Microstructure Characterization of Zr-2.5Nb Alloy Pressure Tube Manufactured by Novel Fabrication Routes and Irradiated Pressure Tubes by Transmission Electron Microscopy

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Abstract

The Indian Pressurized Heavy Water Reactors (PHWRs) use Zr-2.5% Nb pressure tubes (PT) and the in-reactor performance of these tubes, such as irradiation creep and growth behaviour, depends strongly on their microstructure and texture. The microstructure and texture of the tube essentially depends on the combination of different thermomechanical treatments imparted during manufacturing of these pressure tubes. In the present paper, the microstructure evolution has been studied for pressure tubes, produced by novel heat treated condition. The finished microstructure consisted of fine α lamalle with distribution of nanometer size β phase, along the α/α interfaces having composition 75-80wt%Nb. Microstructure evolution in new optimized fabrication routes for manufacture of cold worked pressure tubes has been described here. The microstructure comprises of long α grain with higher aspect ratio and β being present as thin film having composition 15-20wt%Nb. This new fabrication route is proposed to be used, to manufacture pressure tubes for all upcoming PHWR reactors. The paper also shows irradiated microstructure characterization at different locations of the first pressure tube, obtained from the KAPS2 reactor. The study shows substantial modification in β morphology.

Introduction

The present generation of Indian PHWRs use Zr-2.5% Nb pressure tubes. KAPS-2 is the lead reactor in Indian PHWR reactors, using pilgered Zr-2.5% Nb PTs fabricated by the Nuclear Fuel Complex, Hyderabad. The satisfactory performance and the life of the pressure tubes depend mainly upon the extent of degradation of the mechanical properties, corrosion under the influence of high temperature coolant, neutron flux and dimensional instability as a result of irradiation creep and growth in the reactor condition [1-3]. Irradiation growth and irradiation creep govern the performance of the cladding and the pressure tube material to a great extent. Since dimensional changes and mechanical property degradation can limit the life of PT, it is essential to study and develop an understanding of the microstructural evolution at different levels of manufacturing and its exposure to neutron irradiation. Creep and growth are greatly influenced by the microstructural parameters such as shape, size and aspect ratio of the α-lamellae, the distribution and morphology of the β-phase, the interfacial structure, dislocation density and the crystallographic texture [1-3]. These aspects are sensitive to the manufacturing process of the pressure tube.
The present paper describes the microstructure evolution in a new fabrication route, for cold worked pressure tubes. This route has been developed after an industrial scale trial of nine different routes at NFC. Heat treated pressure tubes are known to show improved creep performance [4]. The present paper describes the microstructure assessment at different stages of fabrication of heat treated pressure tube. Samples taken from an irradiated PT in KAPS2 are evaluated and compared with the microstructure of the unirradiated, as-fabricated material.

Results and Discussion

**Microstructural Study of Zr-2.5Nb alloy Heat Treated Pressure Tube Fabrication**

TEM micrographs after first extrusion showed dynamic recrystallization of the \( \alpha \) grain. Refined and uniform microstructure suggested that, cast structure was completely removed during hot extrusion process, \( \beta \) phase was present at the \( \alpha/\alpha \) interface Fig. 1(a) [5-6]. The billet was \( \beta \) quenched to obtain homogenous microstructure, texture and chemical homogeneity. \( \beta \) quenched microstructure showed martensitic microstructure consisting of coarse lath martensites (Fig. 1(b)). The quenched billet was extruded to hollow blank. Microstructure after second hot extrusion showed that \( \alpha \) grains become elongated in the axial direction of the tube and thin in the radial direction (Fig. 1(c)). The longitudinal section exhibited \( \alpha \) phase of lamellar morphology aligned along the direction of the extrusion. Transverse section exhibited \( \alpha \) lamellae, whose width and thickness were not similar. The average length, width and thickness of \( \alpha \) lamellae were in the range of 15-20\( \mu \)m, 1-2.5\( \mu \)m and 0.3-0.5\( \mu \)m respectively. \( \beta \) phase was present as thin wafer between two \( \alpha \) lamellae, having average thickness of 0.02-0.05 \( \mu \)m. The extruded tube was further pilgered, which was showing heavily deformed microstructure. Subsequent to pilgering, \( \alpha+\beta \) quenching was carried out, which is the most critical step in the fabrication of heat treated pressure tube. Quenching treatments were carried out in a controlled fashion, using quenching dilatometer, to study the evolution of microstructures as a function of different cooling rates. Systematic studies on transformation from \( \alpha+\beta \) phase field as a function of cooling rate are far from being extensive in published literature. In addition to cooling rate, effect of soaking temperature in the \( \alpha+\beta \) phase field has been also examined in the present study. The samples cooled from 883\(^\circ\)C at different cooling rates in argon, showed primary \( \alpha \) and transformed \( \beta \) phases. Samples cooled at the rate of 100\(^\circ\)C/sec and 50\(^\circ\)C/sec showed predominantly internally twinned martensite and internally slipped martensite structure respectively. Below this cooling rate, transformed \( \beta \) product was observed to be Widmanstatten \( \alpha+\beta \). On the basis of these observations it could be concluded, that in order to obtain primary \( \alpha \) and martensitic microstructure after \( \alpha+\beta \) quenching, a minimum cooling rate is required which is a function of soaking temperature. On the basis of the results obtained during controlled heat treatment performed in quenching dilatometer, 883\(^\circ\)C was selected as soaking temperature and water as the quenching medium for \( \alpha+\beta \) quenching operation for large dimension tube (100 mm OD X 4.5 mm WT X 500 mm length). TEM micrographs revealed \( \alpha+\beta \) quenched microstructure consisting of fine martensite phase along with 20-25% primary \( \alpha \) volume fraction (Fig. 1(d)). The \( \alpha+\beta \) quenched samples were cold deformed (by pilgering) to the extent of 23%. Cold deformation of martensitic microstructure resulted in considerable increase in the dislocation density and dislocation substructure. (Fig. 1(e)). These defects acted as nucleation sites for precipitation of \( \beta_{\alpha} \) from the supersaturated martensitic phase during subsequent aging process. Ageing temperatures 540\(^\circ\)C below the recrystallization temperature were used in the present study, after the 2\(^{nd} \) pilgering operation. Complete tempering of martensite at 540\(^\circ\)C was observed whereas no recrystallization of the microstructure was observed. \( \beta \) precipitates (size less than 15nm) were observed only at the \( \alpha/\alpha \) lath interface (Fig. 1(f-g)). Energy Dispersive Spectroscopy (EDS) analysis in TEM of samples, aged at 540\(^\circ\)C, showed 75-80% Nb in \( \beta \) phase. This study thus shows that a temperature of 540\(^\circ\)C is required to attain equilibrium concentration of Nb (85%) in \( \beta \) precipitates during aging. Finally autoclaving treatment was carried out at 290\(^\circ\)C for 120 hrs, which did not modify the microstructure to any noticeable extent (Fig. 1(h)).
The conventional method (CANDU) for fabrication of cold worked Zr-2.5Nb alloy pressure tubes, involves hot extrusion followed by 25 pct cold drawing. Zr-2.5% Nb alloy pressure tubes in NFC fabrication, involves hot extrusion (with lower extrusion ratio) and two-stage pilgering with an intermediate annealing treatment followed by autoclaving. The microstructure of the pressure tube fabricated at NFC has considerably different microstructure than those reported for CANDU pressure tube. The α grains are much finer and aspect ratio is also small [7]. The β phase is mostly present in the globalised form unlike stringer in CANDU tube and it is substantially richer in Niobium. In order to examine the role of individual fabrication stages, in NFC, a campaign was carried out to fabricate pressure tube with different fabrication parameters. In all, pressure tubes were fabricated by nine different fabrication routes. The objective was to examine which route gives the best desirable microstructural, textural and mechanical properties. The important variations in Zr-2.5Nb pressure tube fabricated through different routes are given below:

![TEM Microstructure evolution at different stages of new fabrication route of cold worked Zr-2.5Nb alloy pressure tubes](image)

**Microstructural Study of Zr-2.5Nb alloy Cold Worked Pressure Tube Fabrication by Optimized Fabrication Routes**

The conventional method (CANDU) for fabrication of cold worked Zr-2.5Nb alloy pressure tubes, involves hot extrusion followed by 25 pct cold drawing. Zr-2.5% Nb alloy pressure tubes in NFC fabrication, involves hot extrusion (with lower extrusion ratio) and two-stage pilgering with an intermediate annealing treatment followed by autoclaving. The microstructure of the pressure tube fabricated at NFC has considerably different microstructure than those reported for CANDU pressure tube. The α grains are much finer and aspect ratio is also small [7]. The β phase is mostly present in the globalised form unlike stringer in CANDU tube and it is substantially richer in Niobium. In order to examine the role of individual fabrication stages, in NFC, a campaign was carried out to fabricate pressure tube with different fabrication parameters. In all, pressure tubes were fabricated by nine different fabrication routes. The objective was to examine which route gives the best desirable microstructural, textural and mechanical properties. The important variations in Zr-2.5Nb pressure tube fabricated through different routes are given below:

![EDS analysis of β phase](image)
(i) Single stage forging and two step forging vs single stage extrusion process for breaking the ingot cast structure.
(ii) Comparison of tube produced with different extrusion ratios.
(iii) Comparison of the tube produced with two stage pilgering and with single stage pilgering
(iv) Role of stress relief treatment after extrusion
(v) Final 25% cold work by single stage drawing versus single stage pilgering.

The important microstructural observations in the pressure tubes fabricated through above routes are summarized below:

1. Two step forging route has shown more homogenous microstructure. Grains were observed to be longer and aspect ratio was much higher, which is preferable.
2. There was variation in microstructure from leading end to trailing end in single stage forging and extrusion processes of breaking cast structure. In general, trailing end microstructure was more lamellar and homogenous.
3. Higher extrusion ratio has resulted in still higher aspect ratio (desired). The variation in microstructure from leading end to trailing end was much smaller in this case and microstructure appeared more uniform. Single pass always resulted in continuous $\beta$ along the $\alpha/\alpha$ interface.
4. Intermediate annealing treatment resulted in the globulization of $\beta$ phase in several regions. The composition analysis has shown that these $\beta$ were enriched to the level of 25 to 45 wt% of Nb.
5. Cold drawn process has shown that microstructure was very close to that observed in the tube produced by CANDU route.
6. The $\alpha$ lamellae in CANDU pressure tubes were much longer up to 20 micron length and $\beta$ phase was continuous and the composition was always in the range of 15 to 20 wt%.
7. NFC produced Pressure Tubes: The $\alpha$ grains were not lamellar in all the regions; the grain size was much smaller. The $\beta$ was not continuous in many regions and they globulaized both at the $\alpha/\alpha$ interface as well as within the $\alpha$ grains. The $\beta$ phase composition varied from 15-45%.

On the basis of this study described in detail in report [8], it has been proposed to fabricate Zr-2.5Nb alloy pressure, by the following fabrication route which involves two stages forming to billet followed by $\beta$ quenching and extrusion to blank with higher extrusion ratio. The blank is then pilgered to final size and autoclaved at 400°C for 24 hours. The microstructure developed during each stage of this fabrication route is described in Fig. 2. The forged microstructure showed Widmanstatten $\alpha$ phase and $\beta$ phase being present at $\alpha/\alpha$ interface (Fig. 2(a)). The grains were uniformly distributed from one end to the other end of the forged billet. The microstructure was also uniform from center to the edge of the billet. The $\beta$ quenched structure showed complete martensitic microstructure. The extrusion of billet with higher extrusion ratio resulted in much longer $\alpha$ phase (15-20 $\mu$m) and the aspect ratio was in the range of 1:10:70. The $\beta$ phase was present as thin film between $\alpha$ lamellae. The single pilgering was done to obtain pressure tube in 25% cold worked condition. Microstructure showed higher density of dislocations within $\alpha$ grain. Autoclaving treatment reduced the dislocation density to some extent. However, the finished tube exhibited lamellar $\alpha$ with stringer of $\beta$ phase in between. No globulisation of $\beta$ phase was seen in this route and EDS analysis has shown that $\beta$ composition was in the range of 15-20%Nb.

**Microstructural Studies on Irradiated KAPS2 Zr-2.5Nb alloy Pressure Tube**

TEM micrographs of the unirradiated S-07 pressure tube off cuts of KAPS2 reactor, is shown in Fig. 3(a). The samples showed typical lamellar microstructure of $\alpha$ and
grains. However, many of the regions exhibited non-lamellar microstructure as well. The β-phase morphology varied from continuous fiber to globular. Niobium content was 10-12% in the continuous film of β and 20-40% percent in case of globulised β-phase. TEM micrographs of the samples obtained from the different locations of the irradiated pressure tube are shown in Figs. 3(b)-(l) [10]. Unlike in the case of the reported irradiated Zircaloy-2 pressure tube samples [9], no significant defects, generated due to irradiation, could be seen. Hydride platelets were also absent in the irradiated Zr-2.5%Nb pressure tube, unlike the irradiated Zircaloy-2 pressure tube samples, which showed significant number of hydride platelets. The grain morphology of α-Zr phase did not change appreciably after irradiation in the Zr-2.5%Nb alloy pressure tube. The major observations in the microstructure in different locations were as follows. In locations from inlet to 381 mm of PT, the microstructure did not change appreciably (Fig. 3(b)). Recovery of a part of the cold work could be seen. The α and β-phase morphology appeared to be similar as seen in the un-irradiated samples. In the region of 381 to 2000 mm from inlet, it was seen that lamellar structure of the α-phase was retained (Fig. 3(c)). Considerable recovery of the cold worked structure could be seen. The β phase had globulised in large number of regions but remained at the α/α interface. Further away (2000 to 4250 mm), the microstructures appeared to have changed considerably and extensive modification in β? morphology was seen (Fig. 3(d-g)). It appeared that β phase had dissolved and re-precipitated within the α-Zr matrix. The volume fraction of the β phase had considerably reduced, suggesting that it had got enriched in Nb in most of the cases. Considering the reduction in the volume fraction of β phase, it is expected that β had got enriched to the level of 50-60% Nb. The β phase had completely globulised and was present within the α lamellae and no β-phase could be seen at α/α interfaces in many regions (Fig. 3(h-j)). The region 4250 mm from inlet to outlet at the end of the microstructure appeared to be similar to those seen near inlet. However, the extent of recovery of the cold worked structure was much higher and the β-Zr was marginally modified (Fig. 3(k-l)).

Conclusions

The present study shows the microstructure at different stages of fabrication characterized by TEM. The microstructure is shown for the heat treated pressure tube fabricated for the first time at NFC Hyderabad. The finished microstructure consisted of fine α lamellae with distribution of nanometer size β phase, along the α/α
interfaces having composition 75-80wt%Nb. The microstructure developed during each stage of the new fabrication route, for fabrication of pressure tube of 700MW PHWR reactor, is also described. The final microstructure comprises of long $\alpha$ grain with higher aspect ratio and $\beta$ being present as thin film having composition 15-20wt%Nb. The study also shows irradiated microstructure characterization at different locations of the first pressure tube obtained from the KAPS2 reactor. The study shows substantial modification in $\beta$ morphology.

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References