Dimensional Change by Aging Treatment at 300-400℃ up to 20kH in Zr-2.5%Nb CANDU Pressure Tube Material

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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 [2].

The pressure tube is exposed to fast neutron (E>1MeV) emitted by nuclear fission in the operating environment. For this reason, the length and lateral diameter of the pressure tube expand as the reactor operates. This phenomenon is called irradiation creep and growth. Changes in dimension of pressure tubes should be monitored periodically to ensure that they do not deviate from the allowable range.

Changes in the pressure tube in the operating environment are confirmed by periodic monitoring, but the cause of this is not understood yet clear. This is because even in Zr-2.5%Nb alloys, irradiation-induced growth and creep behavior are reported differently depending on the temperature of the neutron irradiation environment, neutron flux, and material processing conditions [3].

Since the pressure tube in CANDU reactor is known to undergo length changes in the operating environment, longitudinal elongation or diametral (transverse) creep of the pressure tube is reflected in the design. In particular, lengthening in the longitudinal direction is permitted up to about 5 mm per year. Transverse deformation is a very important factor in determining operating conditions because there is a risk that the coolant may reduce the cooling of the nuclear fuel.

Therefore, in this study, the dimensional change of Zr-2.5%Nb alloy was investigated in an environment without neutron irradiation. In order to obtain the accelerated effect of the aging treatment, aging was performed at 300-400°C, which is slightly higher than the operating temperature, for up to 20,000 hours, and the lattice change was observed using the neutron diffraction method to understand the behavior of dimensional change purely by thermal treatment

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.

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

This pressure tube material was thermally treated in air at 300, 350, and 400°C for 3,000, 10,000, and 20,000 hours, respectively. In order to confirm whether the dimensional change appears during the thermal treatment, the lattice change of this specimen was observed using a high-resolution neutron diffraction (HRPD) analyzer installed at Hanaro in KAERI (Korea Atomic Energy Research Institute).

Table 2. Summary of thermal treatment for D084 (Quadruple melt Zr-2.5%Nb).

Time(H) / Temp. (°C)	3,000	10,000	20,000
300	O	O	O
350	O	O	O
400	O	O	O

The lattice change was measured as compared to the lattice of the as-received specimen without thermal treatment. The relationship of ($d_{\text{as received}}$-$d_{\text{thermal treated}}$)/$d_{\text{as received}}$ was used. Since neutron diffraction measures the average lattice of the material used for diffraction, it is very meaningful to be able to observe the anisotropic lattice change for each (kkl).

3. Results

Figure 1 shows the microstructure observed by TEM in the as received Zr-2.5%Nb pressure tube material. . The elongated crystal is α-Zr, and the black portion between the grains is the β-Zr phase. The pressure tube has a two-phase structure of α-Zr and β-Zr because the manufacturing process is high-temperature extrusion at about 830°C, cold drawing of about 20-25% at room temperature, and stress relief treatment at 400°C steam for 24 hours

Figure 2 shows a comparison of the (10$\underline{1}$0) lattice change of a specimen thermally treated at 300-400°C for 20,000 hours. At 300 and 350 °C, a contraction occurs the (10$\underline{1}$0) lattice. However, at 400°C, the expansion of about 0.02% occurs at 10,000 hours, but after that, it becomes saturated.

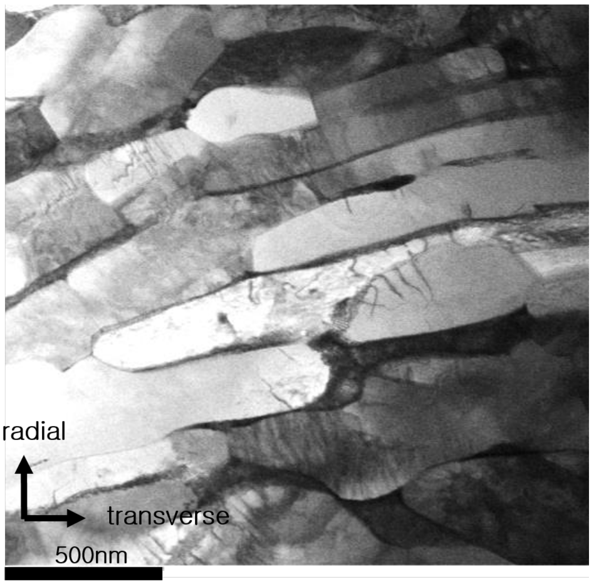

Fig. 1. TEM microstructure of as received Zr-2.5%Nb pressure tube material.

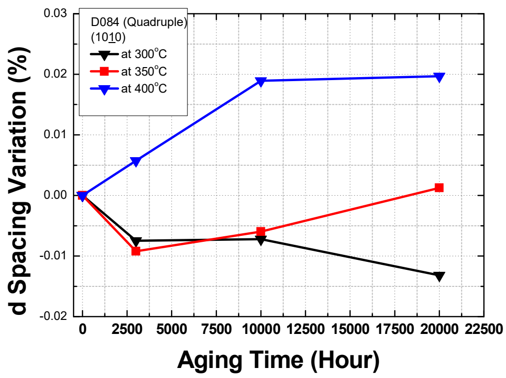

Fig. 2. (10$\underline{1}$0) lattice variation measure by neutron diffraction in D084 (Quadruple melt) pressure tube by aging at 300-400°C up to 20,000 hours.

Figure 3 shows a comparison of the (0002) lattice changes of the specimens thermally treated at 300-400°C for 20,000 hours. The change of the (0002) plane hardly appears at 300 and 350°C. However, at 400°C, a lattice change of about 0.08% appears at 3000 hours. This is about 4 times greater than the (10$\underline{1}$0) lattice change even though it is shorter time. After that, as time increases, the lattice expansion saturates without further increasing.

Figure 4 compares the lattice changes of the {1010} and {0002} families together. There is little difference between the grid changes of (10$\underline{1}$0) and (20$\underline{2}$0), while the lattice changes of (2020) and (0004) show a large difference by nearly 3 times. The lattice change of the (0004) plane is about 1/3 that of (0002). This is a result of anisotropy of crystals.

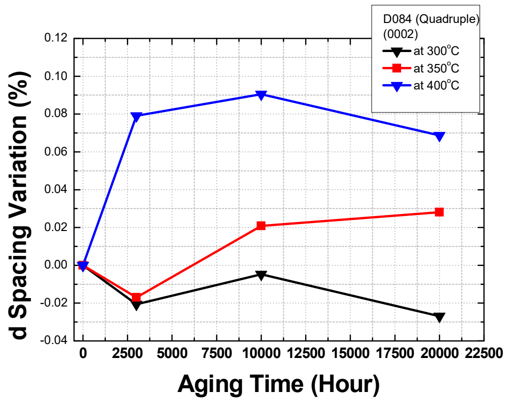

Fig. 3. (0002) lattice variation measure by neutron diffraction in D084 (Quadruple melt) pressure tube by aging at 300-400°C up to 20,000 hours.

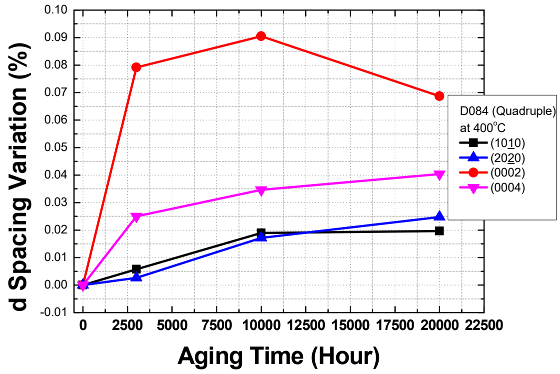

Fig. 4. {10$\underline{1}$0} and {0002} lattice variation measure by neutron diffraction in D084 (Quadruple melt) pressure tube by aging at 400°C up to 20,000 hours.

4. Discussions

When a material is exposed to a temperature range where atoms can diffuse, atoms are aligned through diffusion and entropy decreases [4]. However, the effect of fast neutron (E>1MeV) irradiation in the nuclear reactor environment shifts the position of atoms and increases entropy by increasing lattice defects. The lattice expands when entropy increases as the atomic arrangement is disturbed for whatever reason.

However, the thermal treatment for Zr-2.5%Nb pressure tube shows a change within 0.01% at 300 and 350°C even if the thermal treatment time is continued up to 20kH. On the other hand, at 400°C, the (1010) plane expands by 0.02% and the (0002) plane expands by about 0.08% without neutron irradiation, and then it becomes saturated.

First, the reason that lattice expansion hardly occurs below 350°C and lattice expansion is evident only at 400°C is because the diffusion rate of atoms is slow at 300°C and 350°C, or the critical activation energy required for atoms to diffuse to cause lattice expansion is 400°C. It seems to be because it is supplied at a temperature above 400°C.

The reason that the lattice expansion occurs at 400°C and becomes saturated at 3 kHz or higher is because the internal driving force that causes the lattice expansion is exhausted. When exposed to 400°C without neutron irradiation, it does not expand any more after the driving force that causes lattice expansion is exhausted. It is judged that the driving force causing lattice expansion at 400°C is supplied by the disappearance of Nb supersaturated in α-Zr of the Zr-2.5%Nb material and the reduction of the remaining entropy during the manufacturing process of the pressure tube.

However, in the environment of CANDU reactor pressure tube, the atomic arrangement of the pressure tube material is continuously disturbed by fast neutron (E>1MeV) irradiation, and entropy increases. At the same time, the pressure tube material has a thermal annealing effect that reduces entropy in the operating environment around 300℃. That is, the reactor operating environment is a state in which (a) entropy increase due to neutron irradiation and (b) entropy decrease due to thermal effect compete. Both these effects cause lattice expansion. It is very important for understanding transformation.

In this way, entropy increase by fast neutron (E>1MeV) irradiation itself causes lattice expansion, and the entropy formed by irradiation causes lattice expansion due to the thermal treatment effect of the driving force of atomic diffusion. Therefore, the lattice expansion of the Zr-2.5%Nb pressure tube material exposed to the fast neutron irradiation environment is not saturated and expands continuously. That is, entropy increase due to fast neutron (E>1MeV) irradiation continues, because this is the driving force of the lattice change.

As a result of the thermal treatment experiment applied in this study, the lattice change was insignificant at 300 and 350 °C. However, the lattice change occurred by 0.02-0.08% depending on the direction at 400 °C. As confirmed in this study, it is reviewed whether the effect of thermal treatment shown in the 400℃ out-of-reactor experiment occurs in the operating environment of the CANDU pressure tube at abround 300℃. As discussed above, the driving force of lattice expansion by fast neutron (E>1MeV) irradiation or lattice expansion by thermal treatment is entropy. Therefore, the higher the entropy, the greater the driving force. Therefore, if entropy is increased by neutron irradiation, it is natural that the temperature required for atomic diffusion is lowered. That is, if the temperature required for atomic diffusion in a low entropy state as in the out-of-reactor experiment is about 400°C, the high-entropy fast neutron (E>1MeV) irradiation environment sufficiently enables atomic diffusion at around 300°C. Therefore, although the operating temperature of the CANDU reactor is around 300°C, the lattice expansion observed in the 400°C out-of-reactor experiment can sufficiently occur.

The effect of generating lattice defects or increasing entropy by fast neutron (E>1MeV) irradiation is similar to that of cold working. The ordering reaction that occurs with entropy reduction in cold-worked materials appears at lower temperatures and has been confirmed in several materials. It has been reported that cold working in Alloy 600, a Ni-based alloy, lowers the temperature of the ordering reaction by about 300°C [5]. This is because entropy supplied by cold working acts as a driving force for atomic diffusion.

The results of this study can explain the reason why the longitudinal elongation or transverse creep strain in the actual CANDU reactor environment is much larger than that measured outside the furnace. The neutron irradiation environment continuously increases entropy as the arrangement of atoms is disturbed, so it provides an acceleration effect similar to that of a pressure tube exposed to a higher temperature. That is, the operating temperature of the CANDU reactor is around 300°C, but the fast neutron (E>1MeV) irradiation activates the diffusion of atoms in the pressure tube, so the pressure tube material is operated at 400°C or higher. This helps to explain the phenomenon that the elongation and creep behavior of the CANDU reactor pressure tube material is much larger than the values predicted by the out-of-reactor experiments.

4. Conclusions

1. Thermal treatment of quadruple melt Zr-2.5%Nb pressure tube material causes lattice expansion in any direction above 400℃.
2. The lattice change of the (10$\underline{1}$0) plane expands and saturates by 0.02% in about 10,000 hours. On the other hand, the lattice change of the (0002) plane expands and saturates by 0.08% at 3,000 hours.
3. Since the (0002) plane is mainly oriented in the transverse direction of the pressure tube, the reason for the high transverse creep of the CANDU pressure pipe is due to the lattice expansion of the (0002) plane.
4. Since the fast neutron irradiation environment of the CANDU reactor disturbs the atomic arrangement and causes a continuous increase in entropy, it is judged that the diffusion behavior of the Zr-2.5%Nb pressure tube will have an accelerating effect like operating at 400℃ or higher.

Acknowledgments

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