

Internal distribution code:

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

**Datasheet for the decision
of 17 June 2010**

Case Number: T 0437/07 - 3.3.05

Application Number: 02731829.4

Publication Number: 1393393

IPC: H01M 4/24

Language of the proceedings: EN

Title of invention:

Ionically conductive additive for zinc-based anode in alkaline electrochemical cells

Patentee:

RAYOVAC CORPORATION

Opponent:

-

Headword:

Electrochemical cell/RAYOVAC CORP.

Relevant legal provisions:

EPC Art. 56

Relevant legal provisions (EPC 1973):

-

Keyword:

"Inventive step (yes) - evidence for improvement in the application in suit"

Decisions cited:

-

Catchword:

-



Case Number: T 0437/07 - 3.3.05

D E C I S I O N
of the Technical Board of Appeal 3.3.05
of 17 June 2010

Appellant:

RAYOVAC CORPORATION
601 Rayovac Drive
Madison
Wisconsin 53711 (US)

Representative:

Chiva, Andrew Peter
Dehns
St Bride's House
10 Salisbury Square
London EC4Y 8JD (GB)

Decision under appeal:

Decision of the Examining Division of the
European Patent Office posted 24 October 2006
refusing European application No. 02731829.4
pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: G. Raths
Members: H. Engl
C. Vallet

Summary of Facts and Submissions

- I. The appeal lies against the decision of the examining division refusing European patent application EP02731829.4 on the ground of lack of inventive step.
- II. The examining division relied *inter alia* on the following documents:
- D1: EP-A-0 518 407
 - D2: US-A-4 614 026
 - D3: US-A-6 464 709
 - D6: EP-A-0 768 723
 - D7: Ep-A-0 700 104
- III. Starting from document D6 as the closest prior art, the examining division defined the technical problem underlying the application under appeal as providing an environmentally friendly mercury-free button type cell having a long service life and high operating voltages. The examining division argued in the contested decision that it would have been obvious to the person skilled in the art to use clay additives of the kind disclosed in D1 and D2 in order to improve a battery's discharge behaviour. It was observed that a decreased self-discharge (ascribed in D1 to the clay additive) evidently resulted in a longer service life since charge losses due to parasitic effects were reduced. These beneficial effects were to be expected not only in rechargeable accumulators with metal suspension half cells, such as disclosed in D1, but also in gelled zinc anode batteries. The application was therefore refused.

IV. The notice of appeal and statement of grounds of appeal were received with letters dated 3 January 2007 and 2 March 2007, respectively. The appellant also filed new sets of claims as a main request and auxiliary request.

V. In a communication in preparation of oral proceedings the board made provisional comments and, starting from document D3 as the closest prior art, defined the technical problem underlying the application as improving the cell voltage and service life discharge profile of an electrochemical cell of the type disclosed in D3.

Further sets of amended claims as a main request and first and second auxiliary requests were received with letter dated 17 May 2010.

VI. With telefax of 11 June 2010 the appellant withdrew the main request previously on file. It filed a new set of claims as a main request, amended pages 1, 2, 5, 6 and 7 of the description and an amended Figure 1. The second auxiliary request of 17 May 2010 was retained as new first auxiliary request.

VII. The independent claims of the said new main request are worded as follows:

"1. An electrochemical cell (10) comprising:
a container;
an anode (22);
a cathode (40) disposed within the container, wherein the cathode has an inner surface that defines a centrally disposed void (45) filled with a gelled anode

mixture (26); and
a separator (74) disposed between the inner surface
(32) of the cathode (40) and the gelled anode mixture;

said gelled anode mixture comprising a gelling agent
selected from the group consisting of carboxymethyl
cellulose, polyacrylic acid, sodium polyacrylate, and
salts thereof, an electrolyte, and a mercury-free anode
active material including zinc; and characterized in
that said gelled anode mixture further comprises an
ionically conductive clay additive that is dispersed
throughout the gelled anode mixture, wherein the
ionically conductive clay additive is the synthetic
clay Laponite having the empirical formula
 $[\text{Na}_{0.70}^{0.7+}[(\text{Si}_8\text{Mg}_{5.5}\text{Li}_{0.3})\text{O}_{20}(\text{OH})_4]^{0.7-}]$.

"7. A method for producing a mercury-free gelled anode
(22) for an electrochemical cell (9) in accordance with
any preceding claim, the method comprising

- (A) providing a mercury-free zinc alloy powder;
- (B) blending the zinc alloy powder with a gelling agent
to form a gelled mixture;
- (C) providing an alkaline electrolyte;
- (D) mixing an ionically conductive clay additive with
at least one of: the zinc alloy powder of method step
(A), the gelled mixture of method step (B), and the
alkaline electrolyte of method step (C); and
- (E) mixing the gelled mixture with the alkaline
electrolyte;

wherein said gelling agent is selected from the group
consisting of carboxymethyl cellulose, polyacrylic
acid, sodium polyacrylate, and salts thereof; and
wherein the ionically conductive clay additive is the

synthetic clay Laponite having the empirical formula
 $[\text{Na}_{0.70}^{0.7+}[(\text{Si}_8\text{Mg}_{5.5}\text{Li}_{0.3})\text{O}_{20}(\text{OH})_4]^{0.7-}]$."

VIII. Summary of the appellant's arguments

The problem solved by the application was to provide a zinc/air cell having an improved voltage and service life discharge profile, as was evident from Figure 3 of the application.

The appellant essentially argued that there was no motivation to modify the electrochemical cells of D3 of D6 so as to arrive at the claimed invention. D1 did not aim to improve the voltage and service life discharge profile of a cell. Although D1 recognised that a chain-forming clay (however not Laponite) may lower the resistance of the anode suspension, this did not necessarily lead to an improved voltage and service life discharge profile. D1 taught to use the chain-forming clay primarily as the gelling agent. Therefore, if the skilled person did turn to D1, he/she would be taught to replace the gelling agent with a clay, rather than providing a gelling agent and an ionically conductive clay.

D2 provided no motivation to use an ionically conductive clay and so the skilled person would not turn to this document. D2 did not relate to mercury-free anodes as in the present invention and in D3. The problems relating to corrosion of zinc and poor cell performance were not addressed in D2. Although D2 disclosed anode coating composition containing a small proportion of Bentone LT, no reason was given as to why this material was present. Therefore, there was no

motivation for the skilled person to attempt to incorporate this material into the cell of D3 in order to solve the problem addressed by the application.

IX. Requests:

The appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the set of claims filed with letter of 11 June 2010 as a main request, or, in the alternative, on the basis of the set of claims filed with letter of 17 May 2010 labelled as second auxiliary request.

Reasons for the Decision

1. *Amendments (main request)*

Claim 1 is based on the disclosure of claims 1 and 9 as originally filed (published as WO-A-02/09 850) and the description, page 5, lines 13, 14, and 18 to 20.

Method claim 7 is based on originally filed claims 14 and the description, page 5, lines 13, 14, and 18 to 20.

Claims 2 to 4 are based on claims 8 to 10 as published.

Claim 5 is based on the description, paragraph [29].

Claim 6 is based on the description, page 5, lines 14 to 16.

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

2. *Article 84 EPC*

Laponite^R appears to be a registered mark of Rockwood Inc. and several types or grades of Laponite appear to exist. The claim is nevertheless considered to be sufficiently clear as to the nature of the said Laponite due to the empirical chemical formula of the clay which is recited in the claim.

3. *Novelty (main request)*

- 3.1 Document D6 discloses alkaline electrochemical cells, for example metal-air cells, having a gelled zinc anode. The preferred gelling agents (carboxypolymethylene [CarbopolTM, a polyacrylate]) are the same as in the present application; the anode mixture optionally contains 0.2% to 12% of mercury.

Mercury-free alkaline cells comprising a gelled zinc anode, indium hydroxide, CarbowaxTM and CarbopolTM (a polyacrylate) as a binder are, however, already known from document D3 (example 1; column 4, lines 12 to 44; column 6, lines 31 to 35).

Having regard to D6 and D3, the claimed subject matter is novel because said documents do not disclose gelled anode mixtures comprising as an additive an ionically conductive clay, and in particular not the synthetic clay Laponite.

- 3.2 D1 discloses certain natural and synthetic clays, such as the kaolinitic clays attapulgite, sepiolite, and palygorskite, as chain-forming additives for metal suspension half-cells comprising zinc and an alkaline

electrolyte. The accumulators disclosed in D1 comprising such metal suspension half-cells differ in structure from the electrochemical cells claimed in the instant application, *inter alia* in that the zinc anode is not a gelled anode mixture, but a viscous, flowable and/or stirrable suspension comprising zinc metal particles. Laponite is not among the list of clays disclosed in D1. Therefore, the subject matter of claim 1 is novel having regard to D1.

- 3.3 D2 discloses the clay Bentone™ LT (an organic derivative of hydrous magnesium aluminium silicate) in an amount of 0.14 wt.-% as a component of a zinc anode composition for a laminar electrochemical cell (see column, 9, lines 1 to 23). Document D2 neither discloses the specific gelling agents specified in the claims of the application under appeal, nor does it disclose Laponite clay as an additive.

D7 relates to a zinc alkaline battery having a gelled anode, but does not mention any clay additives.

- 3.4 The board therefore concludes that the claimed subject matter of claim 1 of the main request is novel having regard to the cited prior art. Independent method claim 7 recites all the features of claim 1, in particular the presence of the Laponite additive; the subject matter of claim 7 as well as of dependent claims 2 to 6 and 8 is therefore novel for the same reasons.

The requirements of Article 54 EPC are met.

4. *Inventive step (main request)*

4.1 The application in suit relates to alkaline electrochemical cells and in particular to the preparation of a mercury-free gelled zinc anode for an electrochemical cell.

4.2 Mercury-free alkaline cells comprising a gelled zinc anode, indium hydroxide and CarbowaxTM (an ethylene oxide polymer) as additives and CarbopolTM (a polyacrylate) as a binder are known from document D3 (example 1; column 4, lines 12 to 44; column 5, lines 52 to 55; column 6, lines 31 to 35).

The board notes that the examining division took D6 (rather than D3) as representing the closest prior art. Said document discloses an alkaline electrochemical cell, for example a metal-air cell, having a gelled zinc anode and the same gelling agents as the application under appeal. According to claim 1 of D6, the anode mixture of D6 contains optionally 0.2% to 12% of mercury. However, in the illustrated embodiments of D6, the anode composition includes mercury as a functional component, in amounts of 0.2 to 12% based on the weight of Zn (3 wt.-% in the working example) (page 6, lines 53 to 57; page 11, line 19). The claimed subject matter thus differs from D6 in that the anode mixture is mercury-free and in that it contains an ionically conductive clay additive. For this reason, the board considers that D6 is further removed from the application under appeal.

According to D3, indium hydroxide and the ethylene oxide polymer are employed to inhibit the corrosion of

zinc and to replace mercury which is conventionally added for this purpose (column 1, lines 29 to 34). The mercury-free alkaline cells of D3 bear a close structural similarity to the ones claimed in the application under appeal. The board therefore regards D3 as the closest prior art.

4.3 The technical problem underlying the application under appeal in the light of D3 is to improve the cell voltage and service life discharge profile of a mercury-free zinc cell of the type disclosed in D3.

4.4 As a solution to the above defined technical problem the application in suit proposes an electrochemical cell according to claim 1 of the main request characterised in that the cell's gelled anode mixture comprises the ionically conductive synthetic clay Laponite having the empirical formula $[\text{Na}_{0.70}^{0.7+}[(\text{Si}_8\text{Mg}_{5.5}\text{Li}_{0.3})\text{O}_{20}(\text{OH})_4]^{0.7-}]$ as an additive.

4.5 It has to be examined whether the technical problem has been actually solved.

According to paragraph 26 of the application, the ionically conductive clay additive improves the transport of hydroxyl ions inside the zinc anode matrix during discharge and, thus, delays anode passivation and decreases polarization, resulting in greater cell performance.

Figure 3 of the application shows a higher cell voltage over a longer service life for a mercury-free Laponite based cell, compared with a cell containing no Laponite. Zinc/air cells containing Laponite clay as an additive

exhibit an improved discharge profile, due to the beneficial effect attributed to the gelled zinc anode.

The board therefore accepts that the above defined technical problem has been solved.

4.6 It remains to be decided whether the claimed solution is obvious in view of the prior art.

4.6.1 The examining division argued in the contested decision that it would have been obvious to the person skilled in the art to use clay additives of the kind disclosed in D1 and D2 in order to improve a battery's discharge behaviour.

4.6.2 These arguments are however not sufficient to deny the presence of an inventive step, having in particular regard to the claims as amended during appeal proceedings, for the following reasons.

Document D1 discloses certain natural and synthetic clays, such as the kaolinitic clays attapulgite, sepiolite, and palygorskite as gel-forming additives for zinc anode compositions of a metal suspension half-cell. These clays are described in D1 as having the effects of counteracting self-discharge, increasing charge transfer between the metal particles and increasing conductivity within the anode (column 2, lines 13 to 27; col. 12, lines 5 to 45; column 3, line 50 to column 4, line 35; claims 4 to 6).

Increasing conductivity within the zinc anode at low zinc concentrations and restricted zinc corrosion and self-discharge were also observed (column 12, lines 32 to 51; Figures 10, 11) in connection with the clay

additive palygorskite, compared with cells containing boron carbide.

- 4.6.3 D1 is concerned with metal suspension half cells and accumulators containing same. The board observes that a zinc metal suspension half cell is fundamentally different from the gelled zinc anode employed in the application under appeal in that the zinc in the anode is present as a suspension. It is therefore doubtful whether the skilled person would have considered D1 to be pertinent prior art in view of the problem posed, which relates to mercury-free alkaline cells having a gelled zinc anode.

But even if D1 had been taken into account, the combination of D3 and D1 would not lead to the claimed subject matter as D1 teaches to replace the gelling agent with a clay, rather than providing a gelling agent in addition to an ionically conductive clay. Furthermore, D1 does not disclose or suggest the particular synthetic clay Laponite as an ionically conductive clay additive.

The contested decision states that a decreased self-discharge would evidently result in a longer service life since charge losses due to parasitic effects were reduced and that these beneficial effects were to be expected not only in rechargeable accumulators with a metal suspension half cell, such as disclosed in D1, but also in gelled zinc anode batteries.

However, although the various clays used according to D1 are described to counteract self-discharge, to increase charge transfer between the suspended metal

particles and to increase conductivity within the anode suspension, these effects are not necessarily linked to a voltage increase and to a prolonged service life (improved discharge profile) of a gelled anode battery.

- 4.6.4 As mentioned above, D2 discloses the clay BentoneTM LT (an organic derivative of hydrous magnesium aluminium silicate) in an amount of 0.14 wt.-% as a component of a zinc anode composition for a laminar electrochemical cell (see column, 9, lines 1 to 23). Laponite clay is not disclosed.

Laponite^R, the preferred clay to be used according to the present application, is a layered silicate which also acts as a thickener and is insofar similar to the modified clay BentoneTM LT used in accordance with document D2. D2 is, however, silent on the purpose and effect of BentoneTM LT in the anode mixture. In particular, there is no suggestion in D2 that BentoneTM LT (or Laponite) improves the discharge voltage and the service life of a battery. Therefore, the skilled person would not have been incited by D2 to add BentoneTM LT to the zinc anode mixture, for the purpose of the present invention, let alone to replace BentoneTM LT with Laponite. The object of D2 is to facilitate the mass production of multiple cell laminar batteries, and to reduce the number and complexity of process steps involved (D2, column 1, line 68 to column 2, line 3).

- 4.6.5 D7 discloses a mercury-free alkaline battery comprising a gelled zinc anode, a fibrous material mixed with the alkaline electrolyte and indium oxide adhered to the zinc powder to improve corrosion and discharge behaviour. This document is representative of the prior

art technology of using a metal additive (indium oxide) to increase the performance of the cell. There is no suggestion of a clay additive, so that D7 cannot render the claimed subject matter obvious.

4.6.6 Starting from D6 as closest prior art, the technical problem would be identical to the one defined above under point 4.3. The reasoning would be *mutatis mutandis* the same as under points 4.6.2 to 4.6.5. In summary, neither of D1 and D2 points to the beneficial effects of Laponite clay on discharge voltage and service life.

4.7 In conclusion, the subject matter of claim 1 of the main request is not derivable in an obvious manner from the prior art.

The independent method claim recites all the product features of the electrochemical cell claimed in claim 1 and thus involves an inventive step for the same reasons given in respect of the product claim.

The dependent claims 2 to 6 and 8 define preferred embodiments of the inventive electrochemical cell and the method of its manufacture, respectively. These claims therefore also involve an inventive step.

The requirements of Article 56 EPC are thus met.

4.8 As the main request can be allowed, there is no need to consider the auxiliary request.

4.9 The appellant has provisionally requested oral proceedings only in the event that a decision be

reached other than acceptance of the main request (letter of 2 March 2007). Therefore, the present decision can be delivered on the basis of the written submissions. According to the appellant's suggestion expressed in its telefax of 11 June 2010, oral proceedings initially appointed for 16 June 2010 have been cancelled.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the department of first instance with the order to grant a patent on the basis of the following application documents:
 - Claims 1 to 8 (main request),
 - description, pages 1, 2, 5 to 7,
 - Figure 1,all filed with letter of 11 June 2010;
 - description, pages 3 and 4, and Figures 2 and 3, as published.

The Registrar

The Chairman

C. Vodz

G. Rath