

A		B		C	X
---	--	---	--	---	---

File Number: T 383/90 - 3.2.2

Application No.: 83 106 001.7

Publication No.: 0 098 996

Title of invention: Zirconium alloy having superior corrosion resistance

Classification: C22F 1/18, C22C 16/00, G21C 3/06

D E C I S I O N
of 22 March 1993

Proprietor of the patent: Hitachi Ltd

Opponent: OI) Sandvik Aktiebolag
OII) Siemens Aktiengesellschaft, Berlin und München

Headword: Zirconium alloy for nuclear fuel elements

EPC Articles 56 and 84

Keyword: "Product-by-process claim (yes)"



Case Number : T 383/90 - 3.2.2

D E C I S I O N
of the Technical Board of Appeal 3.2.2
of 22 March 1993

Appellant :
(Opponent II)

Siemens Aktiengesellschaft,
Berlin und München
Postfach 22 16 34
W - 8000 München 22 (DE)

Other party :
(Opponent I)

Sandvik Aktiebolag
Fack
S - 811 81 Sandviken 1 (SE)

Representative :

Östlund, Alf Olof Anders
Sandvik AB Central Service
Patents and Licences
S - 811 81 Sandviken (SE)

Respondent :
(Proprietor of the patent)

HITACHI, LTD.
6, Kanda Surugadai 4-chome
Chiyoda-ku,
Tokyo 100 (JP)

Representative :

Patentanwälte
Beetz sen. - Beetz jun.
Timpe - Siegfried - Schmitt-Fumian
Steinsdorfstraße 10
W - 8000 München 22 (DE)

Decision under appeal :

Interlocutory decision of the Opposition Division
2.1.06.017 of the European Patent Office dated
31 January 1990, posted on 21 March 1990,
concerning maintenance of European patent
No. 0 098 996 in amended form.

Composition of the Board :

Chairman : G.S.A. Szabo
Members : W.D. Weiß
M.K.S. Aúz Castro

Summary of Facts and Submissions

I. European patent No. 0 098 996 was granted with effect of 30 December 1986 on the basis of European patent application 83 106 001.7, filed on 20 June 1983.

II. Two oppositions were filed against the patent on the grounds of lack of novelty and inventive step (Article 100a EPC) in the light of the following documents:

(1a) ASTM-B 353-60T;

(1b) ASTM-B 353-83;

(2) US-A-3 865 635;

(3) Journal of Nuclear Materials 105 (1982), pages 132, 140, 141;

(4a) Trans. ASM (1960), Volume 52, page 1135;

(4c) Conference paper ASTM STP 754, 1982, page 88, presented at the ASTM 5th Int. Conference on Zirconium in the Nuclear Industry, Boston August 4 to 7, 1980;

(5) US-A-2 894 866;

(6) US-A-4 238 251.

III. During oral proceedings, held on 31 January 1990, an amended version of the patent in suit was submitted on the basis of which the Opposition Division orally gave the decision to maintain the patent as amended. The written grounds for this decision were posted on 21 March 1990.

IV. On 8 May 1990, the Appellant (Opponent II) filed an appeal with statement of grounds against this decision and paid the appeal fee on the same date. Document

(9) B. Lustman, "The Metallurgy of Zirconium", McGraw-Hill 1955, pages 631 to 634, and 742,

was cited as a new state of the art.

An auxiliary request for an oral proceedings was withdrawn by telecopy of 25 February 1993.

V. In a communication dated 17 September 1992 the Board drew the attention to the fact that the amended Claim 1 on which the decision under appeal was based was objectionable with respect to clarity because its formulation also included embodiments by which the basic technical problem was not solved. Moreover, it expressed as its provisional opinion that the prior art cited by the Appellant could not put into question that the subject-matter of Claim 1 involved an inventive step.

VI. On 5 February 1993, the Respondent filed a new set of three claims together with amended parts of the description. The independent Claim 1 in this version reads as follows:

"1. A zirconium alloy having superior corrosion resistance, which alloy consists, by weight, of 1-2% Sn, at least one kind of Fe and Cr selected from the group consisting of 0.05-0.3% Fe and 0.05-0.2% Cr; 0-0.1% Ni, and the balance Zr and inevitable impurities, the total amount of said at least one kind and Ni all existing in the solid-solution of the zirconium alloy being not less than 0.26%, the zirconium alloy being subjected to a solution heat treatment at a temperature at which both the α phase and β phase thereof are included in the zirconium alloy, plastic working, consisting of cold plastic working or both hot and cold plastic working, and annealing at a temperature of 400 - 700°C."

The dependent Claims 2 and 3 refer to particular embodiments of the zirconium alloy according to Claim 1.

VII. The Appellant's arguments can be summarised as follows:

According to document (5), a standardised Zircaloy 2 alloy pursuant to the specifications given in document (9) was treated. This specification showed that also the sum of the contents of nickel, iron and chromium met the requirements of the patent in suit. It was only an obvious measure to treat an alloy of this known composition by a method disclosed in document (5), column 5, lines 37 to 45. This particular known method, which included a solution annealing in the α - plus β -region, had obviously been overlooked by the Opposition Division and, therefore, not been considered in its decision.

Moreover, the category as a product-by-process claim was wrongly chosen and did not follow the Guidelines C, III, 4.7a and 4.7b. The clarity objection had to be considered, because the claims had been amended.

VIII. The Respondent presented essentially the following arguments:

The objection with respect to the category of Claim 1 had to be disregarded, because it did not fall under the reasons of Article 100a EPC.

According to the patent in suit, the method to which the basic alloy was subjected resulted in a microstructure by which it had not only a superior corrosion resistance but also an enhanced value of the elongation allowing for the swelling of the uranium fuel occurring during its nuclear burning.

The "previously accepted procedure" disclosed in column 5, lines 37 to 45, of document (5) inevitably led to the

formation of Fe, Cr and Ni precipitates in the Zr alloy, so that the amount of solid-solutioned Fe, Cr and Ni could not arrive at the required level of 0.26%. Furthermore, neither cold plastic working after hot forging nor annealing after cold plastic working was disclosed there. On the other hand, the method recommended according to document (5) avoided the two-phase region of the alloy during the solution treatment, used by the "previously accepted procedure" and aimed at a high degree of isotropy. Consequently, the teaching of document (5) created a prejudice against the subject-matter rather than giving an incentive to go into its direction.

IX. The Appellant requests that the decision under appeal be set aside and the patent be revoked in its entirety. It also requests the refund of the appeal fee in view of an alleged substantial procedural violation.

The Respondent requests that the patent be maintained on the basis of the following documents:

- Claims 1 to 3 filed on 5 February 1993;

- Description pages 2, 5 and 6 filed on 5 February 1993
 (with the insertion in column 2, after
 line 37, filed on 2 January 1990);
 pages 3 and 4 of the patent specification;

- Drawings Sheets 1 to 4 of the patent specification.

Reasons for the Decision

1. The appeal is admissible.

2. Amendments

The amended Claim 1 differs from the Claim 1 as granted only in that the restricting feature "at a temperature of 400 - 700°C" has been added at its end and in that the specific term "consisting of cold plastic working or both hot and cold plastic working" is inserted after "plastic working". The change of the upper limit for the chromium content from 2% into 0.2% is a correction of an obvious printing mistake only and not a change of the granted version of the claim.

Moreover, Claim 1 originates from Claims 1, 2 and 4, and from Figures 5 and 6 as filed.

The dependent claims are also founded on the original disclosure.

There is, therefore, no objection to the current claims under Article 123 EPC.

3. Clarity of the Claims

Clarity of the claims does not belong to the reasons on which an opposition may be founded pursuant to Article 100 EPC. Whenever amendments have been made by the proprietor, Article 102(3) EPC confers upon the Opposition Division as well as the Boards of Appeal jurisdiction to consider all conditions for patentability. In the decision in case T 301/87 (OJ EPO 1990, 335) it was held (point 3.8 of the Reasons) that when amendments are made to a patent during an opposition proceedings, Article 102(3) EPC requires consideration by either instance as to whether the

amendments introduce any contravention of any requirement of the Convention, including Article 84 EPC. However, the Board also held that Article 102(3) EPC did not allow fresh objections to be based upon Article 84 EPC if such objections did not arise out of the amendments to be made.

In the present case, it is objected that Claim 1 is unclear because this product claim contains process features which may not render a known alloy composition novel, and that its category is, therefore, wrongly chosen.

Claim 1 as granted was already formulated as a product claim and comprised a mixture of composition and process features. The amendments made to this claim after grant only reduce the scope of the process features which are contained in the granted version but are not such that they introduce themselves ambiguity.

The Board, therefore, sees no reason why the clarity of Claim 1 should be challenged, when its subject-matter proves to be novel and inventive in any case. This is not one of those cases either, where ambiguity would be a problem even without amendments because some other legitimate ground of objection cannot be resolved in view of the uncertainty of the scope of the claim.

4. Novelty

4.1 The (process) features contained in Claim 1 result in particular modifications in the microstructure of the alloy which can be unambiguously identified and have to be taken into consideration, when its subject-matter is compared with the prior art cited.

The requirement that "the total amount of iron, chromium and nickel all existing in the solid solution of the

zirconium alloy be not less than 0.26%", brings about that the alloy has to be cooled with a considerable speed from the temperature at which the solution treatment is performed. This high cooling velocity additionally involves that the microstructure of the alloy comprises a mixture of mild granula α phase and needle-like α' phase being present at room temperature whereby the subsequent cold working is facilitated (EP-B-0 098 996, column 3, lines 4 to 21). The final plastic working comprising cold working and final annealing results in a recrystallised pure α structure (column 3, lines 22 to 27) which, according to the general knowledge of the metallurgical engineer, is anisotropic and results in an elongation which is higher in the direction transverse to the forming direction than parallel thereto (Respondent's letter dated 15 October 1990, the paragraph bridging pages 2 and 3).

- 4.2 Document (5), on which the Respondent has put the highest emphasis, discloses two separate treatments of a zirconium alloy of the generally known standard type Zircaloy 2 admitting alloy compositions in which the sum of the chromium, nickel and iron contents may lie below or above 0.26% by weight. These two treatments which are rated as "previously accepted" (column 5, lines to 57) and as "achieving the object of the invention" (claims), respectively, have to be considered as separate states of the art when assessing novelty and inventive step.

The "previously accepted" method disclosed in document (5) comprises a final anneal at 843°C which provokes a precipitation of iron, nickel and chromium and prevents that an amount of at least 0.26% of these elements remains in solid solution, even if the composition should have been chosen such that the sum of their contents exceeds this minimum value. The recommended other method of document (5) as well as the method disclosed by document

(2) exclude the α plus β phase region for solution annealing.

Document (6) discloses to precipitate intermetallic phases after a solution annealing in the α plus β phase region.

The other documents enumerated above are cited as a background art only and also fail to disclose an alloy exhibiting the combination of features indicated in Claim 1.

Consequently, the subject-matter of Claim 1 is novel.

5. Closest prior art and difference

None of the cited documents explicitly states that the total amount of iron, chromium and nickel all existing in the solid-solution should be more than 0.26%. However, the documents (5), (2), and (6) disclose zirconium alloys of generally known standard types admitting alloy compositions in which the sum of the chromium, nickel and iron contents may lie below or above 0.26% by weight and which, therefore, embrace basic compositions which enable this requirement to be met provided that the basic composition is selected accordingly and afterwards treated in a manner to keep these elements in solid-solution. Only document (2) of these three sources describes a method for the treatment of these zirconium alloys which is apt to keep the chromium, nickel and iron contents in solid-solution.

The Board, therefore, considers document (2) to be the closest prior art.

This document discloses zirconium alloys which consist, by weight, of 1.2-1.7% Sn, 0.07-0.24% Fe, 0.05 - 0.15% Cr, 0 - 0.08% Ni, balance zirconium and customary impurities,

of which the oxygen is adjusted to 0.09 - 0.16%. These compositions embrace standard compositions which are known as "Zircaloy 2" and "Zircaloy 4". These known alloys are subjected to a treatment which comprises a solution heat treatment at a temperature at which exclusively the β phase is included in the zirconium alloy, plastic working, consisting of hot and cold plastic working, and annealing at a temperature of 400 - 700°C (see the claims). The heat treatment to which this known alloy is subjected is such as to increase the content of alloying elements in solution (column 2, lines 37 to 44).

6. Technical Problem and Solution

The zirconium alloys according to document (2), as well as the alloy according to the patent in suit, are intended to be used as a structural material in a nuclear reactor which material is to be used in contact with water of a high temperature under high pressure. A typical application which falls under this specification are canning or cladding tubes or other constructional parts of a nuclear fuel assembly.

These materials are required to possess both strength and ductility together with a high corrosion resistance. Even when they are used in contact with the water or steam at a high temperature and under a high pressure for a long time no nodular corrosion should occur and the tendency to form a thick and peeling off oxide layer should be small (document (2), column 2, lines 12 to 19; EP-B-0 098 996, lines 40 to 47). When used as a tube for the fuel element, the material must have a particularly high elongation value in the transverse direction of the tube to allow without burst for the swelling of the nuclear fuel occurring during the burning time of the reactor.

The basic composition ranges of the alloy disclosed in document (2) allow to chose such Fe/Cr/Ni contents which lie above 0.26% as well as such which lie below this value. The comparative tests reported by the Respondent in the Exhibit No. 2 to his letter of 12 July 1988 prove that the treatment disclosed in document (2) is also apt to maintain at least 0.26% of the said alloying elements in solution once the basic composition has - by chance - been chosen adequately. In this case, all but one of the requirements for a nuclear fuel tube specified above are also met by the material disclosed in document (2). Only the rupture elongation is reduced even when compared to still older prior art materials which would result in a shorter life time when this material is used to house fuel elements.

Starting from the closest prior art described above, the basic technical problem of the patent in suit is, therefore, to create a structural material for nuclear fuel assemblies which combines a reliably predictable high corrosion resistance with an increased rupture elongation of the material, in particular in transverse direction, leaving the rest of the material characteristics of the known material unaltered.

This problem is solved by the distinguishing features of the subject-matter of Claim 1 which differs from the closest prior art as represented by document (2) in that the total amount of the iron, chromium and nickel contents all existing in the solid-solution of the zirconium alloy is not less than 0.26% and in that the solution heat treatment is carried through at a temperature at which both the α phase and the β phase are included in the zirconium alloy. In this context reference is also made to the Respondent's letter dated 12 July 1988, paragraph II, and Exhibit No. 2, Table 1).

7. Inventive Step

Although the author of document (2) has already recognised (column 2, lines 12 to 19) that a material for the manufacture of nuclear fuel canning tubes requires high values of both strength and ductility (measured by rupture elongation), this document invests its major efforts in increasing the creep strength only (column 2, lines 20 to 43) and accepts that the ductility of the strengthened material is even lower than that of hitherto conventional materials (letter of 12 July 1988 by the Respondent, Exhibit 2, Figure 1). It is readily welcomed that the maintenance of the alloying elements in solid-solution results in an increased creep strength (column 2, lines 37 to 43). Document (2), however, does not contain any hint that these alloying elements increase the resistance against (nodular) corrosion when they are dissolved in the alloy lattice in a total amount of at least 0.26%. The author is rather content that the corrosion properties are not worse than in previously known materials (column 3, lines 16 to 23). Moreover, the solution treatment before the last cold working step is taught to be carried out strictly through at a temperature at which exclusively the β phase exists.

Document (5), onto which the highest emphasis was put by the Appellant during the appeal proceedings, discloses two separate methods to produce a structural material for the fuel system of a nuclear reactor (see the first two paragraphs under 4.2. above).

The historically older "previously accepted" method (column 5, lines 35 to 43), owing to being continuously worked during cooling from the α plus β range and due to a final (recrystallisation) anneal at a temperature in the upper extent of the α range (843°C), provides a microstructure which is characterised by precipitated

intermetallic compounds and so called stringers at the grain boundaries (column 1, line 69, to column 2, line 32) which has an embrittling influence on the material.

The method which is recommended to overcome the disadvantages of the "previously accepted" method strongly teaches to avoid the α plus β range during the solution treatment and to anneal exclusively in the β range; moreover, the final anneal is executed in the upper extent of the α range (Claim 1). These measures result in a fine grained multiphase structure with essentially isotropic mechanical properties. Consequently, the microstructures of the materials ensuing from the "previously accepted" as well as from the recommended method are quite distinct of the structure of the zirconium alloy according to the patent in suit (point 4.1. above).

Document (6) aims at solving the same problem as the patent in suit, placing the avoidance of nodular (pustular) corrosion in the foreground of its intentions (column 2, line 5, to column 3, line 47). The resulting material which is obtained accordingly represents quite another way as a solution, because the alloying elements are precipitated in form of particularly arranged arrays (column 3, lines 49 to 67) instead of being kept in solid-solution to a high extent.

The documents (1 a and b), (3), (4 a and c) and (9) as well as further documents submitted by the Respondent during the opposition proceedings give only background information and are further away from the subject-matter of Claim 1 than are the documents discussed in detail above.

The Board, therefore, comes to the conclusion that the subject-matter of Claim 1 cannot be derived in an obvious

manner from the documents cited by the Respondents and must accordingly be seen as involving an inventive step in the meaning of Article 52(1) in combination with Article 56 EPC.

8. The independent Claim 1, together with the Claims 2 and 3 appended and the revised description adapted thereto, can, therefore, form the basis for maintaining the patent as amended.

9. Refund of the appeal fee

The Opposition Division has properly discussed the "previously accepted" method of document (5) separately from the recommended method of the same document, see points 5. and 7. of the decision under appeal. The Board cannot recognise any reason why this correct proceedings should be rated as a substantial procedural violation.

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 maintain the patent on the basis of the following documents:

Claims 1 to 3 filed on 5 February 1993;

Description pages 2, 5 and 6 filed on 5 February 1993
(with the insertion in column 2, after

line 37, filed on 2 January 1990);
pages 3 and 4 of the patent specification;
Drawings Sheets 1 to 4 of the patent specification.

3. The request to refund the appeal fee is rejected.

The Registrar



S. Fabiani

The Chairman


G. Szabo

01245
J.D.W. 7. 4. 93
11/1/93