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DECISION of 1 February 2005

| Case Number: | T 1021/00 - 3.4.1 |
|---------------------|-------------------|
| Application Number: | 91119732.5 |
| Publication Number: | 0488027 |
| IPC: | G21C 3/07 |
| | |

Language of the proceedings: EN

Title of invention:

Zirconium based alloy member of low irradiation growth, method of manufacturing the same, fuel channel box and assembly, and use of the same

Patentee:

Hitachi, Ltd.

Opponent:

Siemens AG

Headword:

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Relevant legal provisions: EPC Art. 54, 56, 84, 123(2)

Keyword:

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Decisions cited:

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Catchword:

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Boards of Appeal

Chambres de recours

Case Number: T 1021/00 - 3.4.1

D E C I S I O N of the Technical Board of Appeal 3.4.1 of 1 February 2005

| Appellant: (Proprietor of the patent) | Hitachi, Ltd. 6, Kanda Surugadai 4-chome Chiyoda-ku, Tokyo 101 (JP) | |
|----------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------|--|
| Representative: | Beetz & Partner Patentanwälte Steinsdorfstrasse 10 D-80538 München (DE) | |
| Respondent: (Opponent) | Siemens AG Postfach 22 16 34 D-80506 München (DE) | |
| Representative: | Tergau & Pohl Patentanwälte Mögeldorfer Hauptstrasse 51 D-90482 Nürnberg (DE) | |
| Decision under appeal: | Decision of the Opposition Division of the European Patent Office posted 4 August 2000 revoking European patent No. 0488027 pursuant to Article 102(1) EPC. | |

Composition of the Board:

| Chairman: | G. | Assi | |
|-----------|----|--------------|--|
| Members: | R. | Q. Bekkering | |
| | Α. | Pignatelli | |

Summary of Facts and Submissions

- I. The appellant (patentee) lodged an appeal against the decision of the opposition division, dispatched on 4 August 2000, revoking the European patent No. 0 488 027. The notice of appeal was received on 2 October 2000, the appeal fee being paid on the same day, and the statement setting out the grounds of appeal was received on 14 December 2000.
- II. Opposition had been filed against the patent as a whole, based on Articles 100(a), (b) and (c) EPC. The opposition division found in the decision under appeal that the subject-matter of amended claim 1 according to the main request as well as according to any of the three auxiliary requests then on file did not involve an inventive step with respect to the cited prior art and revoked the patent accordingly.
- III. Oral proceedings, requested by both the appellant and the respondent (opponent), were held on 1 February 2005.
- IV. Reference was in particular made to the following documents:
 - B: V. Fidleris, "The irradiation creep and growth phenomena", Journal of Nuclear Materials, vol. 159 (1988), pages 22 to 42

K: JP-A-59 229 475 & English translation.

The day before the oral proceedings the respondent submitted the following further document:

M: EP-B-0 385 719

V. In the oral proceedings, the appellant requested that the decision under appeal be set aside and the patent maintained in amended formed based on the following documents:

> Single claim 1 as filed in the oral proceedings on 1 February 2005; Description and drawings to be adapted.

The respondent requested that the appeal be dismissed.

VI. Claim 1 according the appellant's request reads as follows:

"A method of manufacturing a fuel channel box using a zirconium-based alloy plate formed through a cold rolling process and an annealing process, and containing 5 wt-% or less Sn and/or 5 wt-% or less Nb, and not less than 90 wt-% Zr, comprising the steps of:

- forming two generally U shaped-members of the zirconium-based alloy plate and welding the members for fabricating a square tubular member,
- heating the tubular member at a temperature within the β single phase temperature range,
- retaining the tubular member at the temperature,
- hardening the tubular member by forced quenching using a cooling medium, and

- annealing the tubular member thereafter, wherein the retention time and the temperature of the heat treatment in the β single phase region are controlled by growing the crystal grains until the grain size in the β single phase is at least 100 μ m, so that in the final fuel channel box

- the <0001> orientation of the hexagonal crystals of zirconium with respect to the direction perpendicular to the surface of the tubular member produces an Fr value determined by X-ray diffraction measurement ranging from 0.333 - 0.35,
- the <0001> orientation of the hexagonal crystals of zirconium with respect to the longitudinal direction of the tubular member produces an Fl value determined by X-ray diffraction measurement ranging from 0.25 to 0.36, and
- the <0001> orientation of the hexagonal crystals of zirconium with respect to the circumferential direction of the tubular member produces an Ft value determined by X-ray diffraction measurement ranging from 0.25 to 0.36, and

wherein the annealing of the tubular member is performed in a manner that

- a mandrel of a material having a thermal expansion coefficient larger than that of the Zr-based alloy is inserted into the tubular member and
- the ends of the tubular member and the mandrel are fixed to each other,
- said mandrel being formed such that the mandrel and the tubular member partly contact each other preventing the inner surface of the tubular member from a whole-surface-contact with the mandrel, and then
- the tubular member is heated and retained at the annealing temperature".

VII. The appellant submitted in particular the following arguments:

None of the cited prior art documents disclosed or even suggested a method of manufacturing a fuel channel box with no irradiation growth when subjected to neutron irradiation in a nuclear reactor as specified in claim 1. Document K addressed the problem of eliminating bending of zirconium alloy fuel channel boxes due to irradiation growth, but failed to provide manufacturing details such as the heat treatment in the β -phase controlled by the grain size growth and the use of a mandrel during anneal as specified in the claim. Document B, providing an overview of the knowledge concerning in particular irradiation growth of zirconium alloys, merely mentioned β -guenching as a possible means for reducing irradiation growth, without, however, giving any details as to retention time or temperature of such a treatment. Document M should not be admitted into the proceedings because it was filed too late and was not highly relevant. The document disclosed the thermal sizing of a fuel channel box by means of a mandrel inserted in the box. However, the sizing mandrel required a whole-surface contact with the box and was therefore not comparable with the mandrel as claimed.

VIII. The respondent's arguments may be summarised as follows:

Claim 1 did not define a proper control method since no feed-back was provided from the grain size measurement to the retention time and temperature, and therefore lacked clarity (Article 84 EPC). As such, in the claim merely the result to be achieved by the heat treatment in terms of grain size was defined. Moreover, it was technically impossible to measure the grain size in the β -phase in view of the high temperatures and the short retention times involved. The grain size could only be measured after the heat treatment, ie in the α -phase, typically by microscopy analysis of cross sections of the material. Accordingly, a control of the heat treatment based on a measurement of the grain size in the β -phase was not derivable from the application as originally filed (Article 123(2) EPC).

The subject-matter of claim 1 was rendered obvious by the teaching of document K in combination with the teachings of document B, as far as the thermal treatment was concerned, and document M, as far as the forming of the tubular member and the use of the mandrel during annealing was concerned. In particular, document K suggested eliminating irradiation growth in a fuel channel box by randomizing the crystal orientation of the zirconium alloy. Document B disclosed that this could be achieved by a β -quenching treatment and also indicated that large crystal grains in the zirconium alloy resulted in low irradiation growth. Document M was highly relevant and disclosed, in particular, thermal sizing of fuel channel boxes using a mandrel inserted in the box. The mere prevention of a whole-surface contact between box and mandrel as claimed, could not provide a sufficient distinction over this prior art.

Reasons for the Decision

 The appeal complies with the requirements of Articles 106 to 108 and Rule 64 EPC and is therefore admissible.

2. Procedural issues

Document M was submitted late, as it was filed by the respondent only the day before the oral proceedings. Nonetheless, the appellant confirmed on request that it had had sufficient time to study the document. The board may disregard evidence which is submitted late in accordance with Article 114(2) EPC, if it considers that this is necessary for reasons of procedural economy or to guarantee fair proceedings. Since, in the present case, the late submission did not cause any delay in the proceedings and the appellant had the opportunity to present its comments on it, the board had no reason to disregard the document.

3. Amended claim 1

3.1 Claim 1 according to the appellant's sole request is based on claim 6 of the application as originally filed and parts of the original description. In particular, the formation of the U-shaped members and the welding thereof is disclosed on page 6, lines 38 to 40 of the application as published. The control of the retention time and temperature of the heat treatment in the β phase and the resulting values of the fractions Fr, Fl and Ft determined by X-ray diffraction are derivable from page 5, lines 22 to 23, lines 32 to 38 and lines 50 to 53 of the published application. Finally, the annealing step and the use of the mandrel during the annealing are derivable from page 6, lines 20 to 32 of the published application.

Therefore, the amendments originate from the application documents as filed, so that the requirements of Article 123(2) EPC are considered to be met.

Moreover, the amendments provide additional limiting features having regard to independent method claim 4 of the patent as granted, so that the requirements of Article 123(3) EPC are considered to be met as well.

3.2 From the wording of claim 1 it is clear that the grain size controls the retention time and temperature of the heat treatment in the β -phase to the extent that the heat treatment at the given temperature in the β -phase, and the growth of the crystal grains connected therewith, is continued up to the point in time at which the grain size as measured is at least 100 μ m, after which quenching is performed.

The fact of growing the grains **until** a certain grain size is reached implies the termination of the heat treatment at this point in time. This provides a control of the retention time and of the functionally related temperature, or, in respondent's words, a "feed-back" of the control method. The respondent's objections as to clarity in this respect are, therefore, not convincing.

As regards the grain size in the β -phase, according to the appellant this measurement was known to the skilled

person and could be performed without difficulty by, for example, high temperature X-ray diffraction. It is noted that although the technical feasibility of the grain size measurement in the β -phase has been an issue in the opposition proceedings from the onset, the respondent has not provided any convincing arguments supported by evidence. The respondent's doubts expressed in this respect are, therefore, considered to be unfounded.

Therefore, the requirements of Article 84 EPC are met.

4. Novelty

The closest prior art may be considered to be provided by document K, concerned with the same technical problem underlying the patent in suit of providing a fuel channel box of zirconium alloy (Zircaloy) showing no bending deformation when exposed to neutron irradiation in a nuclear reactor core (see translation, page 4, line 10 to page 5, line 33). Like the patent, document K identifies irradiation growth of the zirconium alloy due to the neutron flux as being the cause of the bending. For fuel channel boxes produced from cold worked plates which are subsequently subjected to recrystallization annealing, the irradiation growth is proportional to $1\mathchar`-3f_{\rm i}$ for any direction i within the plate, wherein f_i is the resolved fraction of the basal poles <0001> of the hexagonal Zr crystal in the direction i. Three orthogonal directions are considered, eg the lengthwise direction of box (direction of rolling) 1, the transverse direction t and the thickness direction r. In particular, according to document K, if the value of the resolved fraction in

the lengthwise direction of the box f_1 is approximately 0.333, no irradiation growth is produced in this direction and the box will not bend.

No further details are however provided in document K as regards the manufacturing process of the fuel channel box.

The subject-matter of claim 1 is, thus, novel with respect to document K (Article 54(1) and (2) EPC). Novelty is also given with respect to the remaining, more remote cited prior art.

5. Inventive step

In view of the above, the problem to be solved underlying the patent in suit may be seen as devising suitable manufacturing steps.

From document B, providing an overview of the irradiation creep and growth phenomena of zirconium alloys used in nuclear reactors, it is known that most β -quenched Zircaloys possess an approximately random distribution of basal poles, ie $f_d^c \approx 0.33$, and their growth (proportional to $1-3f_d^c$) is very low or zero (see page 33, left-hand column, lines 32 to 35), wherein f_d^c is the resolved fraction of basal poles f^c in the direction d. As such, the steps of heating the box at a temperature within the β -phase and of hardening the box by forced quenching are, thus, suggested in document B for obtaining eg the f_1 value of approximately 0.33 sought in document K. It is, however, noted in document B (see page 33, right-hand column, lines 1 to 7 and

figure 19) that β -quenched samples deviate markedly from the 1-3f correlation at higher fluences.

Furthermore, from document M it is known to construct a fuel channel box from two U-shaped zirconium alloy plates welded together and to anneal the box thereafter. The annealing is performed by inserting a mandrel in the box, having a thermal expansion coefficient larger than that of the box, and heating the mandrel with the box using, for instance, an inert gas. The difference in thermal expansion coefficients between the mandrel and the box ensures that the mandrel, which before heating will loosely fit within the box, thereby facilitating its insertion, upon heating will expand more than the box, thereby shaping and dimensioning the box as required.

However, none of the above prior art suggests relying on the grain size of the crystal grains in the β -phase of the zirconium alloy for controlling the retention time and temperature of the heat treatment in the β -phase.

Although, as pointed out by the respondent, from document B (see figure 22 and corresponding description) it is known that recrystallized zirconium material with 225 μ m grains shows very low irradiation growth compared to material with smaller grains, there is no indication that the specified grain size is obtained by recrystallization in the β -phase. In the absence of any suggestion in document B, or in any of the other cited prior art documents, as to the control of the heat treatment in the β -phase using the grain size as specified in claim 1 in suit, an inventive step has to be recognised.

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Hence, the subject-matter of claim 1 according to the appellant's request involves an inventive step (Article 56 EPC).

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 with the single claim filed during the oral proceedings on 1 February 2005 and a description and drawings to be adapted.

The Registrar:

The Chairman:

R. Schumacher

G. Assi