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Microstructural Evoluation in Wolsong 1 Zr-2.5Nb tube with Neutron Irradiation

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Abstract

With the aim of assessing the degradation of Zr-2.5Nb pressure tubes operating in the Wolsong Unit-1 nuclear power plant, characterization tests are being conducted on irradiated Zr-2.5Nb tubes removed after a 10-year operation. The examined tube had been exposed to temperatures ranging from 264 to 306 ^{o}C and a neutron fluence of $8.9x10^{25}$ n/m^2 (E>1MeV). A change in the a-type and c-type dislocations were determined using TEM at the three different locations of the irradiated Wolsong 1 Zr-2.5Nb tube to evaluate the neutron irradiation effect. Further, the phase decomposition of the β-Zr phase with the neutron irradiation were also investigated using selected area diffraction patterns on the foil specimens and the electron diffraction spectroscopy analysis on the extracted particles using carbon replicas. Neutron irradiation up to $8.9x10^{25}$ n/m^2 (E>1MeV) yielded an increase in an a-type dislocation density to $7.5x10^{14}$ n/m^2 from $4.0x10^{14}$ n/m^2, corresponding to the a-type dislocation density of the unirradiated Zr-2.5Nb tube. An increase in the a-type dislocation density was found to be the largest at the inlet part of the irradiated tube that was exposed to the lower temperature. However, there is no change in the c-type dislocation density at the neutron fluence of up to $8.9x10^{25}$ n/m^2 (E>1MeV). The decomposition of the β-Zr phase was higher at the outlet part of the tube exposed to higher temperature compared to the other parts while the lower Nb concentration in the β-Zr grains was observed at the inlet part region. The effect of the microstructural evolution of Zr-2.5Nb tubes on the in-reactor performances is discussed.

1. Introduction

Zr-2.5Nb pressure tubes that have a function to carry fuel bundles and coolant inside are the most critical components to the safety and capacity factors of the heavy pressurized water-cooled reactors. One of the current hot issues is evaluation of mechanical integrity of

Zr-2.5Nb tubes irradiated to the fluence of $30x10^{21}$ n/m^2 corresponding to their design lifetime. It is because the Zr-2.5Nb tubes have an enhanced degradation with time, causing their earlier replacement before their reaching the design lifetime [1]. The main factors that are known to govern the integrity of the Zr-2.5Nb tubes are firstly, their dimensional change by irradiation growth and creep, secondly, embrittlement by neutron irradiation and delayed hydride cracking at the surface flaws or near the rolled joint regions with very high residual tensile stress. A problem is little understanding of what govern the degradation of the Zr-2.5Nb tubes with reactor operating conditions. Thus, the best strategy chosen by the CANDU nuclear industry is periodic surveillance tests on the Zr-2.5Nb tubes to check if there is a sudden change or acceleration of their in-reactor performances with time or neutron fluence. To this end, we are conducting characterization tests on the Zr-2.5Nb tubes that are withdrawn from the Wolsong1 nuclear power plants (NPPs). In this paper, we investigated a microstructural change in the Zr-2.5Nb tube removed after 9.3 EFPY operation in the Wolsong1 NPPs as a function of temperature and fluence and its correlation with the mechanical properties.

2. Experimental Procedures

The examined Zr-2.5Nb tube was operated in the M-11 channel of the Wolsong1 NPP for 9.3 effective full power years. To investigate the effect of temperature and neutron fluence on the microstructures of the Zr-2.5Nb tubes, we took 3 tube rings of 170 mm long from the inlet, the middle and the outlet end of the tube and conducted the microstructural examinations, tensile tests, DHC tests, fracture toughness tests. Fig. 1 shows the distribution of the averaged temperatures and neutron fluence of three tube rings. The inlet tube ring was exposed to the lowest temperature and relatively higher fluence which is higher than that of the outlet tube ring. However, the middle tube ring had the highest neutron fluence and its operating temperature is in the middle between the those of the inlet and outlet tube rings. the microstructural examination, TEM was used to analyze the dislocation density and the Nb concentration in the β-Zr grains. Thin foils of 3mm in diameter and 0.1 mm thick were electropolished in a solution of 10% perchloric acid and 90% ethanol at about –35 ^{o}C at the 20 voltages using a Tenupol-3 twin jet apparatus.

3. Results and Discussion

Fig. 2 shows the elongated grain structures of the Zr-2.5Nb tube before and after neutron irradiation. Here, the Zr-2.5Nb tube before irradiation represents the back-end off-cut of the irradiated Zr-2.5Nb tube. The neutron irradiation did not change the macroscopic elongated shape of α-Zr grains but produced some dark images on them, which results from the small black dots or a-type dislocation density as shown in Fig. 3. In other words, the neutron irradiation produced lots of the small a-type dislocations or dislocation loops in the α-Zr

grains, which were not observed before the neutron irradiation (Fig. 3). Fig. 4 shows the c-type dislocation density in the unirradiated and irradiated tubes. It seems that the c-type dislocation density does not change much even after the neutron irradiation. Fig. 5 shows a summary of the a-type and c-type dislocation density determined at the different locations of the irradiated Zr-2.5Nb tube as well as the β-Nb particles. The neutron irradiation increased the a-type dislocation density of the irradiated tube as much as almost 2 times that of the unirradiated tube, which is the largest at the inlet location subjected to the lowest temperature. This demonstrates that the dominant factor affecting the dislocation density in the irradiated Zr-2.5Nb tubes is temperature not neutron fluence or flux. If the neutron fluence is a dominant factor, the middle tube ring exposed to the highest neutron fluence should have had the highest dislocation density. In reality, the middle tube ring had a lower dislocation density than the inlet tube ring. A thing to note is that the c-type dislocation density shows no change before and after the neutron irradiation and further with the location. Thus, it is reasonable to say that the neutron fluence of 8.9×10^{25} n/m^2 is not enough to produce any change in the c-type dislocation density in the Wolsong1 tubes. As far as the fine β-Nb particles are concerned, they slightly grew at the outlet tube ring due to higher operating temperature. Based on these facts, we conclude that any change in the mechanical properties, or tensile strength and delayed hydride cracking velocity and so on, is attributed to the a-type dislocation density, not the c-type dislocation density.

To correlated the microstructural evolution with the neutron irradiation with the mechanical properties, we are conducting tensile tests and delayed hydride cracking(DHC) tests on the three tube rings taken from the inlet, middle and outlet parts of the tube. Fig. 6 shows the transverse and longitudinal tensile strengths of the irradiated Wolsong1 Zr-2.5Nb tube with the location. For both transverse and longitudinal directions, the highest strength was observed at the inlet part of the tube. These results are quite consistent with the distribution of the a-type dislocation density with the location in the irradiated Wolsong1 tube as shown in Fig. 4. To reduce any scatter in the tensile strengths due to the different circumferential position even at the same location, we compared tensile strengths with the location using the specimens taken from the same circumferential positions, or the bottom and the middle position as shown in Fig. 7. The difference in the tensile strength between the inlet tube ring and the outlet one becomes more noticeable when the location as well as the circumferential position around the same location of the tube is considered.

Since the Zr-2.5Nb tube experiences not only an increase in the dislocation density but also the decomposition of the β-Zr phase with the neutron irradiation, we also investigated the niobium contents in the β-Zr grains using two different methods. One is selected area diffraction patterns obtained using thin foils where the lattice constant of $d_{(110)}$ were determined using the following equation, $d(110) = d(0001)R/r$, where $d(110)$ and $d(0001)$ are the lattice spacing of (110) and (0001) planes, R and r are the radius of the first ring of the diffracted beam as shown in Fig. 8 illustrating a dark field image of the β-Zr and selected area diffraction patterns. The other is a carbon replica method where the beta-Zr particles are extracted and analyzed by electron diffraction spectroscopy [2]. Fig. 9 shows the Nb concentration in the β-Zr phase with the distance from the inlet. Here, we also included the data reported by AECL [3] which were determined macroscopically using an X-ray method from the tubes irradiated for 2 to 12 years in Canadian nuclear power plants. The decomposition of the β-Zr with the location determined by the selected area diffraction

patterns (SADPs) seems to suggest that the decomposition of the β-Zr phase in the Wolsong1 tube has a similar pattern as those in the Canadian Zr-2.5Nb tubes. However, when the carbon replica method was used, the decomposition of the β-Zr with the location had a different pattern from that determined by the SADPs. Furthermore, the Wolsing1 tube was found to have the faster decomposition of the β-Zr at the outlet location compared to the Canadian tubes, or Pickering and Bruce tubes. Comparing the coolant temperatures between the Wolsong 1 and Pickering or Bruce NPPs, these results look quite consistent. One thing to note is that the Nb concentration of the β-Zr phase at the inlet location becomes lower after the neutron irradiation. Considering that the carbon replica method can yield more accurate analysis of the Nb content in the β-Zr grains, we come to the conclusion that the neutron irradiation decreases the Nb concentration of the β-Zr at the inlet or the middle location of the tube below that before irradiation but at the outlet location, the Nb concentration of the β-Zr increases due to the faster thermal decomposition. Thus, neutron irradiation facilitates diffusion of Nb away from the β-Zr grains while thermal effects let Nb diffuse back to the β-Zr grains. Hence, there seems to be a competition of the thermal effect and the neutron irradiation effect as to the decomposition of the β-Zr phase that is consistent with a mechanism for the stability of precipitates under neutron irradiation proposed by Nelson [4]. These results demonstrate that there is continuous movement of Nb between the α-Zr and β-Zr grains during operation in reactor, which is governed by the combined effect of temperature and neutron fluence.

To understand the effect of the phase decomposition on the pick-up of hydrogen and corrosion of the Zr-2.5Nb tube, three different Zr-2.5Nb alloys were subjected to corrosion tests at 400 ^{o}C: the 1st is the beta-quenched Zr-2.5Nb, the 2nd is the aged Zr-2.5Nb obtained by the beta-quenching and aging at the 500 ^{o}C for 10 days and the 3rd is the annealed Zr-2.5Nb obtained by slow cooling from 850 ^{o}C. The aged Zr-2.5Nb has a microstructure of α-Zr and β-Nb particles while the annealed one consists of the α-Zr and β-Zr phases. The annealed one had higher weight gains but lower hydrogen pick-up fraction while the aged one had lower weight gains but much higher hydrogen pick-up fraction as shown in Fig. 10. Based on these results, since neutron irradiation caused the β-Zr to decompose to the β-Nb, the outlet location of the Zr-2.5Nb tube under neutron irradiation would show higher hydrogen pick-up rate than anticipated, which seems to be accelerated with time. This may explain non-linear rate of the hydrogen pick-up of the Zr-2.5Nb tubes.

4. Conclusion

The irradiated Zr-2.5Nb tube in the Wolsong1 nuclear power plant for 9.3 EFPYs had a microstructural evoluation: an change in the dislocation density and the Nb content in the β-Zr grains. At the neutron fluence of 8.9×10^{25} n/m^2, the larger increase in the a-type dislocation density occurred at the inlet location of the tube subjected to the lowest operating temperatures while the c-type dislocation density did not change with the neutron irradiation. The highest increase in the a-type dislocation density at the inlet location leads the Wolsong1 Zr-2.5Nb tube to have the higher tensile strength and higher DHC velocity at the inlet

location. The outlet location of the Wolsong1 Zr-2.5Nb with the highest operating temperature had the higher decomposition rate of the β-Zr, leading to the formation of the β-Nb, while the inlet location had a lesser amount of the Nb concentration in the β-grains compared to that of the unirradiated Zr-2.5Nb tube due to the neutron irradiation. Based on these facts, we conclude that Nb continuously moves between the α-Zr grains and the β-grains in the Zr-2.5Nb tube under combined effects of temperature and neutron fluence during their operation in heavy pressurized water-cooled reactors. Furthermore, the higher decomposition of the β-Zr grains with neutron irradiation may contribute to enhanced hydrogen pick-up rate of the Zr-2.5Nb tubes with their operation period approaching the design lifetime.

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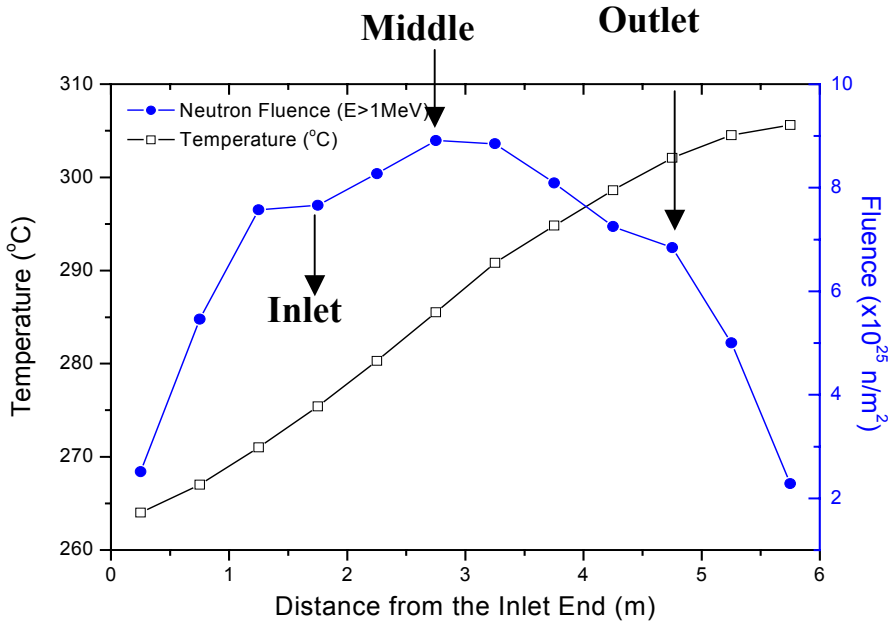

Fig. 1. Temperature and neutron fluence of the three tube rings taken from the M-11 tube.

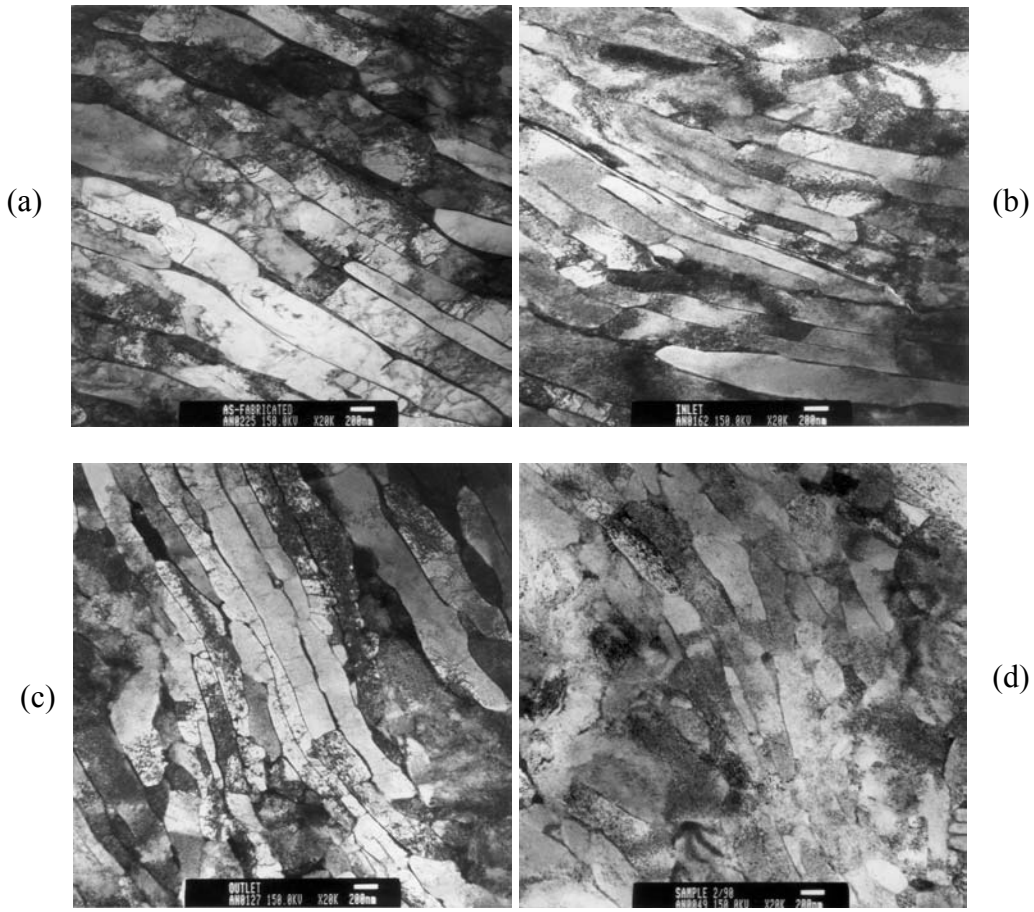

Fig. 2. Microstructures of (a) unirradiated Zr-2.5Nb tube and the (b) inlet, (c) middle and (d) outlet tube rings of the irradiated Zr-2.5Nb tube to the fluence of 8.9×10^{25} n/m^2.

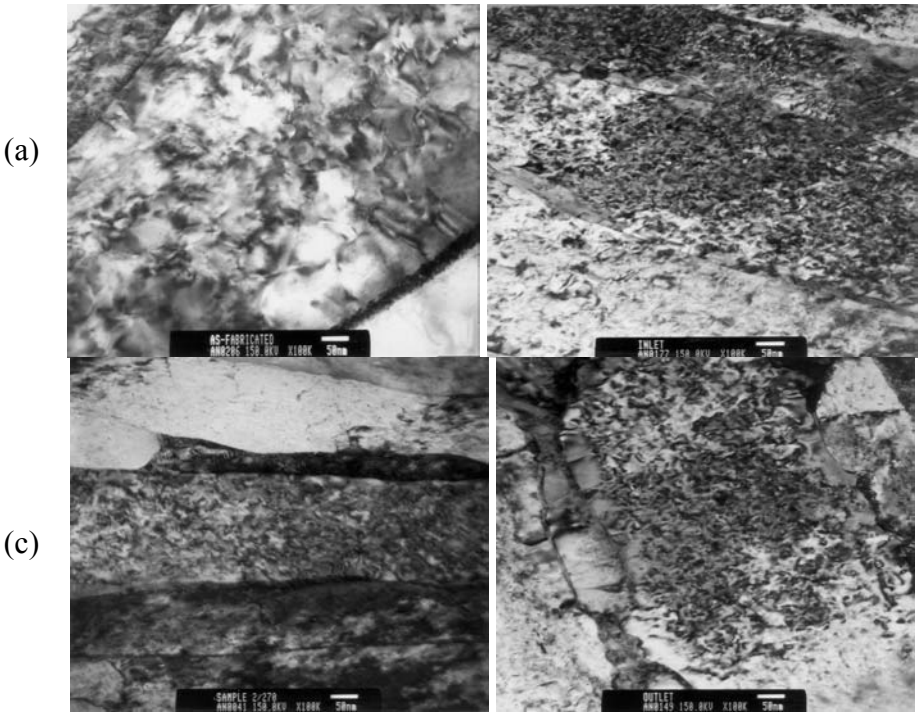

Fig. 3. Enlarged microstructures of α-Zr grains in (a) the Zr-2.5Nb tube before irradiation and (b) the inlet location, (c) the middle location, (d) the outlet location of the irradiated Zr-2.5Nb tube.

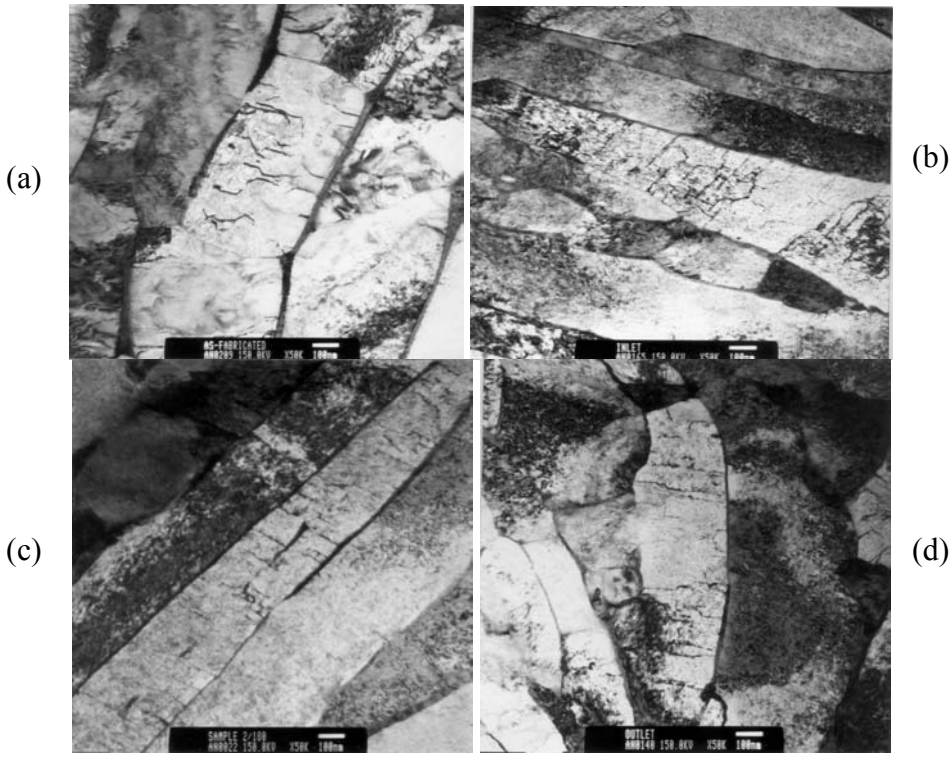

Fig.4. c-type dislocations observed in (a) the Zr-2.5Nb tube before irradiation and (b) the inlet, (c) the middle, (d) the outlet location of the irradiated Zr-2.5Nb tube.

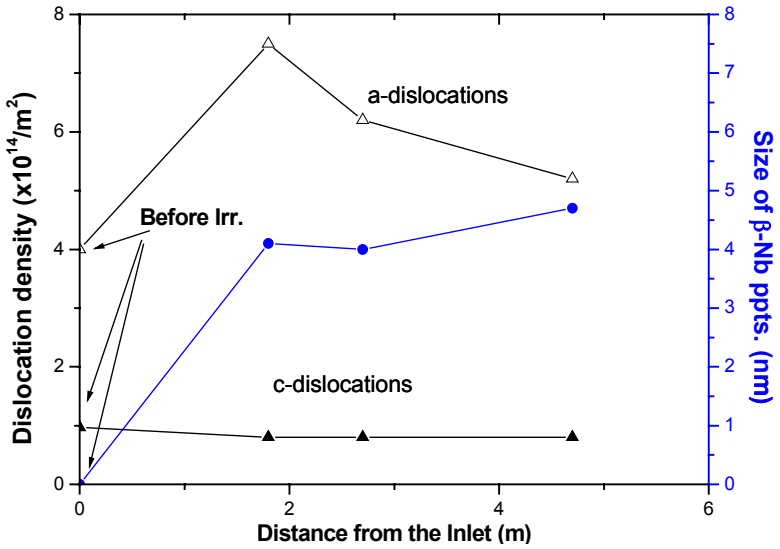

Fig. 5. Axial distribution of a-type and c-type dislocations and β-Nb particles in the irradiated W1 tube.

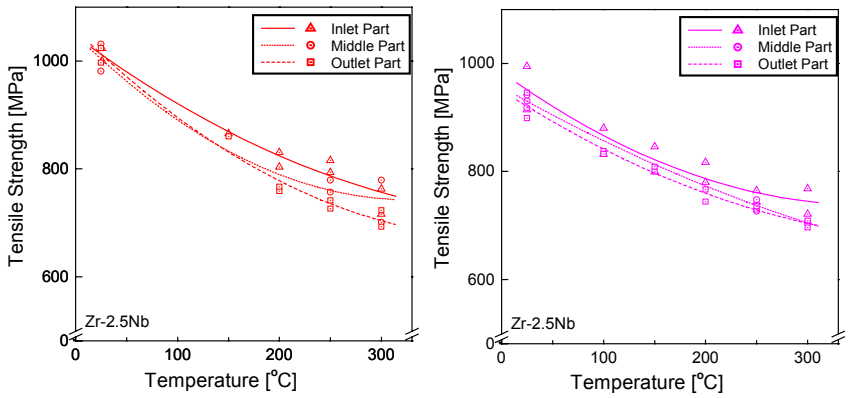

Fig. 6. (a) Transverse and (b) longitudinal tensile strengths of the irradiated W1 Zr-2.5Nb tube with the axial location from the inlet.

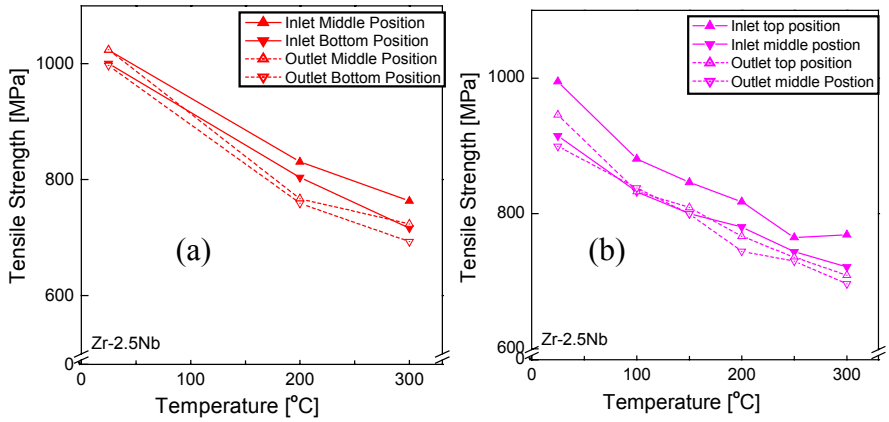

Fig. 7. (a) Transverse and (b) longitudinal tensile strengths of the irradiated W1 Zr-2.5Nb between the inlet and the outlet location at the same circumferential positions.

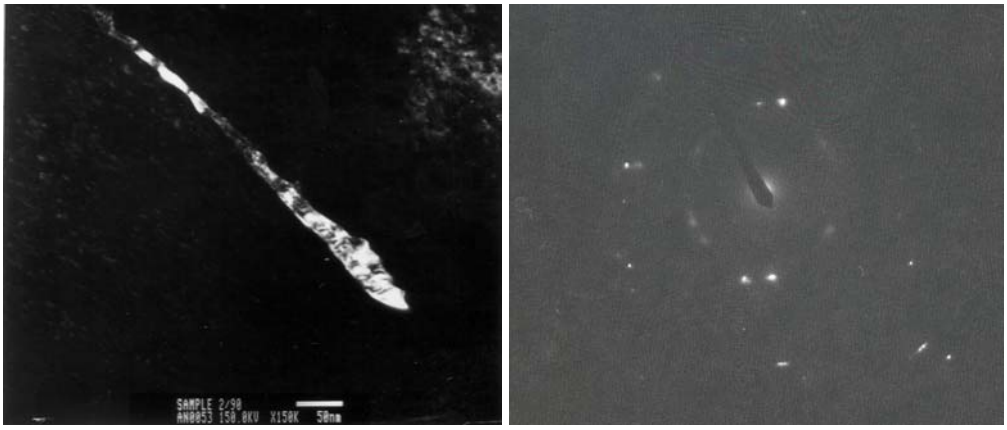

Fig. 8. Dark field image of the β-Zr grains in the irradiated W1 Zr-2.5Nb tube and their selected area diffraction patterns.

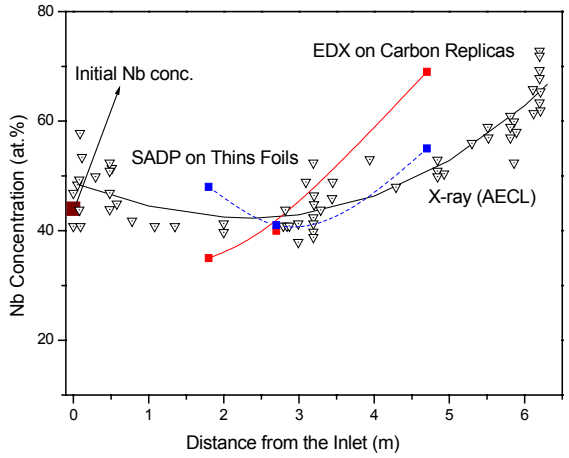

Fig. 9. Decomposition of the β-Zr with the distance from the inlet location. in the irradiated W1 Zr-2.5Nb tube along with that in the Canadian tubes.

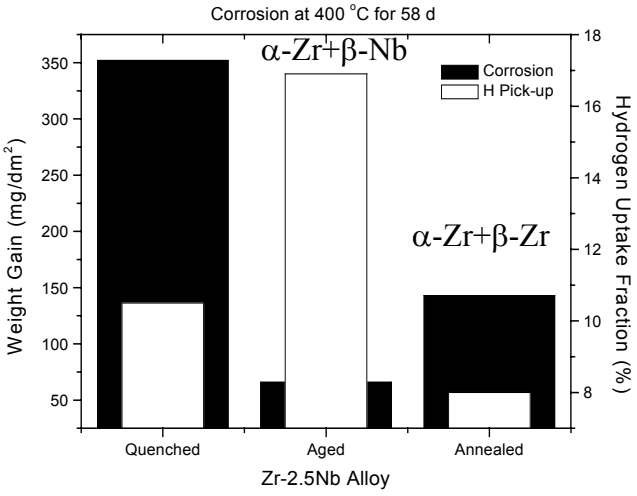

Fig. 10. Weight gains and hydrogen pick-up rate of the Zr-2.5Nb alloy with different microstrucutres