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D E C I S I O N
of 5 November 1997

Case Number: T 0031/95 - 3.4.2

Application Number: 87118066.7

Publication Number: 0271043

IPC: H01M 10/34, H01M 4/24, H01M 4/38, H01M 4/48

Language of the proceedings: EN

Title of invention:
Sealed storage battery and method for making its electrode

Patentee:
MATSUSHITA ELECTRIC INDUSTRIAL CO., LTD.

Opponent:
Alcatel Alsthom Compagnie Générale d'Electricité
Eveready Battery Company

Headword:
-

Relevant legal provisions:
EPC Art. 56, 123(2)

Keyword:
"Inventive step (main request and third auxiliary request - no)"
"Amendments - added subject-matter (first and second auxiliary request - yes)"

Decisions cited:
-

Catchword:
-

Case Number: T 0031/95 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 5 November 1997

Appellant: (Proprietor of the patent)
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Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted 2 November 1994 revoking European patent No. 0 271 043 pursuant to Article 102(1) EPC.**

Composition of the Board:

Chairman: E. Turrini
Members: R. Zottmann
L. C. Mancini

Summary of Facts and Submissions

I. The Appellant (Patentee) lodged an appeal against the decision of the Opposition Division on the revocation of the patent No. 0 271 043 with the application No. 87 118 066.7.

The oppositions were based on the grounds of lack of novelty and lack of inventive step (Article 100(a) EPC).

The Opposition Division held that the claims did not fulfill the requirements of Articles 52(1) and 56 EPC for inventive step.

II. The following documents were among others cited during the appeal procedure:

D1: French translation (pages 1 to 8) of JP-A-61-233 969, provided by Respondent I (Opponent I);

D4: US-A-4 487 817;

D6: English translation (pages 1 to 8) of JP-A-60-70 154, provided by Respondent II (Opponent II);

D12: English translation (pages 1 to 8) of JP-A-61-176 063, provided by Respondent I;

IV. Oral proceedings were held at the end of which the Appellant requested that the decision under appeal be set aside and the patent be maintained in amended form on the basis of a main request or three auxiliary requests.

The Respondents I and II requested that the appeal be dismissed.

V. Claim 1 of the main request reads as follows:

"1. A sealed storage battery comprising:

(a) a positive electrode including a metallic oxide;

(b) a negative electrode including a hydrogen-occlusion-alloy powder containing cobalt (Co), nickel (Ni) and a mixture of rare earth elements, having a crystal structure of $CaCu_5$ type, having a SF-value of 2.5 or below, obtained by rapid cooling of the melted alloy mixture in a cooling apparatus and subsequent heat treatment, and being provided with numerous irregularities on a surface obtained by an acid and/or alkali treatment of the alloy powder, said SF-value representing a plateau characteristic of equilibrium pressure of hydrogen releasing and being calculated by the following formula:

$$SF = \ln(P_{H/M=0.75}/P_{H/M=0.25})$$

wherein $P_{H/M=0.75}$ is the pressure of hydrogen when occluding 75% of hydrogen and $P_{H/M=0.25}$ is the pressure of hydrogen when occluding 25% of hydrogen;

(c) an alkaline electrolyte; and

(d) a separator for separating said positive electrode from said negative electrode, and for absorbing and holding said alkaline electrolyte."

Claim 1 of the first auxiliary request differs from claim 1 of the main request only in that "having a SF-value of 2.5 or below"

is replaced by

"having a SF-value of 2.0 or below".

Claim 1 of the second auxiliary request differs from claim 1 of the main request only in that

"obtained by rapid cooling of the melted alloy mixture in a cooling apparatus and subsequent heat treatment"

is replaced by

"obtained by melting the alloy and transferring it to a cooling apparatus where the alloy is rapidly cooled and by subsequent heat treatment".

Claim 1 of the third auxiliary request reads as follows:

"1. A method for making a sealed storage battery by providing:

(a) a positive electrode including a metallic oxide;

(b) a negative electrode including a hydrogen-occlusion-alloy powder containing cobalt (Co), nickel (Ni) and a mixture of rare earth elements, having a crystal structure of $CaCu_5$ type, having a SF-value of 2.5 or below and being provided with numerous irregularities on a surface, said SF-value representing a plateau characteristic of equilibrium pressure of hydrogen releasing and being calculated by the following formula:

$$SF = \ln(P_{H/M=0.75}/P_{H/M=0.25})$$

wherein $P_{H/M=0.75}$ is the pressure of hydrogen when occluding 75% of hydrogen and $P_{H/M=0.25}$ is the pressure of hydrogen when occluding 25% of hydrogen;

(c) an alkaline electrolyte; and

(d) a separator for separating said positive electrode from said negative electrode, and for absorbing and holding said alkaline electrolyte, in which method the provision of the negative electrode comprises the steps of rapid cooling of the melted alloy mixture in a cooling apparatus and subsequent heat treatment, and the subsequent step of an acid and/or alkali treatment of the alloy powder."

VI. The Appellant's arguing is summarized as follows:

The SF-value (slope factor value) of 2.0 of claim 1 of the first auxiliary request can be taken from Figure 6.

The problems underlying the invention are: maintenance of a high capacity, no generation of hydrogen during operation, high corrosion resistance of the hydrogen-occlusion electrode against oxygen. The essential features of the invention to solve these problems are a low SF-value obtained by rapid cooling, irregularities obtained by alkali and/or acid treatment and Co as a mandatory component of the alloy. The low SF-value brings about an improved crystallinity of the alloy.

A combination of the teachings of D1 and D12 is not obvious since the basic problems are different: To retard dissolution of Mn, D1 seeks to replace Mn by a more suitable element; the method of D12 aims at washing out unalloyed elements precipitating on the separator and worsening its insulating. Even if the skilled person considered both documents, he would obtain no indication as to how he could arrive at the invention. The combination of the rapid cooling step and acid/alkali treatment for ensuring both the structural and physical characteristics of the invention cannot be deduced from said prior art documents.

D1 and D12 do not relate to **sealed** storage batteries. In this respect, the translation of D1 by Respondent I is incorrect. Neither D1 nor D12 disclose the SF-value range and cooling step of the invention. The furnace crucibles of the arc furnaces of said prior art give a temperature decrease of ca. 10 ^{0}C/s ("natural" or "slow" cooling), whereas the cooling according to the patent-in-suit is ca. 100 ^{0}C/s ("active" cooling). In D1 no material for the furnace is disclosed. In D1 eleven elements including Co are mentioned as possible components besides RE metals and Ni. Apart from the Co-containing alloys, there are 13 other examples of alloys listed in the table of D1 not comprising Co.

D4 represents the closest prior art above all since it addresses the oxygen corrosion problem. However, this patent tries to solve this problem with alloys comprising at least one element selected from Al, Cr and Si and does not provide a rapid cooling step and an alkali/acid treatment step during manufacture of the electrode.

Some of the results of Respondent II are of no relevance since the data were measured at different temperatures and at temperatures different from that of the tests of Respondent I. An SF-value lower than 2.5 with the cooling of D1 can be obtained only by chance. Own tests (annex to the letters dated 15 November 1996 and 5 September 1997) indicate that annealing treatment itself shows little effect regarding a reduction of the SF-value. A drastic reduction is achieved only by a combination of rapid cooling and annealing.

VII. The arguing of Respondent I is summarized as follows:

D1 and D12 use quite similar alloys and treat them in a quite similar manner during manufacture and, therefore a combination of said documents is obvious. All features of the independent claims can be deduced from such a combination.

Attempts according to the manufacturing conditions of D1 and with six of the 22 alloy samples of D1 with a good performance show that all yield an SF-value lower than 2.5, all but one case even an SF-value lower than 2.0. An SF-value lower than 2.5 is thus an inherent property of the alloys of D1. The preparation of the alloys in an arc furnace necessarily implies a rapid cooling of the alloy when the arc is stopped. The expression "in a cooling apparatus" is an unimportant detail which does not have any detectable influence on the properties of the alloy. The objection of the Appellant that SF-values were determined by Respondent II at different temperatures is irrelevant in view of the fact that the patent-in-suit does not disclose any temperature value for the measurement of SF.

Before a hydrogen-occlusion electrode is used in a sealed battery it is tested in an open battery. Such electrodes were developed particularly for sealed batteries.

VIII. The arguing of Respondent II is summarized as follows:

For example claim 1 infringes Article 123(3) EPC, as its category has changed from product to product-by-process by introducing the rapid cooling step. The term "rapid cooling" cannot be divorced from "while stirring" and therefore the latter term has to be introduced into the claim. An upper limit of 2.0 of the SF-value range cannot be taken from the patent-in-suit. Thus the corresponding claims do not comply with Article 123(2) EPC. The value 2.0 for the upper limit of SF is nowhere disclosed in the patent-in-suit.

The Appellant stresses the importance of the SF-value. However, it is not disclosed in the patent at what temperature SF is calculated and what is meant by 25 and 75% hydrogen occlusion. Very different values can be determined, depending on whether practical hydrogen occlusion or theoretical hydrogen should be used to calculate the SF-value.

As far as combining D1 and D12 is concerned, there can be no logical reason for the skilled person not to combine documents in the same field. Even if D1 and D12 do not mention that they concern sealed batteries or electrodes for such batteries, it is abundantly clear to the skilled person that hydrogen-occlusion electrodes can only be used in sealed batteries. Otherwise gas would escape. Moreover, such electrodes were developed particularly for sealed storage batteries. Arc melting entails a fast quench of the alloy when the arc is stopped. An arc furnace must contain water cooling tubes. It is usually made of copper which has a melting temperature of 960°C and would melt during the operation of the furnace unless it has a cooling apparatus like water tubes since the melting temperatures of the mixtures used in D12 and D1 are at least 1250°C.

Tests with all alloys of D1 give hydrogen-occlusion electrodes with an SF-value lower than 2.5, mostly much lower than said value.

Reasons for the Decision

1. The appeal is admissible.

2. *Main Request*

2.1 Amendments

Claim 1 has been amended with respect to claim 1 as granted by inserting the features that the indicated SF-value is obtained by rapid cooling of the melted alloy mixture in a cooling apparatus and subsequent heat treatment and by inserting that the **alloy powder** is treated with acid and/or alkali.

It is established case law that a **product** can be defined by its process of preparation (see e.g. decisions T 0150/82, EPO OJ 1984, 309, T 0248/85, EPO OJ 1986, 261 or T 0552/91, EPO OJ 1995, 100). Hence it follows that a product claim into which process features were inserted remains a product claim.

Since these amendments lead to a restricted claim and do not change its category, claim 1 complies with Article 123(3) EPC.

Only lines 6 to 12 of page 13 as originally filed disclose the feature that the melted alloy is rapidly cooled in a cooling apparatus. According to this passage, the melted metal "mixture is transferred to a container having a cooling apparatus while stirring, and then, the mixture is rapidly cooled". Thus, the stirring of the mixture is closely linked with the transfer step but not with the following cooling step. Therefore, when the cooling step is brought into claim 1, "while stirring" may not be inserted into claim 1. When, however, the transfer step is part of the amended claim 1 - as this is the case with claim 1 of the second auxiliary request - "while stirring" has to be inserted together with the transfer step.

Treatment of the alloy powder with acid and/or alkali is disclosed e. g. in the first paragraph of page 19 as originally filed.

Therefore, said amendments comply also with Article 123(2) EPC.

2.2 Inventive step of claim 1

2.2.1 Document D12 is considered as representing the nearest prior art with respect to the subject-matter of claim 1. The prior art of D4 does not come nearer than D12, see for example the last but one paragraph of section V. above.

D12 describes a long-life alkaline storage cell (see claim 1 and page 8 lines 14 to 17) employable in particular for sealed storage cells (see page 8 in the middle) and with a hydrogen-occlusion electrode as the negative electrode. The negative electrode is combined with a known nickel electrode as the positive electrode via a separator to constitute the cell (see page 2 lower part). Known nickel electrodes for such alkaline cells mostly comprise nickel. In sealed storage cells the separator separates the electrodes and regularly serves to absorb and hold the alkaline electrolyte. In the examples $LaNi_{2.5}Co_{2.5}$, a hydrogen-occlusion alloy of the $CaCu_5$ type, is used.

The alloy powder is treated with an aqueous alkaline solution in the same manner as described in the attacked patent (compare Examples 1 and 2 of D12 with the examples of the attacked patent). The alkali treatment is carried out to remove metals soluble by the electrolyte from the alloy powder in order to prevent their precipitation during operation of the cell and to provide a cell having a long life and stable quality (see page 3, first paragraph). Such a treatment necessarily produces, as in the attacked patent, numerous irregularities on the surface of the powder, all the more when using the alloys of D1 which are composed of "pluralized" alloys rather tending to a segregation of the components than the alloy of D12.

From the foregoing follows that the cell of claim 1 of the attacked patent differs from D12 mainly in that the latter document does not disclose a **mixture** of RE metals as one component of the hydrogen-occlusion alloy and the annealing step. Furthermore, the SF-value of the alloy powder is not discussed in D12 and thus not explicitly described there.

The storage cell of D12 has a prolonged cycle life. However, it is well-known in the technical field that pure La is expensive (see for example D6, page 2 at the bottom to page 3 line 4) and that mixtures of RE metals are advantageous under economic aspects.

2.2.2 Thus the problem underlying the subject-matter of claim 1 is, therefore, when starting from prior art D12, to further develop the battery according to this document in such a way that an expensive component of the hydrogen-occlusion alloy need not be used while the high capacity and long cycle life of the battery is at least maintained. The problem of a low corrosion resistance of the alloy against oxygen is not mentioned, cannot be easily derived from D12 and is, moreover, included in the more general problem of increasing or at least maintaining a long cycle life.

2.2.3 According to the single claim and the general description of D12, the negative electrode is made of a powder of a hydrogen-occlusion alloy; it is not restricted to a certain composition of the alloy. As a matter of course, in the **examples** a certain hydrogen-occlusion alloy is used. Thus, though $LaNi_{2.5}Co_{2.5}$ is the only alloy mentioned on D12, the skilled person is not bound to such a choice.

To solve this problem, the skilled person would take into account document D1 since it describes a battery quite similar to that of D12, has a prolonged cycle life and high capacity (see last paragraph of D1) and uses misch metal, a mixture of RE metals which can be acquired cheaply (see for example D6 page 3 second paragraph). When choosing a hydrogen-occlusion alloy mixture of D1, he would prefer to adopt the method for preparation of the alloy as disclosed in D1; however, he would maintain the alkali treatment step since it remains the requirement of removing material precipitating in the separator, all the more with the alloys of D1. According to D1 the alloy mixture is melted several times in an arc furnace and annealed at 1000 ^{0}C (see page 4). In a table (on page 5) 22 preferred hydrogen-occlusion alloys are listed. All of said alloys contain misch metal, nine of them Co. Moreover, the alloy of the examples of D12 contains Co, too. D1 explicitly discloses that the alloy powder has a crystal structure of the $CaCu_5$ type.

Melting in an arc furnace produces rapid cooling of the molten alloy when the power is shut off. It is different from slow cooling in a classical furnace. Additionally, as opposed to a high-frequency furnace, an arc furnace has a metallic crucible for the material to be melted, preferably consisting of copper, and cooling tubes. Due to the high heat conductivity of the crucible and due to the cooling means, the crucible functions as and is deemed to be a cooling apparatus. Moreover, when comparing the cooling step of the attacked claim 1 and the cooling of the alloy in an arc furnace according to D1, it has to be taken into account that it is not disclosed in the patent-in-suit which temperature decrease per time is to be defined by "rapid cooling in a cooling apparatus". A more precise definition of said step by taking into account the condition that the SF-value has a certain upper limit is impossible, since the definition of said value in the patent is very vague. The reader of the patent is only told that the slope factor is measured at a "given temperature" and he is not told whether the amounts of 25% and 75% are relative to the amount of hydrogen which the electrode material measurably absorbs, or whether these quantities are relative to the amount of hydrogen that the material can theoretically absorb. The tests of the Respondents demonstrate that the SF-value depends considerably on these conditions.

The SF-values of the alloys are discussed neither in D1 nor in D12. Therefore, the feature of claim 1 that the SF-value is at the utmost 2.5 is not explicitly disclosed there. However, the tests supplied by Respondent I (see the letter dated 7 August 1995) and Respondent II (see the letter dated 17 October 1995), all being prepared according to D1, yield SF-values below 2.0 when using high temperature desorption and below 2.5 without high temperature desorption. The Appellant argues that the SF-value is dependent on the temperature and thus the measurement of Respondent II at different temperatures are irrelevant. However, the patent-in-suit is silent about the temperature at which the SF-value has to be measured. Moreover, the skilled person is aware of the fact that a low SF-value, in other words a plateau with a low gradient, is advantageous to achieve a high energy density. He would select those alloys and production steps leading to the plateau with a lower slope.

The Appellant alleges that his experiments (according to the annexes of the letters dated 15 November 1996 and 5 September 1997) show that the annealing step alone does not lead to considerable and sufficient reductions of the SF-value, see tests a, b and c according to Figure 1 of said annexes. However, most of the conditions for the production of the alloys of said tests are not mentioned. Moreover, the only tests of the remaining tests d through o carried out with an annealing step but without rapid cooling (i, m, n and o; indicated in lines 2 and 9 to 11 of the annexed table) give SF-values of ca. 2 or below 2, and comparison of test k (Figure 3; line 1 of said table; without annealing and an SF-value of 3.893) and test i (Figure 3; line 2 of said table; with annealing and an SF-value of 2.045) demonstrates that the annealing step does reduce the SF-value.

Therefore, the subject-matter of claim 1 does not involve an inventive step within the meaning of Article 56 EPC.

3. *First auxiliary request*

Claim 1 differs from claim 1 of the main request only in that the upper limit of the SF-value, namely 2.5, is replaced by 2.0. It is nowhere mentioned in the description or in the claims as originally filed that the upper limit of the SF-value should be 2.0, whereas the application as originally filed mentions at several places that an upper limit of 2.5 is of importance for the subject-matter of the patent (see page 15 at the top, page 19 at the top and claims 1 and 6).

A value of 2.0 for SF is mentioned on original page 16 at the bottom (" ... alloys having following SF-value are selected, i. e. 1.5, 2.0, 2.3 and 2.48."). However, it represents only a selection of alloys having certain SF-values below the preferred value of 2.5. This does not mean that the choice of a value of 2.0 as upper limit is advantageous with respect to other values.

Figure 6 is a graph showing a relation between the SF-value of alloys and the number of charge-discharge cycles (until the capacity of the battery deteriorates to a certain extent). SF-values between 1.5 and ca. 3.4 occur, among them also an SF-value of 2.0. The curve trace is roughly approximated by straight lines connecting adjacent points. The real curve trace around the point corresponding to an SF-value of 2.0 is indefinite since the adjacent points (corresponding to SF=1.5 and 2.3) are relatively far away. Therefore, it cannot be derived from Figure 6 that an SF-value of 2.0 is a preferred upper limit.

As a consequence, claim 1 is not allowable under Article 123(2) EPC.

Moreover, not only the SF-value of 2.5 but also the SF-value of 2.0 or less can easily be obtained with electrodes resulting from a combination of the teachings of D1 and D12, see section 2.2.3 above. Therefore, the lowering of the SF-value from 2.5 to 2.0 does not confer inventive step to the subject-matter of claim 1 of the first auxiliary request .

4. *Second auxiliary request*

Claim 1 of the second auxiliary request differs from claim 1 as granted above all in that the following features have been inserted: The indicated SF-value is obtained by transferring the alloy to a cooling apparatus where it is rapidly cooled and by subsequent heat treatment. As already put forward in paragraph 4 of section 2.1 above, insertion of the transfer step into claim 1 as granted while suppressing "while stirring" leads to a claim which extends beyond the content of the application as originally filed.

Claim 1 is thus not allowable under Article 123(2) EPC.

5. *Third auxiliary request*

The features of (method) claim 1 correspond to the features of (apparatus) claim 1 of the main request except for the following differences. The cooling, annealing and acid/alkali treatment steps are not correlated with properties of the alloy. This leads to a more general claim with respect to claim 1 of the main request. Additionally, claim 1 of the third auxiliary request indicates that the acid and/or alkali treatment is **subsequent** to the cooling and heat treatment. Such a sequence is considered as being evident since the elution of disturbing elements at the surface of the alloy makes sense only after formation of the solid alloy.

Therefore, the conclusions with respect to claim 1 of the main request apply to claim 1 of the third auxiliary request, too.

6. Since none of the claims 1 of the four requests is allowable, it is not necessary to examine the remaining

claims of the requests. As a consequence, none of the requests is allowable.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

P. Martorana

The Chairman:

E. Turrini