

Veröffentlichung im Amtsblatt  
Publication in the Official Journal  
Publication au Journal Officiel

Ja/Nein  
Yes/No  
Oui/Non



Aktenzeichen / Case Number / N<sup>o</sup> du recours : T 231/88 - 3.4.1

Anmeldenummer / Filing No / N<sup>o</sup> de la demande : 83 301 558.9

Veröffentlichungs-Nr. / Publication No / N<sup>o</sup> de la publication : 0 089 842

Bezeichnung der Erfindung: Lead acid storage cell and a positive electrode  
Title of invention: therefor  
Titre de l'invention :

Klassifikation / Classification / Classement :

### ENTSCHEIDUNG / DECISION

vom / of / du 27 April 1989

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /  
Titulaire du brevet :

UNITED KINGDOM ATOMIC ENERGY  
AUTHORITY

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence :

EPO / EPC / CBE Art. 56

Schlagwort / Keyword / Mot clé : Inventive step (yes).

Leitsatz / Headnote / Sommaire

Europäisches  
Patentamt

European Patent  
Office

Office européen  
des brevets

Beschwerdekammern

Boards of Appeal

Chambres de recours

Case Number : T 231/88 - 3.4.1



**D E C I S I O N**  
of the Technical Board of Appeal 3.4.1  
of 27 April 1989

**Appellant :** United Kingdom Atomic Energy  
Authority  
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**Representative :** Mansell, Keith Rodney  
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**Decision under appeal :** Decision of Examining Division 019 of the European  
Patent Office dated 30 December 1987 refusing  
European patent application No. 83 301 558.7  
pursuant to Article 97(1) EPC

**Composition of the Board :**

**Chairman :** K. Lederer

**Members :** C. Black

R.L.J. Schulte

### Summary of Facts and Submissions

- I. European patent application No. 83 301 558.9 (publication No. 0 089 842) was refused by decision of the Examining Division.
- II. This decision was based on Claims 1 to 3 received 23 January 1987.
- III. The reason for the refusal was that none of the two subject-matters alternatively claimed by Claim 1 involved an inventive step having regard to the disclosure in J. Appl. Chem. Vol. 16, June 1966, pages 180 to 184 (hereinafter D1), Electrochimica Acta, Vol. 26, No. 10, 1981, pages 1435 to 1438 (hereinafter D2) and GB-A-2 023 558 (hereinafter D4).
- IV. An appeal was lodged against this decision. In the statement of grounds for the appeal the Appellant (Applicant) introduced five further documents (A) to (E),
  - (A) Journal of the Electrochemical Society, Vol. 120, No. 11, November 1973, pp. 1515-1516
  - (B) Journal of the Electrochemical Society, Vol. 122, No. 4, April 1975, pp. 461-466
  - (C) Materials Science and Engineering, 28(1977) pp. 167-199.
  - (D) Journal of Power Sources, 4(1979) pp. 203-213.
  - (E) Paper prepared for presentation at the ILZRO Battery Research Meeting at Munich, Germany, May 6, 1980

and argued that the preparations of  $\text{PbO}_2$  disclosed in D1 were for use as standards in the X-ray investigation of the structure of electroformed  $\beta\text{-PbO}_2$ . D1 does not disclose that chemically prepared  $\beta\text{-PbO}_2$  could be used as the active material in a lead acid storage cell, and in this respect the Examining Division put the wrong interpretation on Conclusions (2) of D1. Further although a standard sample prepared by hydrolysis of lead tetraacetate with acetic acid and an electroformed sample were similar as regards the intensity ratios of their X-ray diffraction patterns, Table II showed that they were chemically different.

Although D2 and D4 disclose chemically prepared  $\text{PbO}_2$  which is electrochemically active, the preparation requires oxidation using ozone under alkaline conditions. D2 states that preparation under acid conditions gives a product with low electrochemical activity. D4 further in its review of prior art states that the electrochemical activity of  $\text{PbO}_2$  made by oxidising  $\text{Pb}_3\text{O}_4$  with nitric acid could be substantially improved but this was to be interpreted in the light of what was already known from documents (A) to (D), that is that chemically prepared lead dioxide had little or no electrochemical activity. He further argued that in testing a material for electrochemical activity it is not incorporated into the positive electrode of a lead-acid cell, therefore that this is not disclosed in D4 in respect of the product of nitric acid oxidation of  $\text{Pb}_3\text{O}_4$ .

Accordingly no combination of D1, D2, and D4, would lead to the subject-matter of Claim 1.

- V. In response to a communication from the Board, the Appellant submitted an amended set of two claims, of which Claim 1, the only independent claim, reads as follows:

A method of making a lead acid electric storage cell comprising the steps of

(i) chemically preparing particulate beta lead dioxide;

(ii) supporting the beta lead dioxide by a carrier therefor adapted to be located in the cell thereby to fabricate an electrode for the cell;

(iii) assembling a lead acid electric storage cell comprising a positive electrode constituted by said beta lead dioxide electrode, a negative electrode constituted by a second electrode, and an electrolyte for the cell, characterised in that, in step (i), the beta lead dioxide is prepared by reacting trilead tetroxide with nitric acid or by hydrolysing lead tetraacetate in acetic acid and is then mixed with either or both of a binder therefor and a particulate electrically conductive material inert to the electrochemical reactions inside the cell; and, in step (ii), the carrier comprises a permeable, electrically insulating tubular container for the beta lead dioxide and into which container a current collecting means extends, the beta lead dioxide having been mixed with at least the electrically conductive material, or the carrier comprises an electrically conductive support structure, the beta lead dioxide having been mixed with at least the binder.

VI. The Appellant requests that the decision refusing the application be cancelled and, as can be interpreted from the content of his response dated 15 December 1988, that a patent be granted on the basis of Claims 1 and 2 accompanying the response.

#### Reasons for the Decision

1. The appeal is admissible.

2. Claim 1 differs from that on which the refusal was based and which already embraced two alternatives based on original Claims 1 plus 13 and 1 plus 15 in that the wording "and is then mixed .... at least the binder" has been added. The effect of this amendment is that Claim 1 now contains four additional features claimed in various combinations. These features are derived from original Claims 2 (carrier is insulating tubular container), 3 (carrier is electrically conductive support structure), 4 (the lead dioxide is mixed with a binder therefor) and 10 (the lead dioxide is mixed with inert particulate electrically conductive material). Moreover, the amended claim contains the proviso that when the carrier is an insulating tubular container the lead dioxide must be mixed with at least the electrically conductive material and when the carrier is a conductive support structure the lead dioxide must be mixed with at least the binder. Claim 1 has therefore been limited to eight combinations of original Claims 1, 2, 3, 4, 10, 13, and 15 with the further, clearly allowable restriction to a method of making a lead acid storage cell rather than a positive electrode therefor, and no objection arises in respect of Article 123(2).

3. **Novelty**

3.1 D4 discloses a method of making a lead acid storage cell (see the references to accumulator production on page 1, lines 87 to 91 and page 2, line 46 to 51) wherein electrochemically active lead dioxide ( $\text{PbO}_2$ ) is prepared chemically by subjecting an aqueous alkaline suspension of an oxidizable lead substrate to oxidation using gaseous ozone. The lead dioxide may be mixed with inter alia carbon or plastics material added to the suspension. D4 does not specify  $\beta\text{-PbO}_2$  but this is implicit in that the

PbO<sub>2</sub> is electrochemically active, and has not been disputed by the Appellant. Moreover, D2, whose author is one of the inventors named in D4, contains substantially the same disclosure as regards oxidation with ozone, and does specify β-PbO<sub>2</sub>.

The subject-matter of Claim 1 differs from the disclosure in D4, as set out so far, (a) in that the β-PbO<sub>2</sub> is prepared by reacting trilead tetroxide with nitric acid or by hydrolyzing lead tetraacetate in acetic acid (b) in that the particulate electrically conductive material, e.g. carbon, and/or the binder, e.g. PTFE, is mixed with the β-PbO<sub>2</sub> after its preparation and (c) in the specific requirements associated with the carrier.

D4 also discloses, in its introductory paragraph, that the known art includes the preparation of PbO<sub>2</sub> by oxidation of minium (trilead tetroxide) with nitric acid. It goes on to state that the specific electrochemical activity could be very substantially improved, which would seem to mean that the said activity was too low for practical purposes. It then states that PbO<sub>2</sub> made in this way is used very little for electrochemical purposes. The Appellant argues, that the expression "for electrochemical purposes" does not mean that the material has been used in an accumulator, because, in the interest of economy, it is standard practice to screen materials for electrochemical activity in a half-cell rather than in an accumulator. This argument appears plausible and can be accepted by the Board. Accordingly the introductory paragraph of D4 is not seen as disclosing that PbO<sub>2</sub> prepared by reacting trilead tetroxide with nitric acid has been used in lead acid storage cells.

- 3.2 D1 discloses the preparation of β-PbO<sub>2</sub> by hydrolysis of lead tetraacetate with acetic acid and by oxidation of trilead tetroxide with nitric acid - see Preparation of

samples, (vi) and (vii) - that is, the two methods embraced by Claim 1 of the application in suit. The aim in D1 of the preparation is, however, to provide standard samples for comparison purposes in X-ray studies of the electroformed  $\beta$ -PbO<sub>2</sub> in lead acid cell positive plates and not the use of chemically prepared  $\beta$ -PbO<sub>2</sub> in the manufacture of such plates. In this respect the Board's interpretation of the disclosure of D1, in particular the final sentence under "Conclusions", differs from that of the Examining Division.

3.3 All other documents to be considered by the Board are more remote from the subject-matter of Claim 1, which is accordingly novel in the sense of Article 54 EPC.

4. **Inventive step.**

4.1 The process disclosed in D4 corresponds to the prior art portion of Claim 1 and constitutes in the Board's view the closest prior art to its subject-matter. In particular both D4 and the application in suit seek to overcome the disadvantages of the conventional electroforming step in the manufacture of lead acid storage cells by using chemically prepared lead dioxide. Accordingly, in the light of the disclosure in D4, the technical problem which the method defined in Claim 1 seeks to solve can be seen as providing an alternative, possibly improved method of making lead acid storage cells using chemically prepared lead dioxide.

4.2 This problem is solved by the features of the characterising portion of Claim 1, which, as indicated above, confer novelty on its subject-matter.

4.3 Claim 1 requires that  $\beta$ -lead dioxide is prepared chemically either by reacting trilead tetroxide with nitric acid or by hydrolysing lead tetraacetate with

acetic acid. As regards the first of these possibilities, the Board accepts that the introductory paragraph of D4 would not encourage the average skilled person to investigate further this route to the preparation of electrochemically active lead dioxide. In fact D4 and the related D2 are the only ones of the cited documents which disclose chemically prepared  $\beta$ -PbO<sub>2</sub> as having at least some electrochemical activity, documents (A) to (E) in particular referring consistently to the lack of electrochemical activity thereof. On the other hand, document D1, which concerns the preparation of standard samples for X-ray studies (see paragraph 3.2 above) would not suggest, in the Board's view, the use of these materials in the manufacture of positive cell plates. Moreover, the  $\beta$ -PbO<sub>2</sub> produced by oxidation of Pb<sub>3</sub>O<sub>4</sub> with HNO<sub>3</sub> was even rejected as a standard sample because it gave very poor X-ray patterns. Further D2, as pointed out by the Appellant, indicates that reaction under acid conditions gives a product with low electrochemical activity.

Therefore to the extent that Claim 1 requires preparation of  $\beta$ -PbO<sub>2</sub> by reaction of trilead tetroxide with nitric acid its subject-matter can be seen as involving an inventive step.

As indicated in paragraph 3.2, D1 discloses both of the methods of chemically preparing  $\beta$ -PbO<sub>2</sub> featured in Claim 1 but for the preparation of standard samples. D1 also discloses in Table II that the hydrogen content of  $\beta$ -PbO<sub>2</sub> prepared by reacting trilead tetroxide with nitric acid is 0.045%, by reacting lead tetraacetate with acetic acid is 0.083% and by electroformation 0.106%, all of these values being subject to an error of 20%. Now the average skilled person knows from (A), page 1516 and (E) page 3 that the loss in capacity of secondary batteries during repeated cycling appears to be associated with loss of hydrogen from the  $\beta$ -PbO<sub>2</sub> and as a corollary that electrochemical

activity is associated with the presence of hydrogen. Since from D1 it is known that the hydrogen content of  $\beta$ - $\text{PbO}_2$  prepared by hydrolysing lead tetraacetate with acetic acid lies roughly midway between that of  $\beta$ - $\text{PbO}_2$  prepared by reacting trilead tetroxide with nitric acid and that prepared electrochemically, the question arises as to whether the average skilled person would be lead to investigate the electrochemical activity of  $\beta$ - $\text{PbO}_2$  prepared by hydrolysis of lead tetraacetate. In concluding that it would not have been obvious to do so the Board has been influenced by the facts that although the authors of (A), published June 1966, were aware of D1 (published November 1973), because (A) contains a reference to D1, neither (A) nor (E), by two of the authors of (A) and published May 1980, contain any suggestion to use  $\beta$ - $\text{PbO}_2$  prepared by hydrolysis of lead tetraacetate in lead acid cells, or that thus prepared  $\beta$ - $\text{PbO}_2$  might be electrochemically active. In fact both (A) and (E) maintain that chemically prepared  $\beta$ - $\text{PbO}_2$  is electrochemically inactive. Moreover as pointed out by the Appellant, (A) in its opening paragraph refers to four other possible causes of loss in capacity of secondary batteries.

The Examining Division also considered that an obvious combination of the teachings of D1 and D2/D4 would lead to the subject-matter of Claim 1 in that D1 disclosed that the X-ray diffraction studies of the product of hydrolysing lead tetraacetate showed it to be similar to that obtained by electrochemical formation. However as pointed out by the Appellant, what D1 is saying is that the intensity ratios of their X-ray diffraction patterns are similar; on the other hand, as shown by Table II their chemical compositions are different. Therefore, here also there is no suggestion that lead dioxide made by hydrolysing lead tetraacetate with acetic acid could be used as the active material in a lead-acid storage cell.

4.4 In view of the foregoing it is not necessary to investigate the remaining features of the characterising portion of Claim 1 in respect of their contribution to an inventive step in its subject-matter. These features are seen more as introducing a further distinction between the subject-matter of Claim 1 and the disclosure in the introductory paragraph of D4 and therefore going some way towards removing the inconsistency between the said paragraph, indicating that lead dioxide prepared by reacting trilead tetroxide with nitric acid is electrochemically inactive, and the application in suit, wherein thus prepared lead dioxide forms a constituent of a positive electrode with a significant discharge capacity. The Board, in the communication referred to in paragraph V, had expressed doubts as to whether the presence or absence of binder and/or particulate electrically conductive material could have a role to play as regards the electrochemical activity of the positive electrode; it is however recognised that although lead acid storage cells have a long history, their mode of action, as pointed out by the Appellant, is still not fully understood.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.
2. The case is referred back to the first instance with the order to grant a European patent on the basis of the following documents:

Claims 1, 2 received 19 December 1988;

Pages 1, 4, renumbered 3a, received 26 May 1986;

Page 2 received 23 January 1987;  
Page 3 received 19 December 1988;  
Pages 4 to 9 as originally filed;  
Drawings sheet 1/2, 2/2 as originally filed;  
with the following amendments:-  
Pages 4, 5 and 7 as requested in Appellant's letter  
received 26 May 1986.  
Page 2: correct spelling of "Electrochemica" to  
"Electrochimica".  
Page 3: Delete lines 29 to 34 and substitute:  
"In applications of the invention wherein the beta lead  
dioxide is mixed with a binder therefor, the binder  
preferably comprises polytetrafluoroethylene (hereinafter  
referred to"  
  
as requested in Appellant's letter received 10 March  
1989.

The Registrar:

The Chairman:

F.Klein

K.Lederer