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MECHANICAL BEHAVIOUR OF HIGH BURNUP PHWR CLADS

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Abstract

The clad tubes being the primary barrier between the irradiated fuel and the hot heavy water coolant, their integrity is essential for good fuel and plant performance. During their in-reactor residence the clad tubes are exposed to severe operating conditions i.e., high temperature, corrosion, erosion, irradiation, and stresses. Therefore, the structure and properties of the clad tube change significantly from that of as manufactured unirradiated cladding. Due to the corrosion reaction with hot water, Zircaloy clad picks-up hydrogen/deuterium over the operating time. In addition, in the case of clad tube defects, the coolant enters inside the tube and leads to a large localized clad oxidation and high up-take of hydrogen in the clad alloy. This hydrogen/deuterium in excess of its maximum solubility precipitates as zirconium hydride which may cause severe embrittlement of the clad tubes. Ensuring a minimum amount of strength and ductility in the clad along the circumferential direction is one of the most important ways of ensuring its integrity. These properties are evaluated using burst test, ring tension test and several other non-conventional test techniques.

The present work involves study of mechanical properties and fracture morphology of transverse ring tension specimens taken from PHWR fuel clads, from different fuel bundles with burnup up to 22,000 Mwd/TU. One of the defective bundles had an end-cap weld defect, in one of the outer pins, that lead to clad tube punctures and was discharged at a lower burnup level, was also studied.

The test results indicate an increase in transverse strength and decrease in ductility of the clad due to in-reactor operation. This study showed that the low and high burnup clads, in absence of zirconium hydride precipitates at reactor operating temperature, had higher strengths and retained adequate ductility levels. The circumferential tensile loading resulted in three very distinct type of failure and fracture edges, depending on the hydride platelet density and burnup levels; (a) flat cleavage type rupture, on radial-axial plane, (b) shear band failure at 45 degree to the tensile loading axis, and (c) a more gross deformation along the specimen width and thickness with ductile rupture. Fracture in the irradiated clad is by the shallow ductile rupture of the ligaments between the cracked hydride platelets. The failure in the irradiated fuel clad, occurs with formation of shear band and the fracture surface is inclined nearly 45 degrees to the tensile loading axis. Defective fuel clad, with high hydride density, is severely embrittled at lower test temperature but its ductility improves at higher test temperature.

1 Introduction

Natural uranium oxide fuel is used in Indian pressurized heavy water reactors. The oxide fuel pellets are enclosed in thin zirconium base alloy tubes, 15.2 mm outside diameter and 0.4 mm nominal thickness, and assembled in the form of fuel bundles [1]. Zircaloy-2 (earlier) and Zircaloy-4 (present) alloys have been used for fabricating the fuel clad tubes. The clad tube being the primary barrier between the irradiated fuel and the hot heavy water coolant, its integrity is essential for satisfactory fuel and plant performance. After typical average fuel burnup of around 7000 MWd/T the fuel bundles are discharged from the reactor core [1]. During their in-reactor residence, the clad tubes are exposed to severe operating conditions e. g. , high temperature, corrosion, erosion, irradiation, and stresses. Therefore, the microstructure and properties of the clad tube changes significantly from that of fresh unirradiated cladding. Due to the corrosion reaction with hot water coolant, zirconium alloy picks-up hydrogen over the time. In addition, in the case of clad tube puncture or leakage, the coolant enters inside the clad tube and leads to large localized clad oxidation and up-take of hydrogen in the clad alloy. Hydrogen in excess of its maximum solid solubility in the alloy, precipitates as zirconium hydride which may cause severe embrittlement of the clad tubes. Initially the clad is under compressive load due to high coolant pressure, the thin clad slowly collapses on to the hard ceramic fuel pellet and the pellet-clad gap becomes nearly zero. The clad tube under such conditions is subjected to tensile hoop stresses during power transients due to differential thermal expansion of the fuel and clad tube. The point of pellet-clad contact may lead to multiaxial loading and had been a cause of fuel failure. The clad tubes must have some residual ductility to accommodate the strains induced due to service loads.

The present work involves study of transverse (circumferential) tensile deformation and fracture behaviour of irradiated Zircaloy-4 clad tubes taken from different fuel bundles of 220 MWe Indian PHWRs, that were discharged after different burnup levels. One of the fuel pins of a fuel bundle had end-cap weld defect that lead to secondary hydriding and developed leakage and was discharged at a low burnup level. The effect of fuel pin leakage has also been studied. The transverse tensile tests were performed using the the ring tension test methodology.

2 Experimental

2.1 Material

The fuel clad specimens were taken from the outer pins of following three fuel bundles, from 220 MWe PHWRs, which have 19 pin configuration; (a) a fuel bundle which had seen average burnup of around 10,000 MWd/tU (b) another fuel bundle which was deliberately irradiated for longer time till higher burnup of 22,000 MWd/tU (c) a fuel bundle, which had end-cap weld defect in one of the outer pins and the bundle was discharged at a lower burnup level of around 4500 MWd/tU. The failed fuel pin had an end-cap lack of fusion weld defect which led to the secondary hydriding and puncture of clad tube at two locations. A small puncture at one end of the fuel pin near the end-cap weld and another larger hole (~3 mm diameter) beside the bearing pad near the center of the pin. All the specimens, from the defective fuel pin, were taken from the region adjacent to the central hole. The fuel bundles were cut and dismantled earlier to separate out the individual fuel pins.

All these fuel bundles had fuel clad made of cold worked and stress relieved Zircaloy-4 as per the standard fabrication route followed by NFC, Hyderabad. The alloy composition of the standard Zircaloy-4 cladding is approximately (in wt.%) 1.2-1.7 Sn, 0.18-0.24 Fe, and 0.07-0.13 Cr. It also contains hydrogen < 20 wppm and oxygen 0.09-0.14 wt.%. The solubility of Fe and Cr in the alloys is, however, very low. Therefore, most Fe and Cr atoms in unirradiated Zircaloy exist either in a metastable state as solute atoms or as $Zr(Cr,Fe)_2$ precipitates [5]. The Zircaloy is essentially a single phase alloy with fine intermetallic precipitates.

The Zircaloy clad tubes are fabricated at Nuclear Fuel Complex (NFC), Hyderabad, India. The 15.4 mm outside diameter and 0.4 mm wall thickness, Zircaloy fuel clad tubes are manufactured by a combination of thermo-mechanical processes [2]. The clad tubes are made from vacuum arc melted ingot which is hot extruded in billet form, and then billet is given a β-quench treatment. The β-quenched billet is further hot extruded to get hollow tube form. After vacuum annealing of this hollow tube form, further reduction in the tube wall thickness and diameter is achieved in three stages of cold pilgering

Figure 1: Burst tested specimens from As-fabricated clads.

with intermediate annealing operations. After final stage of cold pilgering, which gives high degree of cold work, the clad tubes are stress relieved at 773 K [2]. Anisotropy due to hexagonal crystal structure of the α-zirconium, leads to a strong texture during fabrication process. A high wall thickness and a low diameter reduction during the cold pilgering process leads to radial basal pole texture in the clad tubes. The average values of f-parameter, which is fraction of basal poles around a given direction, is 74%, 22% and 4% along the radial, tangential and axial direction of clad tube respectively [2]. Due to strong radial basal pole texture of the clad tubes, the hydrogen / deuterium present in the clad material precipitates out as zirconium hydride platelets along the circumferential direction.

2.2 Burst Tests of As-Fabricated Clad Tubes

As fabricated PHWR clad tubes are subjected to ambient temperature burst tests as a quality control check at Nuclear Fuel Complex, Hyderabad. The burst tests used specimen length of 200 mm. Burst tests are carried out after sealing both the tube ends, which are flared to fit into tapered end plugs. One end of the tube is supported and other end is kept free. The tubes sections were slowly pressurized using water from a displacement / diaphragm pumps. This is a close end burst test where both hoop and longitudinal stresses are present due to pressurization. Typical appearance of a tested specimen is shown in Fig. 1. A thin metallic tape is wound around the ruptured region of the tube to measure the circumference and to obtain the value of circumferential elongation. The maximum pressure denoted as burst pressure and the total circumferential elongation (TCE) after the test are recorded.

As-Fabricated PHWR clad tubes typically show burst pressure of 51-54 MPa, corresponding to ultimate hoop stress of 940-990 MPa and TCE of 10-13 %.

2.3 Specimen Preparation

The ring specimens (3.5 mm wide) were cut from one end of the fuel pins. In the case of failed low burnup fuel pin the ring specimens were taken from the region adjacent to the hole near the central bearing pad to asses the local changes in the clad properties. The specimens were cut inside hot cell using slow speed cut-off wheel. The cut specimens were defueled and cleaned using a small fixture. In majority of the cases the cracked oxide fuel pieces fell off the clad ring on its own. The width of the ring specimens were measured inside the hot cell using micrometer attachment. The specimens were then transferred to another cell having a screw driven universal testing machine for the ring tension tests.

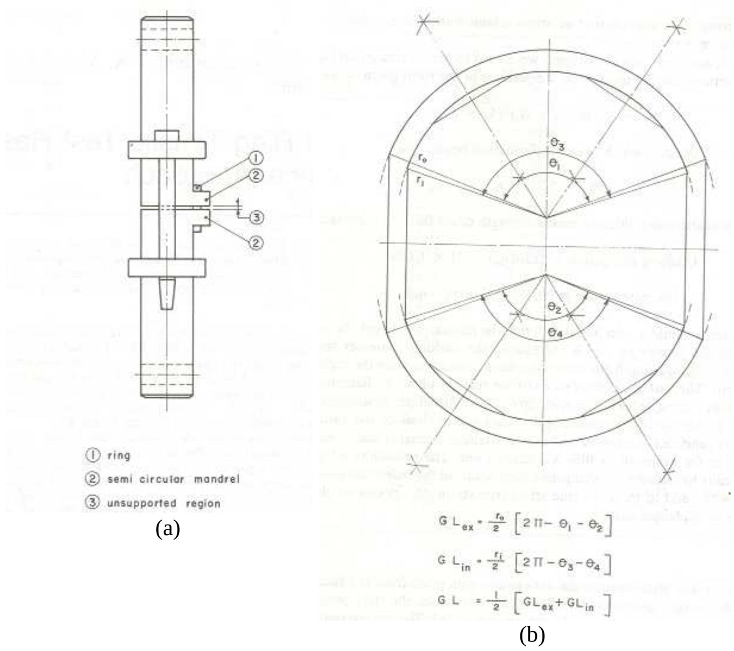


Figure 2: (a) Schematic of ring tension test fixture and (b) effective gage length (GL) determination.

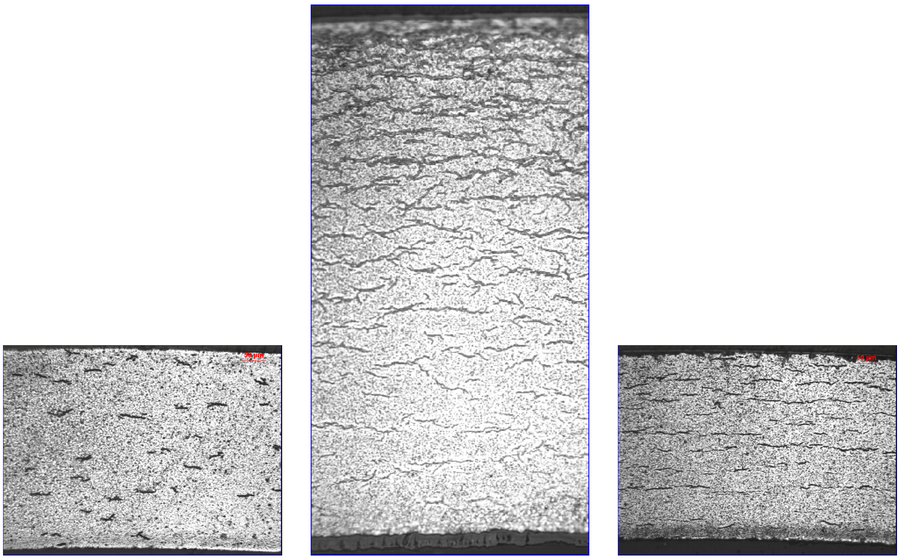
2.4 Ring Tension Test

Ring tension tests (RTT) were carried out using around 3.5 mm wide rings cut from the clad tubes. These rings were pulled along the hoop direction using two split semi-circular mandrels with a curvature that fitted into the inner diameter of the rings as shown in Fig 2. The cross-head speed of the universal screw driven testing machine was kept constant at 0.5 mm/min. The ring specimen has uniform width all along the circumference, without any reduced gage portion, but deformation is mainly concentrated near the center of the split portion of the grip. The top and bottom portions of the ring being in close contact with mandrel do not deform. Several methods like chemical grid etching, photo grid etching, profile projection technique can be used to estimate effective gage length of ring tensile specimen. In present work, profile projection method has been used to determine the undeformed circumference after the test of the ring specimen [3]. Half of the difference between the total original circumference of the ring and the undeformed circumference after the test has been taken as the effective gage length for the specimen. The effective gage length was found to be 5.2 mm in this work and all the elongation values reported here are based on this gage length.

The tests at 583 K were carried out using an electrically heated environment chamber. The universal testing machine had an interface with personal computer for acquisition of load and cross head displacement data during the tests. This data has been using to obtain the tensile stress-strain curve for the clad tubes.

2.5 Metallography, Fractography and H Content Analysis

Few of the ring tension tested specimens were prepared for optical metallography by carrying out metallographic polishing and etching inside hot cells. The microhardness measurement were carried using an attachment of the remote optical microscope. The fractured region of the specimens were also examined using scanning electron microscope. Some of the specimens were used to measure equivalent total hydrogen content in the tested clad specimens using differential scanning calorimetry method.



(a) (b) (c)

Figure 3: The hydride distribution across the clad thickness (bottom is tube inner diameter) for (a) as fabricated unirradiated tube, (b) low burnup defective fuel pin specimen, (c) high burnup clad tube.

3 Results

3.1 Microstructure

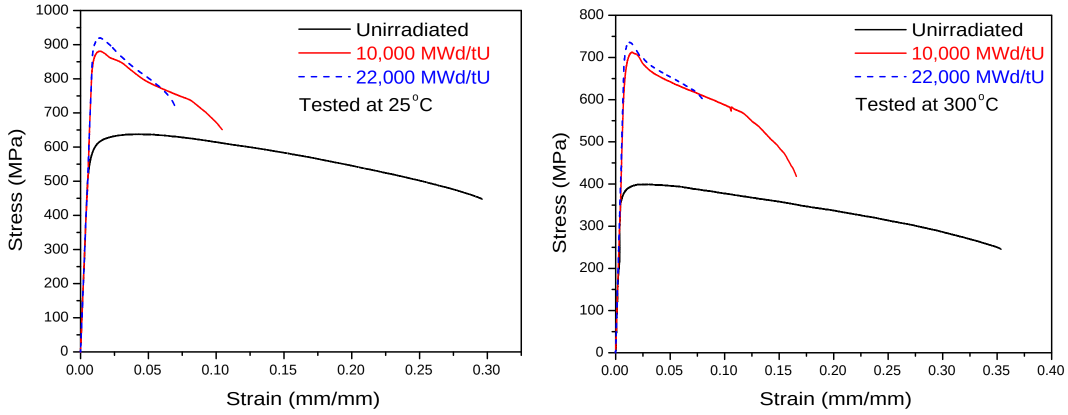
The optical metallographs showing the transverse cross section of the fuel clad and the distribution of hydride are given in Fig. 3. As fabricated clad specimen had around 18 wt. ppm of hydrogen (Table 1) and the hydrogen content in the clad was found to increases with the burnup to a level of 45 ppm for a burnup level of up to around 22,000 MWd/TU. The hydride platelets in clad tubes were found to be oriented predominantly in the circumferential direction of the tube. The as fabricated clad material, Fig. 3(a), had small volume fraction of short hydride platelets distributed uniformly across the tube thickness. The hydride volume fraction increased in the high burnup clad, Fig. 3(c). However, the specimens from the region around the hole in the failed fuel pin had much higher hydrogen level (335 wt. ppm), as indicated by the high hydride density in optical metallograph Fig. 3(b). The defective fuel pin had higher density of hydride platelets at the outer circumference of the tube (coolant side), which decreased toward the clad inner side (fuel side). This is expected behaviour as hydrogen is known to move from higher temperature region towards lower temperature region which is the clad periphery in this case.

The Vickers micro-hardness measurements (using 100 gm load) across the tube thickness gave an average hardness value of 210, 245, and 300 VHN for as fabricated clad, average burnup clad, and high burnup clad respectively. Irradiation increases the hardness values, high burnup clad having the highest hardness value.

The as fabricated clad has negligible oxide layer thickness on the clad inner and outer surfaces. However, during service the outer surface of the fuel clad, which is in contact with hot water coolant, gets oxidized slowly and part of the hydrogen liberated during the corrosion reaction is absorbed by the zirconium metal matrix. A slow build up of oxide layer has been observed on the clad outer surface with increase in burnup levels. The oxide layer thickness on the clad outer diameter was measured to be 2.44 and 2.82 μm for the average and high burnup fuel clads. No measurable oxide layer was found on the inner diameter of the irradiated clads, which is on the fuel side. However, in case of defective fuel pin, in the region near the leaking hole, a thick (9.0 μm) oxide layer was observed on the outer clad surface

Table 1: Circumferential tensile properties and measured total equivalent hydrogen content of the clad tubes.

Material	Test Temperature	H Content (Equivalent)	Yield Strength	Ultimate Tensile Strength	Uniform Elongation	Total Elongation
	(K)	(wt. ppm)	(MPa)	(MPa)	(%)	(%)
As fabricated clad	300	18	570-620	638-700	3.7-4.4	28-30
	583	18	370-375	400-405	2.6-2.8	34-36
Low Burnup Failed clad	300	334	617	621	0.95	1.14
	583	334	550	627	2.0	10
Average burnup clad	300	35	795-955	840-1010	1.4-1.8	6-10
	583	35	670-690	690-695	1-1.5	12-24
High burnup clad	300	45	850-910	890-925	1.4-1.5	5-7
	583	45	625-730	640-760	1.1-1.4	8-22



(a) (b)

Figure 4: Circumferential tensile deformation behaviour of low and high burnup fuel pin clads, at ambient temperature (300 K) and at 583 K.

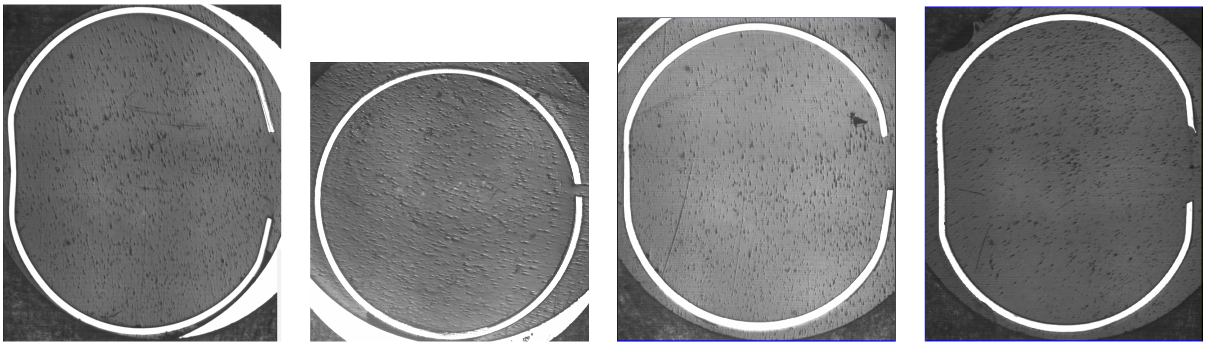
and around 3.0 μm thick oxide layer was found on the clad inner surface. This is clearly due to ingress of the high pressure, hot water coolant inside the clad tube that lead to localized oxidation and hydrogen pick up by the clad near the hole.

3.2 Transverse Tensile Behaviour

The circumferential tensile properties of the clad tube specimens are given in Table 1. The table also gives the total measured equivalent hydrogen content (including deuterium) in the specimens. The engineering stress-strain curves for the different clad tube specimens are shown in Fig. 4.

Ambient Temperature Behaviour

The amount of gross deformation, during RTT at ambient temperature, for different clad tubes can be seen from the shape of the tested ring specimens given in Fig. 5. The close up of the fractured edges of two specimens are shown in Fig. 6. As can be seen in Fig. 5(b) the gross deformation in the gage



(a) (b) (c) (d)

Figure 5: The ring cross section after tests at ambient temperature for (a) unirradiated, (b) low burnup defective fuel pin, (c) high burnup pin and (d) average burnup fuel pin.

portion of the defective fuel clad specimen is severely limited at ambient temperature and the ring specimen remains nearly circular even after failure. The fracture edge of defective fuel clad (Fig. 6(a)) shows negligible deformation at ambient temperature and has a radial-axial fracture plane that is nearly perpendicular to the loading axis. In contrast, as fabricated clad tubes and clad tubes having low and average burnup showed higher elongation at ambient temperature. The observed deformation and ductility level in the high burnup clad at ambient temperature is that of intermediate level, around 25% of that of as fabricated clad material.

At ambient temperature the defective fuel pin, having around 330 ppm of hydrogen in the form of high density of circumferential hydrides, showed small ductility 1.14 %. However at higher test temperature of 583K the ductility level in the defective fuel pin also improved to levels similar to non-defective clads.

Material hardening and loss of clad ductility due to high energy neutron irradiation is evident from ambient temperature tensile stress-strain plot given in Fig. 4(a). The drop in ductility at ambient temperature is more in the non-uniform part of the strain, though uniform elongation also decreased (Table 1). At ambient temperature, the circumferential ductility in low and high burnup fuel clads drop to nearly 25 % of that in as fabricated clad tube.

Behaviour at 583 K

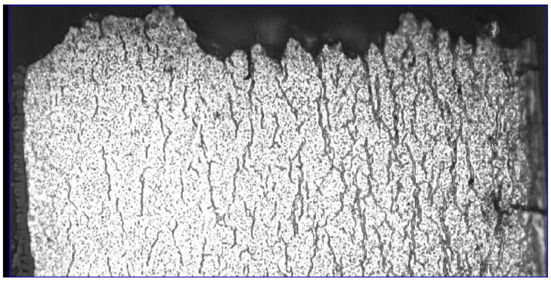
Tests near reactor operating temperature ($\approx$583K) of the clad tubes show an increase in the ductility levels for the low as well high burnup fuel clad tubes. The higher flow stress due to irradiation hardening is evident from the Fig. 4(b) at this temperature. At reactor operating temperature all the clads showed some level of ductility and gross deformation before failure. The defective fuel clad specimens having high hydride platelet densities were also able to undergo gross deformation at these temperature. The brittle to ductile transition in the ductility values for the hydrided zirconium alloy with temperature has been observed in many other studies [4,5].

3.3 Fracture Morphology

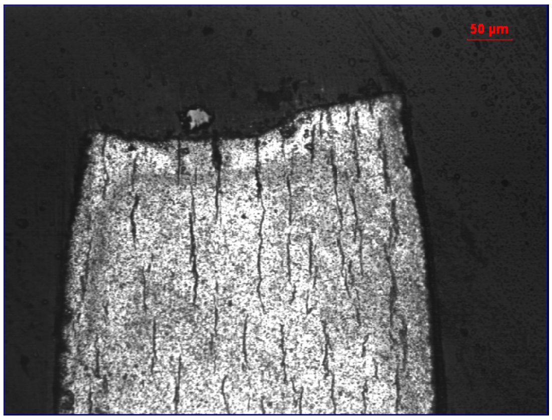
Ambient Temperature

At ambient temperature the circumferential tensile loading resulted in three very distinct type of failure and fracture edges;

- fracture after gross deformation and necking, as seen in as-fabricated clads, in Fig. 6(a)



(a)



(b)

Figure 6: The fractured edge of the ambient temperature tested specimens, showing the full thickness, of (a) defective low burnup fuel clad and (b) for high burnup fuel pin.

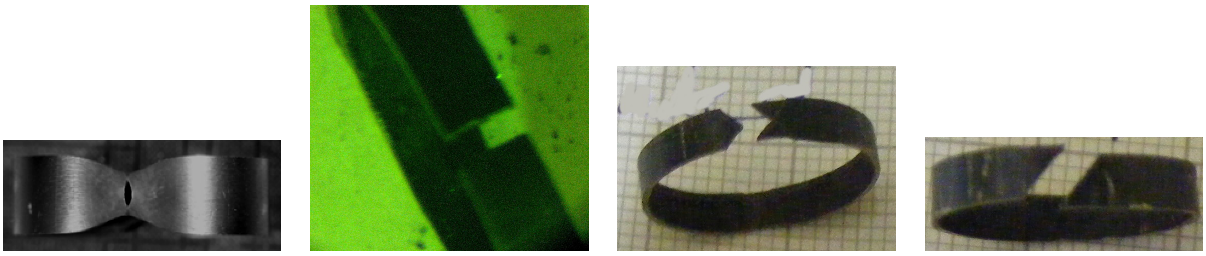
- flat cleavage type rupture, on radial-axial plane, with little deformation along the thickness and axial direction of the specimen, as seen in the defective fuel clad in Fig. 6(b), a material having high density of hydride platelets combined with irradiation damage.
- shear band failure at 45 degree to the tensile loading axis, with some amount of width contraction and limited thickness deformation, as seen in Fig. 7(c) and (d), for low and high burnup fuel clad that had smaller volume fraction of hydride in matrix. The measured ductility is higher when the shear band starts from the center of the specimen, Fig. 7(c), rather from the edge, Fig. 7(d).

It is clear from the RTT results that volume fraction of hydride platelets, their size and distribution play an important role in fracture of the clad tube at ambient temperature. The close up view of the fracture edges, of ambient temperature tested specimens, is shown in Fig. 6. In case of the defective fuel clad, Fig. 6(a), the high hydride platelet density leads to numerous crack formation during loading which get linked up in a zig-zag manner with small amount of slip but maintaining a nearly flat fracture plane perpendicular to loading axis.The high burnup fuel clad, Fig. 6(b), with lower volume fraction of hydride is able to undergo some deformation along the thickness.

The fractographs of the ambient temperature tested ring tension specimens are shown in Fig. 8. As fabricated, unirradiated clad tubes show fully ductile rupture with prominent neck formation and thickness contraction. The flat fracture plane of failed fuel pin shows closely spaced deep hydride cracks joined together with shallow deformed layer. The fracture plane of high burnup fuel clad showed less amount of hydride cracking and more fibrous appearance, though much flatter than the as fabricated clad material. Fracture in the irradiated clads is by the ductile rupture of the ligament between the cracked hydride platelets.

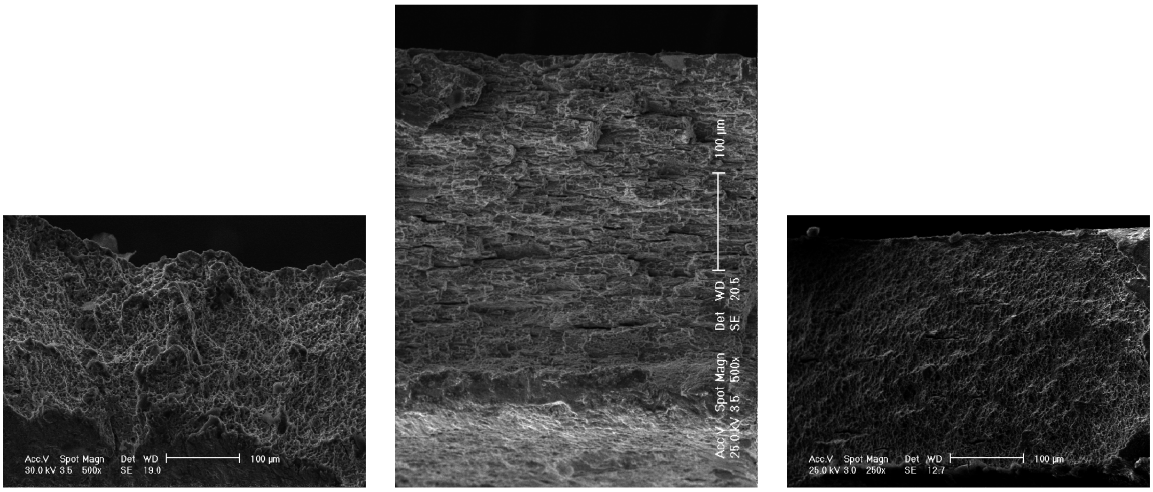
At 583 K

Hydrided Zircaloy show a transition in fracture behaviour from brittle to ductile fracture with increase in test temperature. The transition temperature has been reported to be around 423 to 443K for materials having few hundred ppm of hydrogen [5,6]. This transition is mainly attributed to the softening of zirconium alloy matrix. As the temperature at loading increases the hydride cracking or interfacial cracking density reduces due to strain accommodation in softer matrix and the required fracture energy increases rapidly. The fracture edge of the specimens tested at 583K is shown in Fig. 9. The gross deformation



(a) (b) (c) (d)

Figure 7: The photographs of the ambient temperature tested ring specimens for (a) as-fabricated clad, (b) defective low burnup fuel clad, (c) high burnup pin type-I, and (d) high burnup clad type-II.



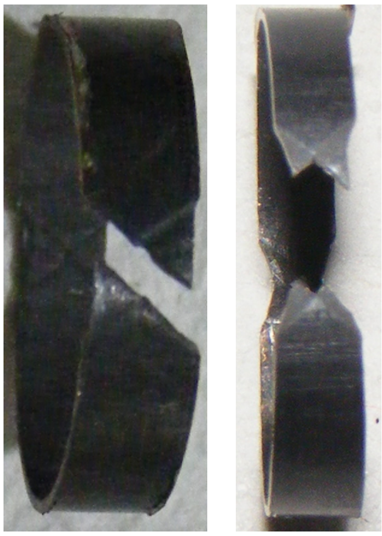
(a) (b) (c)

Figure 8: The SEM fractographs showing radial-axial plane for (a) as fabricated clad material, (b) failed low burnup fuel pin and (c) high burnup fuel clad.

and neck formation before fracture is clearly seen in high burnup clad at 583K, in Fig. 9(b). At this temperature, the fracture becomes more fibrous, with less hydride related crack formation, near the fracture plane. This has also been observed in earlier studies on unirradiated hydrided specimen conducted by author [6]. However, the fracture plane is at 45 degrees to the tensile axis showing shear localization.

4 Discussion

The mechanical properties of fuel rod Zircaloy cladding have been known to change with irradiation because of damage induced from the fast neutron fluence, clad oxidation and hydrogen ingress in the clad matrix. The hydrogen will migrate to the cooler regions of the cladding and precipitate as zirconium hydride when the concentration exceeds the solubility limits for a given temperature. The hydrides have been found to preferentially precipitate perpendicular to the tensile stresses and parallel to the compressive stresses. Also the hydride platelets have been found to prefer plane close to the basal plane of α-zirconium grains. Initially, the fuel cladding hoop stress remains compressive due to higher coolant pressure than inner gas pressure. Therefore, both the radial basal pole texture of fuel clad and compressive hoop stress in the clad tube promote circumferential hydride precipitation in the fuel clads. As long



(a) (b)

Figure 9: The photographs of the ring specimens of high burnup fuel clads tested at 583K showing (a) type I and (b) type II of failure.

as the concentration of hydrogen remains below its solubility limit, its effect on the clad ductility is limited. When the fuel-to-cladding gap closes, the cladding may get subjected to tensile hoop stresses during power transient due to differential thermal expansion and hydrides may become radially orientated with respect to the axial direction. In this situation, the hydrides become crack initiation points perpendicular to the stresses and can penetrate the cladding in the radial direction. The latter phenomenon can lead to cladding ductility near zero.

The irradiated fuel clad has high density of irradiation induced defect density which leads to the increase in flow stress of the irradiated material. Such structure show a manifestation of dislocation channelling and deformation band formation [4]. Under high stresses as dislocations move on a slip plane, during its motion the radiation induced defects are annihilated and subsequent dislocation motion on such defect denuded plane becomes easy. This leads to formation of deformation band that is nearly at 45 degrees to the tensile load axis. This behaviour is clearly seen in the low and high burnup clads. The presence of hydrides at deformation temperature may synergistically interact with these dislocation channels to promote cracking and fracture [5].

The clad materials studied in this work, except the defective fuel pin, had maximum total hydrogen content of ≈45 ppm. This is less than the terminal solid solubility of hydrogen (≈70 ppm) in Zircaloy-4 at 583 K. Thus it is expected that majority of hydrides will get dissolved in the matrix at the reactor operating temperature and shall not deteriorate the clad ductility. This is confirmed by the fact that the clad ductility of the irradiated clads, at reactor operating temperature, has been found to decrease to smaller extent as compared to ambient temperature. This reduction in ductility is mainly due to the irradiation hardening. However, at ambient temperature the presence of circumferential hydrides in the clad matrix leads to a large reduction in ductility levels. The brittle zirconium hydrides crack under stress and acts as numerous crack initiation sites that need little plastic work for linking up.

5 Conclusion

- Low and high burnup clads, in absence of zirconium hydride precipitates at reactor operating temperature, showed higher strength and adequate ductility levels. However, at ambient temperature the ductility levels are limited to around 25 % of that observed in as fabricated condition.
- The failure in the irradiated fuel clad occurs with formation of shear band and the fracture surface is inclined nearly 45 degrees to the tensile loading axis.
- Defective fuel clad, with high hydride density, is severely embrittled at lower test temperature but its ductility improves significantly at higher test temperature.
- Fracture in the irradiated clad is by the shallow ductile rupture of the ligament between the cracked hydride platelets. Presence of large density of hydride platelets at the test temperature has a dominant role in reduction of clad ductility.

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