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D E C I S I O N
of 14 June 2002

Case Number: T 0318/00 - 3.2.2

Application Number: 95307858.1

Publication Number: 0712938

IPC: C22C 16/00

Language of the proceedings: EN

Title of invention:
Zirconium alloy

Applicant:
GENERAL ELECTRIC COMPANY

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54

Keyword:
"Novelty (yes)"

Decisions cited:
-

Catchword:
-

Case Number: T 0318/00 - 3.2.2

D E C I S I O N
of the Technical Board of Appeal 3.2.2
of 14 June 2002

Appellant: GENERAL ELECTRIC COMPANY
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 23 September 1999 refusing European patent application No. 95 307 858.1 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: W. D. Weiß
Members: R. Ries
U. Tronser

Summary of Facts and Submissions

I. This appeal, which was filed on 26 November 1999, lies against the decision of the Examining Division dated 23 September 1999, refusing European patent application No. 95 307 858.1 filed on 3 November 1995 in the name of GENERAL ELECTRIC COMPANY and published under No. EP 0 712 938 A1. The appeal fee was paid on the filing date of the Notice of Appeal, and the Statement of Grounds of Appeal was filed on 31 January 2000.

II. The decision under appeal was based on a set of 23 claims as originally filed. Independent claims 1, 12 and 19 read as follows:

"1. A zirconium-based alloy, comprising 0.05-0.09 weight percent of iron, 0.03-0.05 weight percent of chromium, 0.02-0.04 weight percent of nickel, 1.2-1.7 weight percent of tin and 0-0.15 weight percent oxygen, with a balance of zirconium."

"12. A nuclear fuel element, comprising:
a cladding tube having an inner surface region and an outer surface region, said tube comprising a cross-section of a zirconium-based alloy matrix having alloying elements comprising 0.05-0.09 weight percent of iron, 0.03-0.05 weight percent of chromium, 0.02-0.04 weight percent of nickel, 1.2-1.7 weight percent of tin and 0-0.15 weight percent oxygen, and a balance of zirconium, wherein the iron, chromium and nickel alloying elements are in a sufficient concentration to form a plurality of precipitates; and
nuclear fuel material disposed within said tube.

"19. A nuclear fuel element, comprising:

a cladding tube having an inner surface and an outer surface, said cladding tube comprising an outer cladding alloy and an inner cladding alloy that are metallurgically bonded to one another, wherein the outer cladding alloy comprises 0.05-0.09 weight percent of iron, 0.03-0.05 weight percent of chromium, 0.02-0.04 weight percent of nickel, 1.2-1.7 weight percent of tin and 0-0.15 weight percent oxygen, and a balance of zirconium and the inner cladding alloy is a zirconium-based alloy, wherein the iron, chromium and nickel alloying elements in both the outer cladding alloy and inner cladding alloy are in a sufficient concentration to form a plurality of precipitates; and nuclear fuel material disposed within said tube."

III. The Examining Division found in its decision that the zirconium-based alloy according to claim 1 was anticipated by the disclosure given in document

D1: FR-A-2 693 476.

In the Examining Division's view, the claimed Zr-alloy composition was regarded as being a "selection" from the known Zr-alloy composition disclosed in this document, but the claimed sub-range failed to satisfy the criteria for the novelty of a selection invention.

IV. With the Statement of Grounds of Appeal, the appellant requested that the decision under appeal be set aside and a patent be granted on the basis of the claims as originally filed. Oral proceedings were requested, should a negative decision be contemplated by the Board.

In support of novelty, the appellant drew attention to the fact that document D1 neither deals with minimizing the propensity for uniform corrosion of a Zircaloy-2 type alloy under high burn-up conditions nor teaches that this aim could be achieved by diluting the amounts of Fe, Cr and Ni of a Zircaloy-2 alloy. By contrast, document D1 teaches to increase the Ni-content of a Zircaloy-4 alloy in order to improve the alloy's resistance to nodular corrosion.

Reasons for the Decision

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

2. Document D1 relates to a product having at least an exterior surface formed by a Zr-alloy comprising the following additional elements in wt%: 0.40 to 1.70% Sn, 0.05 to 0.25% Fe, 0.03 to 0.16% Cr, 0.0070 to 0.0300% Ni, 0.05 to 1400 ppm oxygen, other residual impurities, the balance being Zr (cf. D1, claim 1, page 2, lines 17, 18). As can be seen, an overlap exists between the elemental ranges of the Zr-alloy composition claimed in the present application and that given in document D1. However, a closer inspection of document D1 reveals that this alloy is characterized as a "nickel doped Zircaloy-4" which comprises nickel as a voluntary low addition rather than as an impurity. According to document D1, the low amounts of nickel cause a surprising improvement in nodular corrosion resistance without adversely affecting the uniform corrosion resistance and hydrogen absorption rate of the alloy so that its corrosion properties come closer to those of Zircalloy-2 (cf. D1, page 2, lines 19 to 32). Although this improvement has been found to exist

over the whole range of 0.4 to 1.7% Sn, the resistance to uniform and nodular corrosion resistance could be further improved if, in a first embodiment, the Sn amounts are decreased to 0.4 to 0.8%, with the alloy then being particularly suitable for use as an exterior layer or as sheathing tubes (cf. D1, page 3, lines 28 to 31). If, however, in a second embodiment, Sn contents in the range of 1.2 to 1.7 are selected (corresponding to the Sn-range claimed in the present application), the zirconium alloy is of a Zircaloy-4 type modified by the addition of Ni, this alloy comprising 0.18% to 0.24% Fe, 0.07 to 0.13% Cr, with the total of Fe+Cr+Ni being 0.28 to 037% (cf. D1, page 4, lines 6 to 11). As can be immediately noted, these amounts of iron and chromium and the total of Fe+Cr+Ni fall completely outside the corresponding ranges of the zirconium alloy claimed in the present application. Hence there is no reason to choose, on the basis of document D1, a composition of a (1.2-1.7%)Sn-Zr alloy which exhibits the extremely low amounts of Fe, Cr and Ni specified in the present application.

The evaluation of the contents of document D1, therefore, leads to the conclusion that the teaching in this document neither makes the claimed "diluted" Zr-alloy composition (ie the area of overlap) available to the skilled reader nor provides the metallurgist with specific information so that he would seriously contemplate applying the technical facts at his disposal in the range of overlap. Consequently, the subject matter of claim 1 is novel over the teaching given in document D1.

3. The decision further argues that the claimed Zr-alloy was an arbitrary choice since it failed to exhibit any

improved properties under standard conditions with respect to the Zr-alloys known from the prior art, and since the expected improvement under high burn-up was not supported by experimental data.

There is, however, no evidence for such a finding. The application stresses the point that an economically driven shift to higher burn-up (ie to higher neutron fluences) and long fuel cycles may push conventional Zircaloy-2 beyond its inherent capability to resist the corrosion attack in the boiling water (BWR) or pressurized water (PWR) reactors and that little is known about the alloy's response at such high burn-up. Based on theoretical considerations about the metallurgical background of the corrosion behaviour of zirconium alloys in BWR or PWR (cf. the A1 publication page 4, lines 44/45), the present application aims at designing a Zr-alloy composition which exhibits an improved uniform corrosion resistance at high burn-up while maintaining the initial uniform and nodular corrosion resistance at normal conditions. It is well known in metallurgy that even slight variations of its constituents may change the properties of an alloy dramatically and unpredictably. Due to the difficulty in obtaining access to the high neutron fluxes necessary to test the alloy's response under severe industrial conditions, the applicant has performed indirect tests on a lab scale and reported the results in detail in the example and the Figures. As shown in Figure 1, the steam tests did not produce widespread nodular corrosion of the dilute alloy on any of the test coupons. The results from the uniform corrosion tests are shown in Figure 2. Based on these test results, the corrosion behaviour of alloys having a dilution factor between 0.3 to 0.5 is comparable to the corrosion properties of undiluted Zircaloy-2. Already this finding is surprising since higher contents of Fe and Cr than claimed were considered indispensable to protect Zircaloy-2 adequately from corrosion attack. Hence, there is no basis for concluding or implying that a pronounced benefit in terms of improvement to the corrosion properties under high burn-up cannot be achieved with the claimed "dilute" Zr-alloy.

4. It may be argued that the upper limits to the elemental ranges of the claimed composition (ie 1.2-1-7% Sn, 0.09%Fe, 0.05% Cr, 0.04% Ni, balance Zr) fall within the standard specification for Zircaloy-2 (1.2-1.7% Sn, 0.07-0.20% Fe, 0.05-0.15%Cr, 0.03-0.04% Ni, balance Zr). For Zircaloy-2, however, technical prudence would dictate to select an alloy composition in the upper range of the ASTM specification. To this end, the iron content is fixed at 0.12% and the chromium content at 0.1% in conventional Zircaloy-2. Moreover, the total of Fe+Ni+Cr should be in the range of 0.25 to 0.45% as can be seen from Figure 10 of document

D2: ASTM Special Technical Publication, 368, 1963, page 3 to 27 (cf. also D2, pages 10, 11: Zircaloy-2).

This general technical knowledge is also confirmed by the exemplifying Zr-alloys disclosed in document

D3: JP-A-02209443

which is mentioned in the European Search Report: for Zr-alloy having a Sn content in the range of 1.2 to 1.7%, the iron content is between 0.16 to 0.48% and Cr contents between 0.08 to 0.27% (cf. D3, Table 1,

examples 4 to 8).
This statement also applies to document

D4: US-A-5296 058

which is concerned with the production of structural parts made from Zircaloy-2 or Zircaloy-4, this alloy further including specific amounts of oxygen and silicon (cf. D4, claim 15; column 3, lines 26/27).

The evaluation of the contents of documents D1, D3 and D4 shows that none of the cited prior art documents discloses the Zr-alloy composition proposed by the present application. Consequently, the subject-matter of claim 1 of the present application is novel.

5. Given that the reason of lack of novelty set out in the decision of the Examining Division for refusing the application no longer applies, the Board cannot support the decision under appeal and it has, therefore, to be set aside. The first instance has not yet examined whether or not the present application meets the remaining requirements of the EPC, in particular those of Article 56 (inventive step). It is, therefore, considered appropriate in accordance with Article 111(1) EPC to remit the case to the first instance for further prosecution.

6. Since the request for oral proceedings was conditional on the Board's intention to decide negatively, which condition is not met, this request has no relevance.

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 for further prosecution.

The Registrar: The Chairman:

V. Commare W. D. Weiß