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The Characteristics of Hydride Precipitation with Hydrogen Concentration and Homogenization Temperature in Zr-2.5%Nb Pressure Tube Material

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1. Introduction

Zr-2.5%Nb alloy, which is excellent in high-temperature strength, corrosion resistance, and neutron absorption cross-sectional area, is used for pressure tube material in pressurized heavy water reactor (PHWR). Since the primary water flows through the pressure tube during the operation of PHWR, some of the hydrogen reacted with Zr is absorbed into the pressure tube [1]. The pressure tube materials containing hydrogen precipitate hydrides, when the hydrogen concentration exceeds the solubility of hydrogen during cooling process of pressure tube. The precipitation of hydride lowers the fracture toughness of the pressure tube materials. In addition, hydrogen in the pressure tube material tends to be precipitated at the stress concentration site at the crack tip and causes crack initiation and propagation by the delayed hydride cracking mechanism [1].

When zirconium hydride precipitates at low temperatures in dilute, coarse-grained, single phase zirconium alloys, the morphology and habit plane of the hydride are dictated by the shear or displacive nature of the phase transformation [2].

Therefore, it means that the behavior of precipitation of hydrides in the pressure tube material directly is closely related to the integrity of the pressure tube. This study quantitatively analyzed the effects of hydrogen concentration and homogenization temperature on the behavior of precipitation of hydrides in Zr-2.5%Nb pressure tube material.

2. Experimental

The pressure tube material used in the experiment is Zr-2.5% Nb alloy. The chemical composition is shown in Table 1. The material of the pressure tube was cut into 30 × 100 mm pieces and hydrogen was charged in a 1 molar sulfuric acid solution at a current density of 100mA/cm^2. The solution temperature is controlled to be 85 ± 5°C for 24 to 40 hours. After hydrogen charging, the specimen was homogenized in vacuum sealed tubing for 240 hours at 240-550°C.

The hydrogen concentration of the homogenized specimen was analyzed by using vacuum extraction method. The hydrogen concentration is determined by the average of three specimens. Separately from the hydrogen concentration measurement, differential scanning calorimeter (DSC) was used to confirm the actual homogenized temperature at which the hydride specimen was maintained [3, 4].

As a result, the hydrogen concentration did not increase directly with the homogenization treatment temperature. This is presumably because the amount of hydride precipitated on the surface by the electrolysis method was not sufficient. These specimens were not charged at equilibrium concentration of homogenized temperature, but the objective of this experiment is to analyze the precipitation pattern according to both hydrogen concentration and homogenization temperature.

The hydrides were observed by optical microscope after polishing and etching in radial-transverse plane (plane normal to longitudinal direction). The parameter of hydrides like number and length etc. are counted quantitatively with hydrogen concentration. Hydrides were observed at a magnification of 200 times, and 20 photographs of each specimen were taken and analyzed. The effect of hydrogen concentration and treatment temperature on hydride precipitation was investigated.

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

Zr	Nb	Fe	O	N	P
97.4	2.5	0.05	0.097	0.007	0.003

3. Results and Discussions

The hydride of the as-received specimen containing 14 +/- 2 ppm of hydrogen is shown in Figure 1 a). The hydrides of 28 and 86 ppm of hydrogen are shown in Fig. 1 b) and c). As the hydrogen concentration increases, the number of hydrides increases and the length increases. Although the length of hydrides in the transverse direction is a few microns to several microns, the grain size is less than 1 micron in radial direction and is a few microns in transverse direction. This means that the hydride penetrate several grains and precipitates in a transgranular manner. This seems to be due to the hydride nucleates in α/β interface in Zr-2.5%Nb pressure tube material.

Figure 2 shows the histogram of the frequency of hydride length for the specimen shown in Figure 1. As the hydrogen concentration increases, the number of hydrides from 5 to 15 μm increases sharply at 28 ppm of hydrogen as shown in Figure 2 b). As the hydrogen concentration increases to 86 ppm of hydrogen, the number of hydrides longer than 20 microns increases whereas those of 5 to 15 microns decrease. According to DSC analysis, the TSSD (terminal solid solubility od dissolution) of 86 ppm of hydrogen was 324℃, but it seems that the increase of hydrogen concentration at this temperature mainly increased the length of hydride.

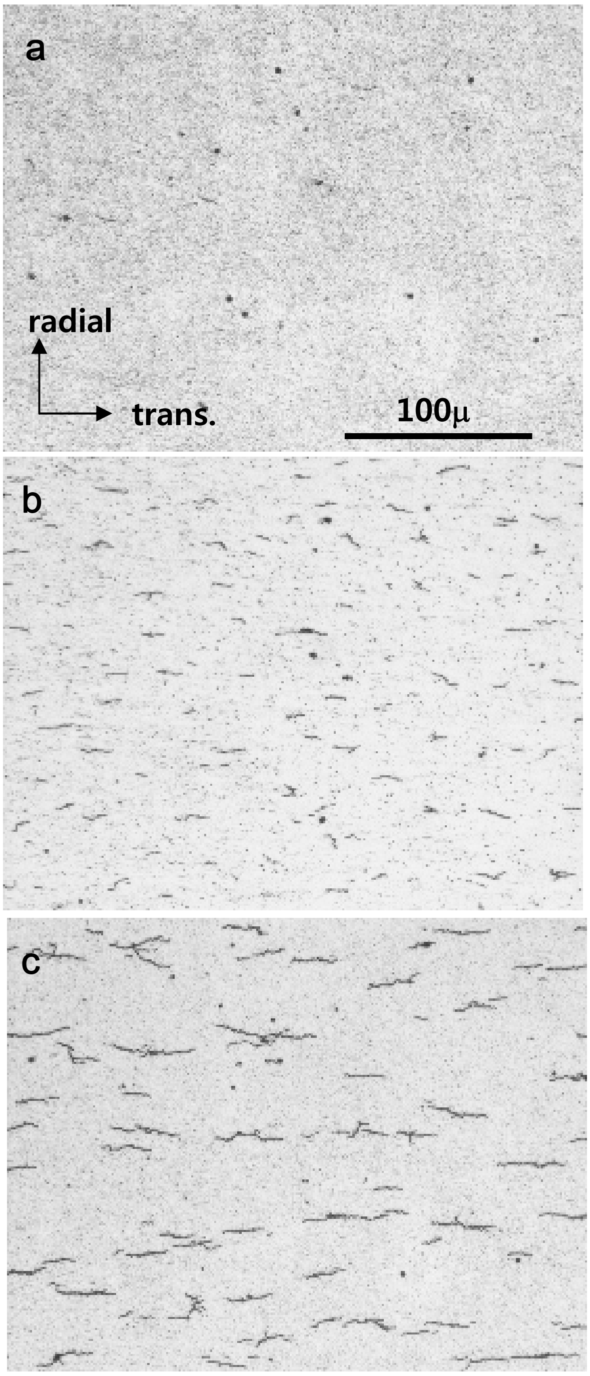

Fig. 1. Morphology of hydrides in Zr-2.5%Nb pressure tube material containing hydrogen, a) 14 ppm, b) 28 ppm, and 86ppm.

As shown in Fig. 3, the fraction of hydrides increased almost linearly with the concentration of hydrogen. The area fraction of hydrides is shown in Figure 3 although the homogenization treatment temperature varies from 300 to 550℃. This is reasonable since the number of hydrides increases with the concentration of hydrogen.

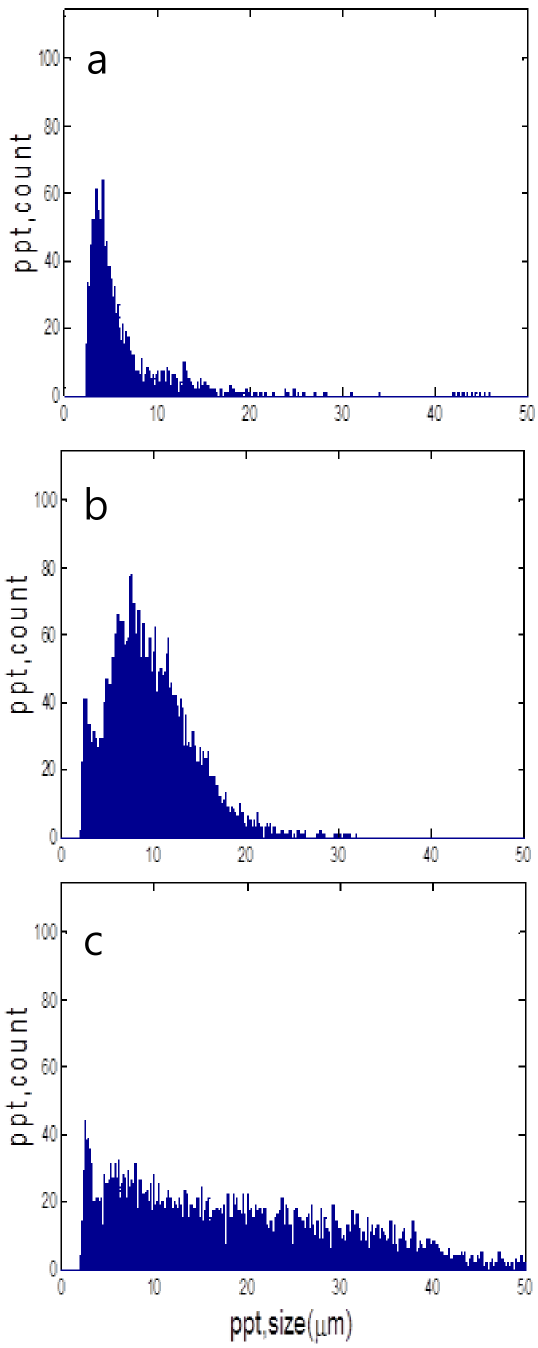

Fig. 2. Distribution of length of hydrides in Zr-2.5%Nb pressure tube material containing hydrogen, a) 14 ppm, b) 28 ppm, and 86ppm.

It is necessary to analyze the influence of the homogenization temperature on the precipitation behavior of hydrides. However, the hydrogen concentration does not increase with homogenization temperature in this study. In order to compare the parameters of hydrides reasonably, it is necessary to normalize the parameters of hydride precipitation.

The following equation was used to normalize the effect of hydrogen concentration to be 50 ppm of hydrogen. For example, the length of 28 ppm of hydrogen normalized to 50 ppm H was calculated as follows.

As the homogenization temperature increases, the ratio of

Normalized length $_{(50\ ppm)}$ = Length $_{(28\ ppm)}$ x 50/28 (1)

Where the length $_{(28\ ppm)}$ is the average hydride length of the specimen, 50 is the hydrogen concentration to be normalized, and 28 ppm is the hydrogen concentration of a specimen.

The ratio of the length to thickness of hydrides calculated by equation (1) is plotted against homogenization temperature in Fig. 4. The ratio of length to thickness decreases gradually. This seems to be due to the fact that the continuity of beta phase breaks down as the homogenization increases at above 400℃, since it is known that the hydride preferentially nucleates in α/β interfaces [2]. The microstructure of as-received Zr-2.5%Nb pressure tube material is consisted of α-Zr surrounded by β- zirconium. This means that the α/β phase in as-received condition is nearly continuous, whereas the α/β interface in homogenized at above 400℃ is discontinuous. It is natural that the discontinuity of α/β interface inhibits the growth of hydrides longer [2].

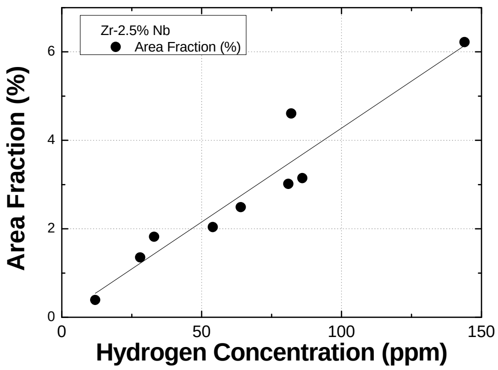

Fig. 3. Area fraction of hydrides with hydrogen concentration in Zr-2.5%Nb pressure tube material.

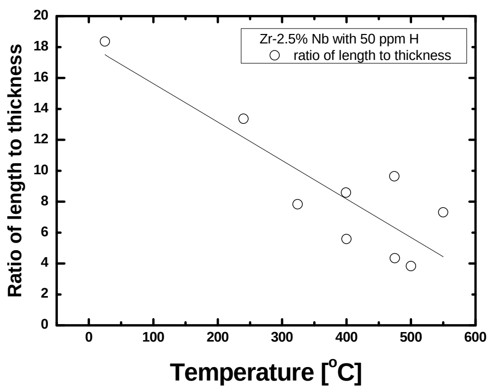

Fig. 4. Ratio of length to thickness of hydride in normalized to be 50ppm H with homogenization temperature in Zr-2.5%Nb pressure tube material.

4. Conclusions

The results of the quantitative analysis of the effects of hydrogen concentration and homogenization temperature on the characteristics of hydride precipitation in Zr - 2.5% Nb alloy pressure tube material were as follows.

1. The fraction of hydrides increases linearly with hydrogen concentration regardless of the homogenization temperature.
2. The increase in hydrogen concentration increases the length of the hydride to some extent, but homogenization at above 400℃ increases the number of hydrides shorter than 30 µm.
3. The ratio of length to thickness of hydrides decreases with increase in homogenization temperature, based on the normalized results to 50 ppm of hydrogen.

Acknowledgments

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