Non-destructive evaluation and characterization of Advanced cladding material for extended burn up application in Indian PHWRs


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Abstract
Nuclear Fuel Complex (NFC) is a unique facility of national importance responsible for manufacturing of all the in-core hardware for nuclear power program in India. High burn up is an important consideration for thermal reactor in general and Pressurized Heavy Reactors (PHWRs) in particular. It is always been an attempt to enhance fuel burn up. However limitation for higher burn up comes mainly from cladding material. Zirconium alloy is used as conventional material of construction for cladding in all the PHWRs. Pellet clad interaction-stress corrosion cracking (PCI-SCC) is one of the primary mechanisms limiting clad life. Duplex cladding containing Zirconium barrier layer and Zr-1Nb materials are known as the candidate material to enhance cladding life of PHWR fuel avoiding PCI-SCC failure. In NFC, dia.~15 mm X 0.9 mm thick Zirconium alloy-4 to Zirconium duplex clad and Zr-1Nb cladding were developed to achieve extended burn up in PHWRs. Tubes are produced by a combination of hot-extrusion at high temperature followed by pilgering at room temperature with intermediate vacuum annealing. Inspection and testing of these tubes, especially duplex clad ensuring detection of through wall defect, de-bonding in between layers, has always been challenge. NFC has developed non-destructive evaluation (NDE) techniques, necessary test arrangements for qualification of these tubes. This paper brings out the NDE capability and characterization detail for these duplex clad and Zr-1Nb advance clad demonstrating successful development of these tubes.

Keywords: Duplex cladding, Zirconium alloy, Clad, Zr-1Nb, Non-destructive evaluation (NDE).

1. Introduction:
Zirconium alloy cladding materials are being used for encapsulation of thermal reactor fuel [1]. High burn up is an important consideration for thermal reactor in general and Pressurized Heavy Reactors (PHWRs) in particular [2]. It is always been an attempt to enhance fuel burn up [3]. However limitation for higher burn up comes mainly from cladding material [3]. Pellet clad interaction arising out of fission gas release and stresses due to power ramp at high temperature have been a life limiting factor for high burn-up application [4]. Zirconium alloy duplex cladding with Zr-barrier layer has been identified as candidate material for high burn-up application [4]. Zr-barrier lining has been chosen as liner in order to prevent cack initiation and crack prorogation. In order to exhibit desired in reactor performance, Zr-Sn barrier layer needs to be metallurgically bonded inside the Zirconium alloy-4 base tube forming integral clad. In NFC, duplex clad has been developed by a combination of co-extrusion at high temperature followed by co-pilgering at room temperature with intermediate vacuum annealing.

Zr-1%Nb is another cladding material being widely used as cladding for high burn-up application in PWRs [3, 5]. Zr-1%Nb is an alpha alloy of Zirconium having niobium as major alloying elements. This material is accepted as potential clad for high burn-up application in PHWRs due to it's high toughness, excellent corrosion resistance and low concomitant hydrogen pick-up [4]. NFC has developed thermo mechanical processes (TMP) for manufacturing of these tubes. The major manufacturing steps includes compaction of reactor grade Zr-sponge with alloying elements, electron beam welding of compacts to form
consumable electrodes, triple vacuum Arc melting, two stages of hot extrusion and manufacturing of seamless tube through multi-pass cold pilgering with intermediate vacuum annealing. The final size tubes were heat treated under vacuum, straightened, cleaned, ID grit blasted and polished to achieve the finish tube quality.

Comprehensive Quality Assurance Plan (QAP) was established starting from ingot to finished product to ensure the finished tube requirement. The inspection and testing includes a combination of material evaluation viz. chemical analysis, Mechanical examination, microstructural examination, texture measurement and non-destructive examination viz. visual examination, ultrasonic testing. NDE technique was developed to ensure detection of through wall defect, de-bonding between layers of duplex clad. NDE results were corroborated with finding of microscopic examination.

2. Experimental:

Manufacturing of these tubes was accomplished through two stage hot extrusion of machined ingot into blanks. In case of duplex clad, two billets of suitable sizes were co-extruded during second extrusion forming mother blanks. These blanks are further pilgered through multiple passes employing intermediate vacuum annealing. At final stage, some of the tubes were stress relieved at 498°C/10hrs and remaining tubes were fully annealed at 575°C/3hrs. A detailed QA plan is given fig.1.

Fig-1: Manufacturing and QA plan of advanced cladding tubes for PHWRs.

Tubes were ultrasonically tested at finished stage using computerized probe rotating immersion UT system employing both angle beam and normal beam to ensure material soundness and enhanced reliability. Samples for detailed characterisation were collected from finished tube of both Stress-relieved and fully annealed condition. X-ray diffraction (XRD), Optical microscopy and scanning electron microscopy (SEM) were used for bulk texture, microstructural examination and distribution precipitates respectively. Bulk texture measurements were conducted on a Panalytical XRD system. For micro-structural examination and beta phase distribution, TSL-OIM SEM was used.
3. Results and Discussion

3.1 Zr-1Nb Cladding:

Fig 2 (a-b) shows the SEM images of stress relieved tubes samples. Micro structural observations suggest that the present TMP steps followed in the fabrication of Zircaloy-1% Nb tube material results in a partially recrystallized microstructure in the finished product. The beta particle size distribution was found uniform of avg. size of 50 nm size (30 nm to 104 nm) and Nb content in Beta phase is found to be 70-80%. On the other hand, Fig 3 (a-b) shows the SEM images of stress relieved tubes samples. Micro structural observations revealed a recrystallized microstructure in the finished product. The beta particle size distribution was found uniform of avg. size of 57 nm(28 nm to 136 nm) and Nb content in Beta phase is found to be 70-80%.

Fig-2(a-b) : Shows the typical SEM images of longitudinal section of the Zr-1% Nb tube (15mm OD X 0.9 mm WT), stress relieved at 498 deg C/10 h , (a) grain structure at 2 kX; b) beta particles at 40 kX.

Fig-3: Shows the typical SEM images of longitudinal section of the Zr-1% Nb tube (15.2 mm OD X 0.9 mm WT), annealed at 575 deg C/3 h,(a) grain structure at 2 kX; b) beta particles at 40 kX.
Table 1 shows the textural results (in terms of Kearns’s factor which is the distribution of basal fiber along given direction of the sample) obtained using XRD. It was observed that basal fibers were mostly distributed in plane defined by axial direction. These textures correspond to typical deformation and annealed textures in zirconium alloy where prismatic slip is the predominant deformation mechanism. Texture measurement was confirmed through artificial hydrogen charging on finished tube samples and Hydride orientation factor estimation in accordance to ASTM-B-811. Optical micrographs of hydrided sample are shown in Fig.4 (a-b) and Fn value was estimated to be 0.02-0.09. Predominance of circumferential hydride was seen in both stress relieved and fully annealed condition.

<table>
<thead>
<tr>
<th>Condition</th>
<th>$f_r$</th>
<th>$f_i$</th>
<th>$f_a$</th>
</tr>
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<tbody>
<tr>
<td>Stress relieved</td>
<td>0.60</td>
<td>0.33</td>
<td>0.06</td>
</tr>
<tr>
<td>Fully annealed</td>
<td>0.63</td>
<td>0.31</td>
<td>0.05</td>
</tr>
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Fig.4: Optical micrograph of hydrogen charged sample for (a) stress relieved and (b) fully annealed tubes revealing predominance of circumferential hydride platelets.

All the finished tubes (both fully annealed and stress relieved) were examined for flaw detection and measured for outer diameter, wall thickness employing an automatic probe rotating UT system. The UT record of reference flaw standard is given in Fig.5. Fig.6 (a-b) showed the UT record for deviated and acceptable tube respectively.

Fig.5: UT chart for Reference flaw standard for Zr-1Nb tube
3.2 Duplex Cladding:

Finished duplex cladding were subjected to non-destructive evaluation on automatic probe rotating UT system. Flaw detection and measurement of outer diameter, wall thickness was carried out on an eight channel system with high frequency probes. Bi-directional angle beam probes were employed for through thickness flaw detection. Normal beam channels were calibrated to ensure detection of de-bonding at interface. Samples were collected from both UT accepted, defective regions and subjected to microscopic examination. UT records of acceptable tubes is shown in fig.8. Typical Optical microstructure of accepted Zirconium alloy-4 to Zr-Sn duplex cladding is shown in fig. 9 (a-b) revealing complete bonding. Both the layers revealed recrystallized equiaxed grains structure in fully annealed condition. Stress relieved samples did not show equiaxed grains in base metal; however liner region had equiaxed grain.

UT chart of duplex tubes indicating de-bonding in the normal beam channel is shown in fig. 10(a-b). The optical micrographs of defective regions indicated by UT were examined under microscopy revealing de-bonding [fig.10 (c-d)]. Thus normal beam UT response could be correlated to optical microscopic examination demonstrating NDE capability for detection of interface de-bonding. Abnormal wall thickness variation along the tube length as indicated in UT were subjected to optical microscopy [Fig. 11(a)]. Fig.11 (b-c) shows optical microstructure of defective regions revealing variation or absence of liner. UT findings were found in line with microscopic observation. Stringent artificial flaw standard was used for
sensitivity setting employing bi-directional angle beam. UT rejected regions indicating circumferential defects in transverse scanning [fig.12(a)] were examined under optical microscope which revealed real transverse flaws on inner surface of tube [fig. 12(b-c)].

![UT chart of acceptable duplex clad (Sample No. S4)](image)

**Fig.8:** UT chart of acceptable duplex clad (Sample No. S4)

![Optical micrographs duplex clad showing complete bonding between base metal and liner (Sample No. S4).](image)

**Fig.9** (a-b): Optical micrographs duplex clad showing complete bonding between base metal and liner (Sample No. S4).

![UT Chart of duplex clad showing indicating de-bonding between liner and base metal in normal beam channel, (b) Zoom out image showing UT echo pattern near de-bonding region.](image)

**Fig.10:** (a) UT Chart of duplex clad showing indicating de-bonding between liner and base metal in normal beam channel, (b) Zoom out image showing UT echo pattern near de-bonding region.
Fig. 10 (c-d): Optical micrographs of UT defective regions showing the de-bonding between liner and base metal.

Fig. 11 (a): Wall thickness pattern on UT indicating variation in liner thickness.

Fig. 11 (b): Optical micrographs showing gradual decrease in liner thickness and finally absence of liner, (c) Variation in liner thickness.

Fig. 12 (a): UT chart indicating circumferential (Transverse) defect in transverse scanning.
Fig. 12 (b-c): Optical micrographs showing defects in liner co-relating UT response.

4. Conclusions

- Micro-structural observations suggest that the present TMP steps followed in the fabrication of Zr-1Nb tube results in a partially and completely recrystallized microstructure in the stress relieved and full annealed tubes respectively. Beta phase was found uniformly distributed with avg. size of 50 nm.
- The process route followed for duplex clad results in successful developed of these tubes ensuring complete metallurgical bonding between base metal and liner.
- UT results were found in agreement with micro-structural examination demonstrating NDE capability to ensure detection of through wall defect and de-bonding between layers.

5. References


