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

SungSoo Kim*, Jong Yeop Jung*, Hyung Sub Kim, and Young Suk Kim**

**Korea Atomic Energy Research Institute,*
** MACTEC(Materials Core Technology Center), 402-1, Nuclear Tech-Biz Center
111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, Korea
**Corresponding author: sskim6@kaeri.re.kr*

1. Introduction

The pressure tube of the heavy water reactor is made of Zr-2.5%Nb alloy [1, 2]. The pressure tube is the primary pressure boundary component of the heavy water reactor that surrounds the fuel and coolant [3]. During reactor operation, the pressure tube exposed to fast neutron irradiation causes dimensional changes, increasing the length of the pressure tube and expanding its diameter [1, 2]. This change in the dimensions of the pressure tube is called irradiation growth and irradiation creep. Growth and creep occur simultaneously in the irradiated environment, making it difficult to separate them. Moreover, the irradiation growth behavior of the Zr-2.5%Nb pressure tube is reported differently depending on the temperature of the neutron irradiation environment, on the neutron flux, and on the processing conditions of the material, and the cause of this phenomenon is still not clearly understood [1].

Since the pressure tube undergoes dimensional change in the operating environment, the longitudinal elongation or diametric creep of the pressure tube is reflected in the reactor design [1, 2]. Nevertheless, the dimensional change of the pressure tube should be monitored periodically to ensure that it does not deviate from the allowable range [5]. The longitudinal extension of the pressure tube is allowed up to about 5 mm per year. It is accommodated by being fixed on one side of the pressure tube and pushed in the opposite direction. Therefore, elongation is not a problem as long as the design tolerances are not exceeded.

On the other hand, the diametric deformation in which the diameter of the pressure tube expands may increase the area through which the coolant flows and may not completely cool the nuclear fuel enough [1, 4]. Therefore, the diametric creep of the pressure tube is a very important factor in determining the operating conditions of a nuclear reactor [1]. According to the operating experience so far, the diameter expansion of the Zr-2.5%Nb alloy pressure tube is much larger than the design estimate. Since this diameter expansion causes a reduction in the power generation capacity of heavy water reactors, it appears as an economic loss in reality [1-3].

Although pressure tubes are a major cause of dimensional change in operating environments with fast neutron irradiation, it is important to understand how dimensional changes appear when aged in the absence of neutron irradiation. Therefore, this study investigated the dimensional change of Zr-2.5%Nb alloy according to aging treatment in an environment without neutron irradiation. In order to obtain the accelerated effect of the aging treatment, aging treatment was performed for up to 20,000 hours at 300°C similar to the operating temperature and at 350 and 400°C slightly higher than the operating temperature, and only thermal treatment was performed by observing the lattice change using the neutron diffraction method. We tried to confirm the lattice change behavior by as an accelerated experiment at 400°C, we discussed the relationship between the experimental results and the actual dimensional change of the heavy water reactor pressure tube.

2. Experimental

Zr-2.5%Nb CANDU pressure tube material is divided into 4 melting and 2 melting materials depending on how many times it is vacuum melted during the ingot manufacturing process. The materials used in this study are a double melt pressure tube (E34) and a four melt pressure tube (D084). The chemical composition of this material is shown in Table 1.

Table 1. Chemical composition of D084 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 measured using a high-resolution neutron diffraction (HRPD) analyzer installed at Hanaro in KAERI (Korea Atomic Energy Research Institute).

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 the 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 (kkil).

3. Results

Figure 1 shows the (10$\bar{1}$0) lattice change according to temperature in the E34 pressure tube aged at 300-400°C for 20,000 hours. When aging at 300°C-10,000 hours, (10$\bar{1}$0) lattice expands to about 0.02% and contracts slightly to about 0.013% after 20,000 hours. When aging at 350°C, it expands rapidly up to about 3,000 hours and expands by 0.03% until 20,000 hours. On the other hand, the 400°C aging treatment expands to about 0.045% at 3,000 hours and remains almost constant.

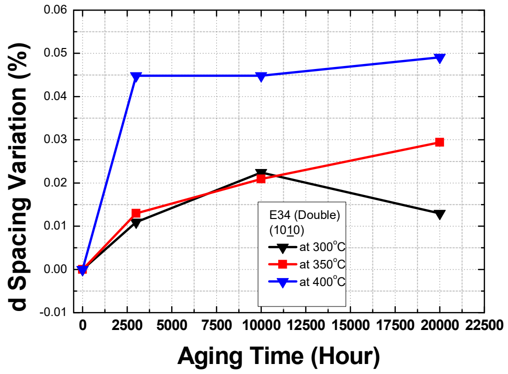

Fig. 1. (10$\bar{1}$0) lattice variation measure by neutron diffraction in D084 (Quadruple melt) pressure tube by aging at 300-400°C up to 20,000 hours.

Figure 2 shows the (0002) lattice change according to temperature in the E34 pressure tube aged at 300-400°C for 20,000 hours. Aging at 300° C appears to fluctuate in the range of about 0.02%. Aging at 350°C expands about 0.01% up to about 10,000 hours, and expands about 0.04% after 20,000 hours.

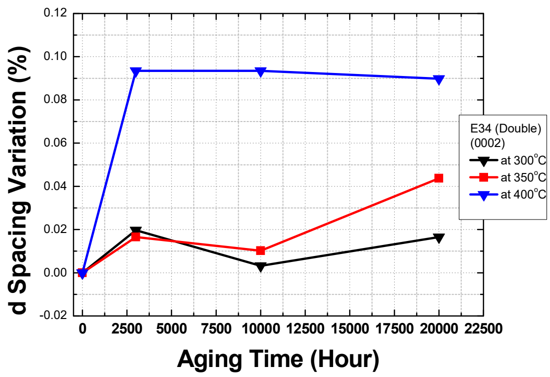

Fig. 2. (0002) lattice variation measure by neutron diffraction in D084 (Quadruple melt) pressure tube by aging at 300-400°C up to 20,000 hours.

Figures 1 and 2 show that the aging treatment at 350°C or lower and the sample treatment behavior at 400°C are completely different. Aging at 400 °C rapidly expands the lattice and remains constant. It seems that the lattice expansion by aging at 400 °C is attributed to an increase in entropy.

Figure 3 shows the lattice change of {10$\bar{1}$0} and {0002} planes according to the aging treatment time at 400°C. The lattice expansion of the (2020) plane is about half that of the (10$\bar{1}$0) plane. The lattice expansion of a (0004) plane is about 1/4-1/3 that of (0002) plane. This result clearly shows that the lattice change of crystal is anisotropic.

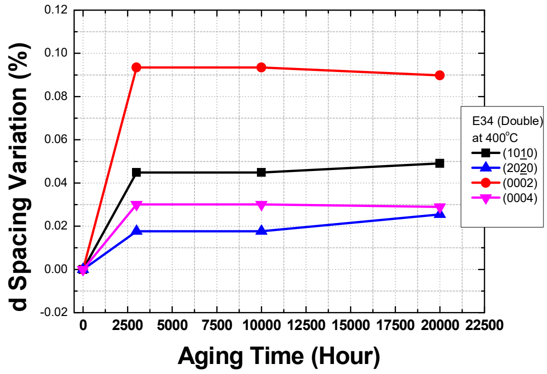

Fig. 3. {10$\bar{1}$0} and {0002} lattice variation measure by neutron diffraction in D084 (Quadruple melt) pressure tube by aging at 400°C up to 20,000 hours.

Figure 4 compares the (10$\bar{1}$0) lattice change of E34 and D084 at 300-400°C. The two-time melting pressure tube (E34) expands at 350° C or lower, while the fourth melting pressure tube (D084) contracts or contracts at 350° C, and then expands and returns to the origin. The (10$\bar{1}$0) lattice expansion according to the aging treatment at 400° C. is as large as twice that of the double melting pressure tube E34 and the quadruple melting pressure tube D084.

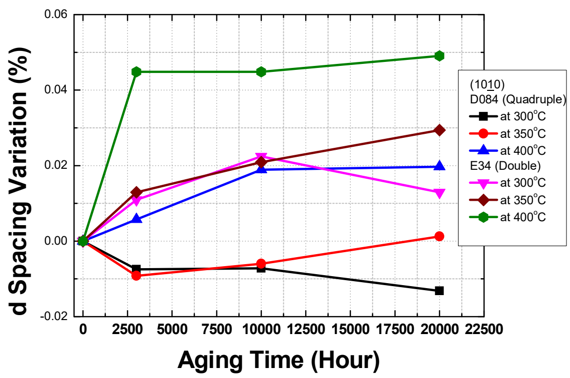

Fig. 4. Comparisons of {10$\bar{1}$0} lattice variation measured by neutron diffraction in E34 (Double melt) and D084 (Quadruple melt) pressure tube by aging at 300-400°C up to 20,000 hours.

Figure 5 compares the (0002) lattice change of E34 and D084 at 300-400°C. The double melting pressure tube E34 expands in an aging treatment at 350° C or less. On the other hand, the four-time melting pressure tube (D084) initially contracts slightly, then continues to contract or expands. The aging treatment at 400°C shows a similar behavior in lattice expansion, which expands and saturates by 0.08-0.09% at 3,000 hours.

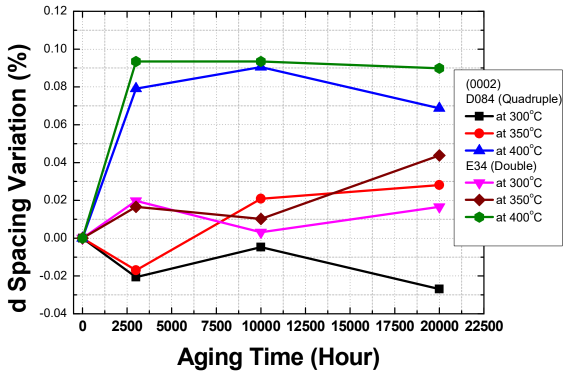

Fig. 5. Comparisons of {0002} lattice variation measured by neutron diffraction in E34 (Double melt) and D084 (Quadruple melt) pressure tube by aging at 300-400℃ up to 20,000 hours.

4. Conclusions

(1) Thermal treatment of double melt Zr-2.5%Nb pressure tube material causes anisotropic expansion at 400℃ or higher regardless of direction, and it is likely due to entropy increase.

(2) The lattice change of the (10$\underline{1}$0) plane expands to about 0.045% at 400°C for 3,000 hours and then becomes saturated, whereas the lattice change of the (0002) plane expands to about 0.09% at 400°C for 3,000 hours or more and then becomes saturated.

(3) Double melting pressure tube (E34) shows lattice expansion regardless of temperature in 300-400℃ aging treatment, whereas quadruple melting pressure tube (D084) causes contraction at 350℃ or lower and lattice expansion at 400℃ or higher .

(4) The reason that the transverse creep of the CANDU pressure tube appears higher than the design estimate is because the (0002) plane is mainly aligned in the transverse direction and the high lattice expansion rate of this lattice plane.

(5) The fast neutron irradiation of the CANDU reactor disturbs the atomic arrangement, which increases entropy and promotes the diffusion of atoms.

(6) In the Zr-2.5%Nb pressure tube material, the driving force of lattice expansion due to aging treatment is the entropy remaining in the manufacturing process, and the driving force of the expansion of the movable pressure tube exposed to neutron irradiation is judged to be due to the continuous increase in entropy due to irradiation.

Acknowledgments

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