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D E C I S I O N
of 7 April 2000

Case Number: T 0926/95 - 3.2.2

Application Number: 91902344.0

Publication Number: 0502127

IPC: C01B 6/00

Language of the proceedings: EN

Title of invention:
Catalytic hydrogen storage electrode materials for use in electrochemical cells incorporating the materials

Appplicant:
ENERGY CONVERSION DEVICES INC.

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56

Keyword:
"Novelty (yes, after amendment)"
"Inventive step (yes, after amendment)"

Decisions cited:
-

Catchword:
-

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Case Number: T 0926/95 - 3.2.2

D E C I S I O N
of the Technical Board of Appeal 3.2.2
of 7 April 2000

Appellant: ENERGY CONVERSION DEVICES, INC.
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 10 July 1995 refusing European patent application No. 91 902 344.0 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: W. D. Weiß
Members: R. Ries
C. Holtz

Summary of Facts and Submissions

I. The present appeal is against the decision of the examining division to refuse European patent application No. 91 902 344.0 (WO91/08167). The examining division reasoned that independent claim 1 of the main request then under consideration lacked novelty and that the alternative claim 1 according to the auxiliary request lacked an inventive step. During examination, reference was made inter alia to the following documents:

D1: US-A-4 849 205

D2: DE-A-3 023 770

D3: US-A-4 728 586

II. In three communications dated 17 November 1998, 17 June 1999 and 11 January 2000 the Board considered document D1 as pertinent prior art and additionally referred to documents

D4: US-A-4 551 400 and

D5: Titanium '95, Science and Technology, Proc. of the Eighth World Conf. on Titanium, Birmingham, 22 to 26 October 1995, The Institute of Metals, pages 1429 to 1437

III. In response, the appellant referred to document

D6: Metals Handbook Desk Edition, American Society for Metals, Chapter 14: "Pure Metals", pages 14-1 to 14-3

and submitted amended sets of claims.

In the appellant's view, the claimed alloy composition is novel with respect to the alloys disclosed in documents D1 to D4. More specifically, alloy No. 5 of group 2 in Table 1 of document D1 comprises 3.2 at% Cu as a modifying agent, which is outside the claimed range of 5 to 30% Cu. Moreover, alloy No. 6 of group 3 of document D1 fails to comprise Cr which is, however, a compulsory component in the claimed alloy to form the intergranular "V-Cr"-phase. Document D4 essentially relates to alloys comprising only four components rather than five elements as claimed. Although document D3 discloses Ni-V-Ti-Zr-Cr alloys and in particular the "control alloy" $Ti_{16}Zr_{16}V_{22}Ni_{39}Cr_7$ used in the invention, it does not disclose nor suggest specifically alloy compositions additionally comprising one or more modifying agents within a range of 5 to 30 at% as does the composition now claimed.

IV. At the oral proceedings held on 4 April 2000, the appellant requested that

- the decision under appeal be set aside and

- the patent be granted on the basis of claims 1 to 8 as submitted in the oral proceedings.

The appellant's former request for reimbursement of the appeal fee was withdrawn.

The wording of claim 1 reads as follows:

"1. Multicomponent and multiphase electrochemical hydrogen storage alloy of the type V-Ti-Zr-Ni-Cr alloy, comprising an intergranular phase of vanadium and chromium, of the following compositions:

$$(V_{y'-y}\,Ni_y\,Ti_{x'-x}\,Zr_x\,Cr_z)_a\,M_{100-a}$$

where $1.80 \leq x' \leq 2.20$
$0.00 < x \leq 1.50$
$3.60 \leq y' \leq 4.40$
$0.60 \leq y \leq 3.50$
$0.00 < z \leq 1.44$ and
$70 \leq a \leq 95$

wherein M is at least one modifying element chosen from the group consisting of:

Al, Si, Mn, Fe, Co, Cu, Mo, W, Sn, Zn,

thereby impeding the formation of the V-Cr phase."

Reasons for the Decision

1. The appeal complies with the provisions mentioned in Rule 65(1) EPC and is, therefore, admissible.

2. *Amendments*

Present claim 1 derives from claim 1 as originally filed and the subject matter present in pages 25 to 27 of the description. It is apparent from page 26, lines 5 to 15 that Ti and Zr are both present in the claimed alloy thus justifying the lower limit of > 0.00 for zirconium. The range of $70 \leq a \leq 95$ for the modifying elements is disclosed on page 26, last paragraph bridging page 27, paragraph 1.

The existence of the intergranular "V-Cr"-phase in the five-component Ni-V-Ti-Zr-Cr host alloy is amply described on page 32, last paragraph bridging page 33, paragraph 1, on page 44, line 14 bridging page 45, paragraph 1 and on page 70. It is immediately evident from the application taken as whole that, in order for

the intergranular "V-Cr"-phase to exist in the Ni-V-Ti-Zr-Cr base alloy, chromium as a component must be deliberately added in sufficient amounts which clearly differ from the impurity level of Cr introduced by the other components.

Dependent claims 2 and 3 find support on page 26, last paragraph bridging page 27 paragraph 1, and dependent claims 4 to 8 correspond to former claims 4, 5, 7, 11 and 12 as filed.

The requirements of Article 123(2) EPC are therefore satisfied.

3. *Clarity*

The wording of present claim 1 makes it clear that the claimed alloy consists of two parts: (i) a five-component (V-Ni-Ti-Zr-Cr) base alloy and (ii) at least one modifying element, both adding up to 100 at%. Given that the V-Ni-Ti-Zr-Cr basic alloy, taken separately, actually comprises a "V-Cr"-phase, chromium must be present in amounts which are effective to form the "V-Cr"-phase. Moreover, it is clear from the basic composition and the description that zirconium is deliberately added and, therefore, is present in amounts higher than zero. Claim 1 in its present form, therefore, meets the requirements of Article 84 EPC.

The compositions of the preferred embodiments of the claimed alloy featuring in the dependent claims 2 to 8 are also clear and likewise satisfy the requirements of Article 84 EPC.

4. *Novelty*

Document D1 discloses in column 8, lines 10 to 28 a group of multicomponent hydrogen storage alloys having high capacity, long life cycles and good rate capability. The alloys are represented by the formula $Ti_aCr_bZr_cNi_dV_{3-a-b-c-d}M_x$ with $0 \leq x \leq 0.2$ and M standing for Al, Si, Mn, Co, Cu, Fe, Nb and rare earth metals. Converted into $\Sigma = 100$ atomic percent, the maximum amount of $M_{0.2}$ in the formula corresponds to max. 6.25 at% M. Compared with the claimed alloy which requires one or more modifiers M in the range of 5 to 30 at%, a small range of overlap exists. Turning to the examples given in Table 1 of document D1, only example 5 of group 2 discloses a five-component alloy $Ni_{0.7}V_{1.2}Ti_{0.3}Zr_{0.5}Cr_{0.3}Cu_{0.1}$ which includes copper as a modifying element. Recalculated to give a total of 100 at%, this composition can be written as

$Ni_{22.58}V_{38.71}Ti_{9.68}Zr_{16.13}Cr_{9,68}Cu_{3,23}$ or
$(Ni_{22.33}V_{40}Ti_{10}Zr_{16.67}Cr_{10})_{96.77}Cu_{3,23}$.

Therefore, the copper content of 3.23 at% falls outside the range of 5 to 30 at% Cu specified in claim 1 of the application.

As to alloy No. 6 of group 3 in Table 1 ($Ti_{0.5}Zr_{0.5}Ni_{1.1}V_{0.7}Cu_{0.2}$), it does not comprise chromium and is, therefore, also outside the claimed compositions. Hence, the alloy compositions disclosed in document D1 do not anticipate the claimed alloys.

Document D4 relates to hydrogen storage alloys represented by the formulae

$(TiV_{2-x}N_x)_{1-y}M_y$, (M = Al or Zr)
$Ti_{2-x}Zr_xV_{4-y}Ni_y$
$Ti_{1-x}Cr_xV_{2-y}Ni_y$

thus consisting of four components rather than five plus one or more modifiers as claimed (cf. in particular D4, column 5, lines 26 to 33, column 6, lines 56 to 63, column 7, lines, 40 to 47, Tables 1 to 3).

Document D3 discloses alloy compositions represented by the formula $(Ti_{2-x}Zr_xV_{4-y}N_y)_{1-z}Cr_z$ with Cr being the preferred modifier and including the claimed "control alloy" $Ti_{16}Zr_{16}V_{22}Ni_{39}Cr_7$ (cf. D3, column 11, lines 9 to 43). However, also document D3 does not specifically disclose Ti-Zr-Ni-V-Cr alloys which additionally comprise one or more of the modifying elements listed in claim 1 of the application.

The remaining documents D2, D5 and D6 are more remote and likewise do not anticipate the hydrogen storage alloys claimed in claim 1 of the present application.

Consequently, the subject matter of claim 1 is novel.

5. *Inventive step*

Document D3 describes in detail the polycrystalline structure and the intergranular phases formed during solidification of the unmodified "control alloy" $Ti_{16}Zr_{16}V_{22}Ni_{39}Cr_7$ which is used in the majority of the examples as a starting material in the present patent application. Therefore, this document which is already amply acknowledged in the description represents the closest prior art (cf. page 11, last paragraph bridging page 12, paragraph 2, page 33, second paragraph and page 43, third paragraph). More specifically, chromium is said to be the preferred (modifying) component to bring about a marked corrosion prevention for the high concentrations of easily corrodible vanadium metal (cf. D3, column 11, line 18, column 12, lines 3 to 9).

However, document D3 fails to realize the presence of the "V-Cr"-phase and its deleterious effect upon the properties of the hydrogen storage alloys.

Starting from this prior art, the problem underlying the present application resides in further improving the overall electrochemical performance of the five-component Ti-Zr-V-Ni-Cr hydrogen storage alloy known from document D3.

The solution to this problem consists in redesigning the basic alloys by the addition of one or more modifying elements selected from the group of Al, Si, Mn, Fe, Co, Cu, Mo, W, Sn or Zn in specific amounts. A list of the overall properties which are improved at least in part by the modified storage alloys according to the invention is given on page 30 of the specification. Specifically, Al, Mn, Mo, Cu, W, Fe, Co and the combination of Co-Mn-Al have been identified as being appropriate modifiers for improving the discharge rate capability, and their presence results in one or more properties including higher midpoint voltage on discharge, decreased polarization during flooded half-cell testing and lower internal resistance when tested in a sealed cell (cf. page 32, paragraphs 1 and 2 of the specification).
In the present application, Cr was found to promote the precipitation of the "V-Cr"-phase during solidification. This "undesirable" phase, however, binds and, therefore, inactivates large amounts of the hydrogen storing element vanadium. Moreover, the hydrogen stored in the "V-Cr"-phase is more prone to damage by oxidation and corrosion. In addition, cycle life is deleteriously affected by the "V-Cr"-phase. Hence, the presence of the "V-Cr"-phase was found to impair the overall hydrogen storage properties rather than improve them.

However, by adding the modifying elements in amounts between 5 to 30 at% to the basic Ti-Zr-Ni-V-Cr alloys as proposed by the present invention, it is possible to minimize or substantially eliminate the "V-Cr"-phase and to improve the discharge voltage and the overall hydrogen storage properties. Reference is made in this context to the application, page 32, last paragraph, bridging page 33, paragraph 1, page 44, last paragraph bridging page 45, paragraph 1, and page 70. Although document D3 in column 12, lines 37 to 45 and also in claims 9, 32 and 55 already mentions a "second modifier" selected from one or more metals of the group Cu, Fe, Mn, Co that may partially substitute for the Ni, and one or more metals of the group Mg, Co, La, Nb, Si and Hf that may partially substitute for the Ti and/or Zr, no information is found anywhere in the this document about the effect associated with the presence of one or more of these elements. Moreover, document D3 neither proposes specific elemental ranges for the "second modifiers" nor discloses any specific example relating to such an alloy. Consequently, document D3 fails to give a clear and unmistakable direction to a skilled person to design the claimed hydrogen storage alloys in view of the problem to be solved by the present invention.

Also document D1 teaches away from the claimed alloy composition since the elemental range for the modifying components in the second group of materials represented by the formula $Ti_aCr_bZr_cNi_dV_{3-a-b-c-d}M_x$ is restricted to at most 6.25%, including 3.23% Cu as an example which is, however, far outside the claimed range.

The remaining documents are even more remote since the problem addressed by the invention is not realized in any of them.

6. Given this situation, the subject matter of claim 1 is novel and involves an inventive step with respect to the prior art represented by documents D1 to D6.

The dependent claims relate to preferred embodiments of the alloy compositions defined in claim 1 and are, therefore, likewise allowable.

7. Having regard to Article 111(1) EPC the Board considers it appropriate to exercise favourably the power of the examining division in the present case because it is satisfied that the claims according to the request of the appellant meet the requirements of the European Patent Convention.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to grant a patent on the basis of claims 1 to 8 as submitted in the oral proceedings on 7 April 2000 and a description and Figures to be adapted thereto.

The Registrar: The Chairman:

V. Commare W. D. Weiß

Checked
R. Rui.

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