample containing  $FeS_2$  having an average particle size of 22 µm. The improvement, measured at room temperature, was more pronounced at high service FEP (see paragraphs [0101] to [0103], Example 7 and Tables 5a and 5b).

Furthermore, Example 10 (see paragraph [0108] and Table 7) compares cell performance of media milled cathodes of different average particle size  $FeS_2$  of 5 to 10  $\mu$ m (according to the invention) and 22  $\mu$ m (control) at standard (21°C) and low temperatures (0°C and -20°C). The lithium cell with a reduced particle size  $FeS_2$  had an improved performance at low temperatures. The differences were most significant at -20°C.

However, the opposed patent does not contain evidence demonstrating similar improvements having regard to the nano-sized FeS2 electrodes of D5 which represent the closest prior art. Such an effect or improvement (for instance in discharge capacity or low temperature performance) is also not plausible, because it would imply a sudden discontinuity of cell properties between electrodes containing  $FeS_2$  of 1 µm average particle size (lower end point of claim 1 of the patent in suit) and cells containing nano-size 0.5  $\mu m$  FeS<sub>2</sub>. In fact, since the particle sizes are average values, one should expect a substantial overlap in the respective particle size distributions such that many particles in both distributions will effectively be of the same size. Accordingly, essentially similar, if not identical electrochemical behavior of the electrodes and the

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batteries is to be expected.

Therefore, the board cannot accept that the above defined technical problem has been successfully solved. The problem has to be reformulated and may be seen as providing an alternative  $\text{Li/FeS}_2$  battery.

# 4.6 Obviousness

It remains to be discussed whether the claimed solution is obvious having regard to the prior art.

With respect to the features of claim 1, the batteries of D5 differ only in that the average particle size of the FeS $_2$  is not within the claimed range of 1 to 19  $\mu$ m, but slightly lower (average particle size of 0.5  $\mu$ m) (see Figure 2).

In the board's view, the gap between a battery comprising nano-sized pyrite electrodes of D5 and the subject-matter of current claim 1 is of an order that would not deter the skilled person from bridging it. As already said before, an alternative battery having essentially the same performance characteristics as in D5 could have been expected using a marginally coarser  $FeS_2$  of 1  $\mu$ m average particle size.

The respondent argued that the skilled person was dissuaded from using particle sizes in the claimed range because D5 stated (see concluding remarks on page A1502) that "it is believed that the ionic conductivity of the electrolyte, rather than  $FeS_2$  crystal sizes, limited the commercial Li/FeS2 performance when the FeS2 had an average crystal size smaller than 1  $\mu$ m." In this respect, it could be inferred from D10 (page 2619, chapter "Physical Characterization"; and page 2616,

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Figure 3) that a sample having an individual crystal size of 1  $\mu m$  corresponds to a particle size larger than 20  $\mu m$  .

The board cannot accept this argument. D5 teaches that the nano-FeS $_2$  electrodes (0.5 µm average particle size) outperformed the relatively coarse 10 µm natural FeS $_2$  electrodes, but were equivalent to the synthetic FeS $_2$  electrodes having an average crystal size of 1 µm. Therefore, D5 suggests using not only ultra-fine FeS $_2$  of 0.5 µm average particle size, but also of up to 10 to 20 µm (corresponding to a crystal size of about 1 µm).

In conclusion, the subject-matter of claim 1 of the main request is obvious having regard to D5 (Article 56 EPC).

## Auxiliary request

## 5. Admissibility

This request was filed during oral proceedings.

The appellant argued that it should not be admitted, because the claims were filed late and not clearly allowable.

The board decided to admit the amended claims because they were filed in reaction to an objection of lack of inventive step based on document D5 which came up for the first time in the oral proceedings before the board. Under such circumstances the board exceptionally found it appropriate to give the respondent a final

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chance for filing restricted claims.

# 6. Inventive step

#### 6.1 Invention

The subject-matter claimed in claim 1 of the auxiliary request differs from the one according to claim 1 of the main request in two respects:

- the average particle size of the iron disulfide is 2 to 19  $\mu m$ ; and
- the iron disulfide is of natural origin (i.e. not synthetic).

The invention concerns an electrochemical battery cell (see claims 1 and 16) and a process for preparing a cathode (see claim 25).

## 6.2 Closest prior art

The closest prior art is the same as set out in point 4.2.

## 6.3 Problem

See 4.3.

## 6.4 Solution

As a solution to the above problem the patent in suit proposes an electrochemical  $\text{Li/FeS}_2$  cell in accordance with claim 1, characterized in that the iron disulfide has an average particle size of 2 to 19  $\mu$ m and in that the iron disulfide is **natural**.

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### 6.5 Success of the solution

The board comes to the same conclusion as under point 4.5 and is satisfied that the reformulated problem i.e. the provision of an alternative FeS<sub>2</sub> battery is solved.

#### 6.6 Obviousness

It remains to be discussed whether the claimed solution is obvious having regard to the prior art.

As discussed before (see 4.6), it can be derived from D5 that the nano-FeS2 electrodes (0.5 µm average particle size) outperform the relatively coarse 10 µm natural FeS2 electrodes, but not synthetic FeS2 electrodes having an average crystal size of 1 µm. However, D5 does not suggest using natural pyrite particles whose particle size is also substantially coarser than those of the nano-FeS2 samples which performed best. Natural pyrite has an average particle size on the order of 50 to 100 µm, substantially larger than synthetic pyrite (e.g. made from Fe<sub>2</sub>O<sub>3</sub> or FeCl<sub>2</sub> (see D4, page 271, Figure 2, microphotographs A, B and C, D; page 274, section 3.5). Therefore, natural pyrite must be milled to obtain a product having an average particle size in the claimed range. Milling of a sensitive material such as  $FeS_2$  to micrometer particles sizes is not trivial for risk of oxidation and/or decomposition. The opposed patent proposes a nonmechanical milling, preferably a jet-mill, which is advantageous in that no temperature increase occurs during milling (see paragraphs [0058] and [0059]). In view of these additional limitations the skilled person would not have chosen natural FeS2 as a starting material for manufacturing an alternative Li/FeS2 cell

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performing similar to the best nano-size  $FeS_2$  cells disclosed in D5.

The subject-matter of claim 1 therefore involves an inventive step having regard to D5 (Article 56 EPC).

Starting for the sake of argument from D2, no other conclusion would be reached. D2 is a paper on nonaqueous Li/FeS<sub>2</sub> AA cells presented at the 41<sup>st</sup> Power Sources Conference in June 2004. The advantages of the Li cell design in comparison with alkaline batteries (Figure 2, 3 and 4; Table 3; Figure 5), especially at low temperatures, are described. However, there is no disclosure or suggestion as to useful particle sizes of the pyrite, nor whether a synthetic or a natural pyrite is used. Therefore, even if one took D2 in combination with D5, one would not arrive at the claimed invention.

The subject-matter of claim 1 therefore also involves an inventive step having regard to D2 (Article 56 EPC).

6.7 As to claims 16 (electrochemical cell) and 25 (process for preparing a cathode), the reasoning under 7.2 to 7.6 applies mutatis mutandis to the subject-matter of these claims which contain the essential features required for the presence of an inventive step, namely the average particle size of 2 to 19 µm of iron disulfide and its natural origin.

Claims 2 to 15, 17 to 24 and 26 and 27 depend on claims 1, 16 and 25 respectively from which they derive their patentability.

The requirements of Article 56 EPC are met.

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## 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 in amended form on the basis of claims 1 to 27 of the auxiliary request filed during oral proceedings, and a description and figures to be adapted.

The Registrar:

The Chairman:



C. Vodz G. Raths

Decision electronically authenticated