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Variation of Mechanical Property due to Short Range Ordering by Thermal Aging up to 20,000 Hours in Zr-2.5%Nb CANDU Pressure Tube

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1. Introduction

The pressure tube made of the Zr-2.5%Nb alloy in CANDU(CANada Deuterium Uranium) reactor consists of the primary pressure boundary with the feeder pipe and steam generator [1]. The pressure tube contains nuclear fuel causing nuclear fission, and heavy water (D_2O) being a coolant and a moderator flow between the pressure tube and the fuel.

Although Zircaloy-2 alloy was initially used for the pressure tube of the heavy reactor, there was a rupture failure during operation due to high ingress rate of hydrogen generated during CANDU reactor operation [2]. On the other hand, the Zr-2.5%Nb alloy has been used as a pressure tube material with relatively high yield strength and low hydrogen absorption.

The change in material that appears when the material is maintained at the reactor operating temperature is generally referred to as an aging phenomenon. Until now, it was common to study the aging phenomena to confirm the change in strength or toughness according to the aging treatment. However, the material has increased entropy in the manufacturing process and a decrease in entropy during service operation in actual structures. In alloys this process is due to the attraction force between specific atoms. Then, the arrangement of atoms has been changed. Certain atoms are located around another specific atom. If its size is as small as 2 nm, it is called a short range order (SRO) because it does not form a super lattice [3, 4].

Aging in a reactor operating environment inevitably involves an SRO phenomenon. That is, the SRO phenomenon is at least one of the main causes of the aging effect. However, to date, the nature of the aging phenomenon is not understood. When the SRO phenomenon occurs during the aging process, the distance between atoms gets closer, the lattice distance and the length of the structure is reduced. Surprisingly, the SRO phenomenon generates additional stress itself within the structure [5, 6, 7].

Therefore, this study aged a quadruple melt pressure tube material from 300-420°C up to 20,000H. The effect of aging on tensile behavior was examined and analyzed. In addition, the effects of strain rate and the effects of short range ordering (SRO) are discussed in detail.

2. Experimental

The CANDU pressure tube material used for this experiment was quadruple-melted (four time melt), the chemical composition of this pressure tube is shown in Table 1.

To simulate the Aging effect, the material was aged in air at 300, 350, and 400°C up to 20,000 hours, respectively. In order to compare the effect of aging, the as-received specimens were tested.

The gage length of tensile specimen in transverse direction is 16mm due to the limitation of pressure tube with 103mm inner diameter. Strain rate for tensile test is applied to be $1x10^{-6}$/s. Tensile test was carried out at 267±2°C, which is the inlet temperature of the pressure tube in CANDU reactor.

Table 1. Chemical composition of quadruple melt Zr-2.5%Nb (wt %).

elements	Zr	Nb	Fe	Ta	Cr	Ti	W	O	H
composition	Balance	2.6%	980 ppm	100 ppm	<100 ppm	<50 ppm	<50 ppm	1100 ppm	<3 ppm

3. Results and Discussions

Fig. 1 shows the change in mechanical properties in transverse direction at 267°C aged at 350°C for up to 20,000 hours. The yield strength (YS) and tensile strength (UTS) decreased by about 11%, even though there is a small fluctuation with aging time. The total elongation (TE) and the uniform elongation (UE) changed little.

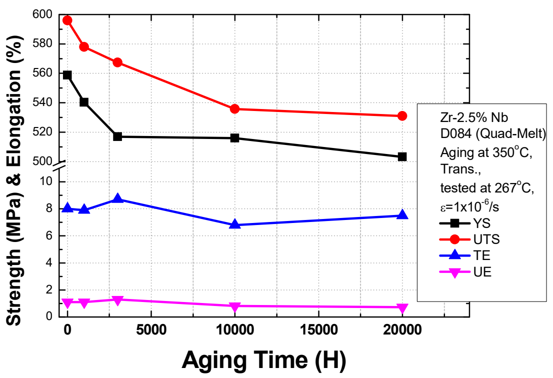


Fig. 1. Variation of mechanical properties in transverse direction at 310°C in Zr-2.5%Nb pressure tube material with aging at 350°C up to 20,000 hours.

Fig. 2 shows the changes in the mechanical properties at 267°C in transverse direction aged at 400°C. The YS and UTS decreased by about 13% and by 14%, respectively after 20,000H aging. The TE changed little and the UE stayed at about 1% from the beginning stage.

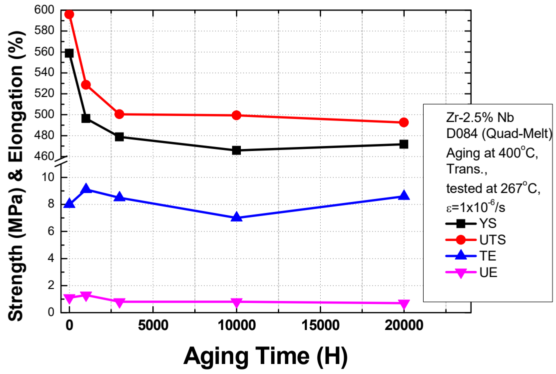


Fig. 2. Variation of mechanical properties in transverse direction at 267°C in Zr-2.5%Nb pressure tube material with aging at 400°C up to 20,000 hours.

Fig. 3 shows the comparison of the mechanical properties at 267°C in the transverse direction with the temperature aged at 300-400°C for 20,000 hours. The YS and UTS decreased fast at above 350°C. The TE increases with temperature and the UE decreases from about 2% in as received to 1% aged at 400°C for 20,000 hour.

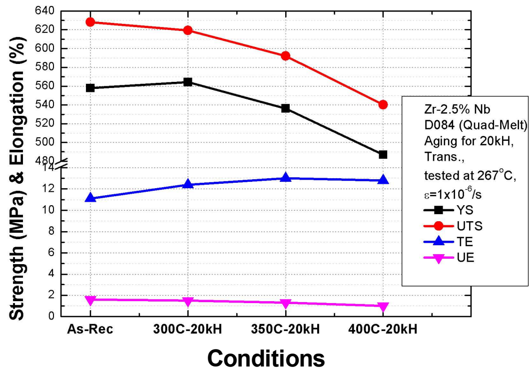


Fig. 3. Variation of mechanical properties in transverse direction at 267°C in Zr-2.5%Nb pressure tube material with aging at 400°C up to 20,000 hours.

Fig. 4 shows the comparison of the change in yield strength at 267℃ with aging treatment time at 300-420℃. The yield strength (YS) increases by 1-2% in specimens aged at 300°C. However, the YS decreases fast with aging temperature at above 350°C and the decreasing rate becomes slower with aging time up to 20,000 hours.

Fig. 5 shows the comparison of the change in tensile strength (UTS) at 267℃ with aging treatment time at 300-420℃. The aging treatment at 300℃ showed little change in UTS and reduction rate within 2%. However, the aging at 350℃ and 400℃ decreased by 6 and 13%, respectively.

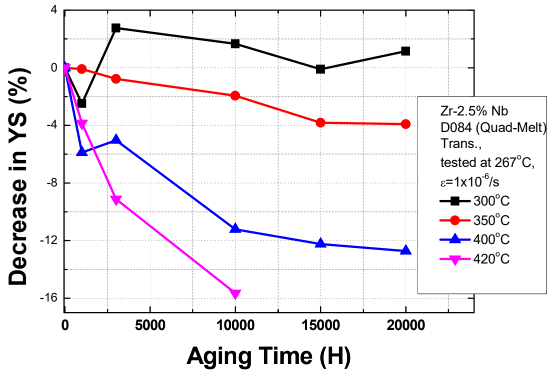


Fig. 4. Variation of yield strength at 267℃ in Zr-2.5%Nb pressure tube material with aging conditions up to 20,000 hours.

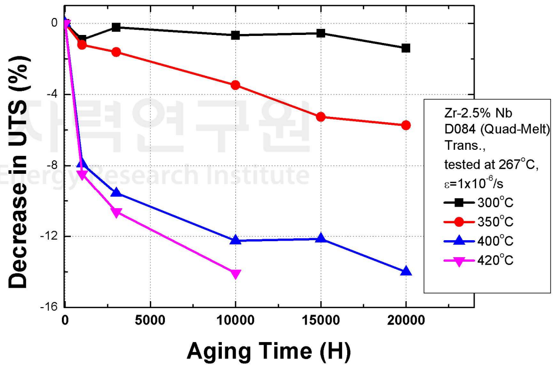
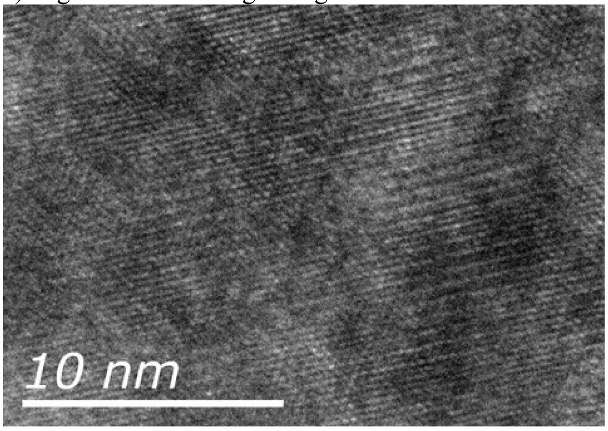


Fig. 5. Variation of tensile strength at 267℃ in Zr-2.5%Nb pressure tube material with aging conditions up to 20,000 hours.

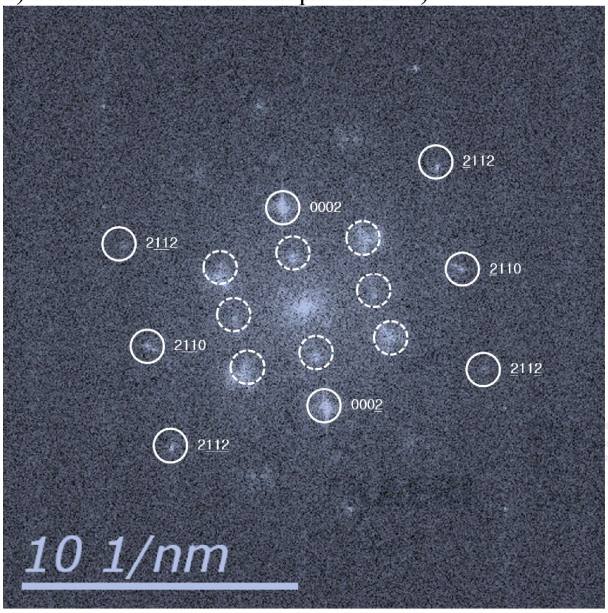
The reason why the mechanical properties vary by thermal aging is due to a short range ordering (SRO) reactionin Zr-2.5%Nb pressure tube. This can be confirmed by various method, such as DSC (differential scanning calorimeter), neutron diffraction, and TEM. The evidence for SRO in Zr-2.5%Nb pressure tube is shown in Fig. 6. When Fig. 6 b) and c) is compared, some extra spots are marked as a broken lined circles. This forbidden spots cannot be seen without SRO reaction.

The effects of SRO in Zr-2.5%Nb material is well explained recently [3]. Generally, the decrease in mechanical properties with thermal aging has been understood as annealing effect. However, the softening of material by thermal aging is mainly occurred by atomic rearrangements. This is due to reduction in entropy by thermal aging, that is, SRO reaction.

a) High resolution image of aged Zr-2.5%Nb



b) Selected area diffraction pattern of a)



c) Relationship of zone axis in HCP

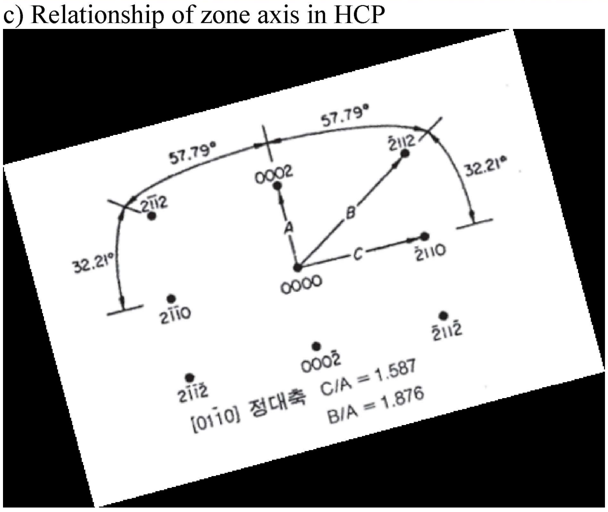


Fig. 6. TEM analysis results showing short range ordering (SRO) in aged at 400°C for 3,000H, a) high resolution image, 2) SADP showing forbidden spots, and c) relationship of diffraction spot in [0110] zone.

This study only analyzed the effect of thermal aging on Zr-2.5% Nb alloy pressure tube. However, since the reactor pressure tube is exposed in a fast neutron irradiation environment, the irradiation effect by the fast neutron affects the rate of SRO reaction. Therefore, the aging effect in this study is not appropriate to be used directly for the evaluation of the properties of operating CANDU pressure tube, but it is meaningful to understand in terms of studying the aging effect of pressure tube materials due to SRO phenomenon and the nature of the aging phenomenon.

4. Conclusions

1. The aged quadruple-melt pressure tube materials show a decrease in YS by 70 MPa at 267°C after aging at 350°C for 20,000 hours at strain rate of $1x10^{-6}$/s.
2. The aged quadruple-melt pressure tube materials show a decrease in UTS by 110 MPa at 267°C after aging at 400°C for 20,000 hours at strain rate of $1x10^{-6}$/s.
3. The YS and UTS in transverse direction at 267°C decreased by 0-13% and 2-14%, respectively, after aging at 300-400°C for 20,000 hours.
4. The variation of mechanical properties by thermal aging comes from the short range ordering (SRO) reaction accompanying reduction in entropy.

Acknowledgments

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