The Effect of Hoop Stress, Hydrogen Content and Cooling Rate on Hydride Reorientation of Oxidized Zr Alloy Cladding Tubes

Ju-Jin Won*, Hyun-Jin Cha, Kyu-Tae Kim
Nuclear and Energy Engineering Department, Dongguk University
123, Dongdea-Ro, Gyeongju, Republic Of Korea, 780-714
*Corresponding author: chuchinwon@dongguk.ac.kr

1. Introduction

The amount of hydrogen content in nuclear fuel cladding is increased, and fuel corrosion accelerated because of the extended fuel cycle. After the burning task is completed at nuclear power plant, the nuclear fuel is moved to fuel pit from nuclear reactor for cool-down. After the cool-down process in the wet storage, the nuclear fuel is stored for more than 50 years in the dry storage. During the dry process, fuel temperature increases from 60 to 400℃, and tensile hoop stress is applied to the cladding because external pressure is less than internal pressure. Hydrogen concentration of the fuel cladding is known to be between 300~600ppm under the burning condition in the nuclear reactor. When a certain threshold tensile hoop stress which is larger than 75~80MPa is applied on the cladding during the cool-down process of the dry storage, the hydrides in circumferential direction are rearranged to the hydride in radial direction. The formation of radial hydrides in the cladding tubes causes the degradation of mechanical properties.

Therefore, the goal of this study is to find the effect of hoop stress, hydrogen content and cooling rate on hydride reorientation of oxidized Zr alloy cladding tubes**.**

2. Experimental Setup

2.1 Specimen Preparation

The Zr-Nb alloy cladding tubes used in the PWRs were tested in this study. The chemical compositions of the Zr-Nb alloy cladding are shown in Table I. As shown in Fig.1, the external diameter and thickness of the ring test specimen are 9.5mm and 0.57mm, respectively. The vacuum furnace was used for charging hydrogen in cladding tubes. The tubes were charged with hydrogen for 24 hours in a vacuum furnace at 420^{o}C containing a mixture gas of hydrogen (150torr) and helium (200torr) to generate a uniform distribution of hydrogen atoms through the tubes. The hydrogen concentrations of the test specimens were analyzed by the LECO hydrogen analyzer. Two kinds of hydrogen contents in the hydrogen-charged specimens were measured to be 250ppm and 500ppm, respectively. After the hydrogen charging, the test specimens were oxidized in a autoclave with a pressure 17.5MPa at 360℃ and water of 70ppm LiOH for 30days. As shown in Fig.2, 2.5μm oxide was generated at the surface of the specimens after the oxidation test.

Table I: Chemical Compositions of Zr-Nb Alloy (wt %)

Nb	Sn	Cr	Fe	Zr
1.0	1.0	-	0.1	Bal.

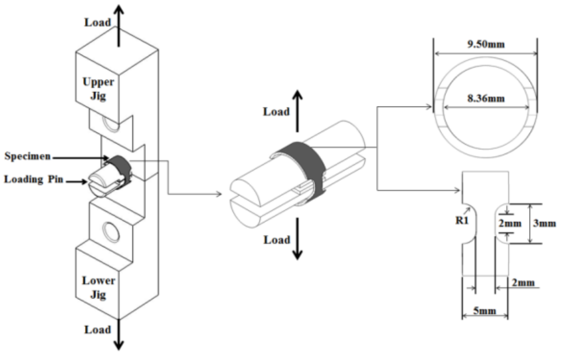

Fig.1. Zr Specimen size and jig used in this study

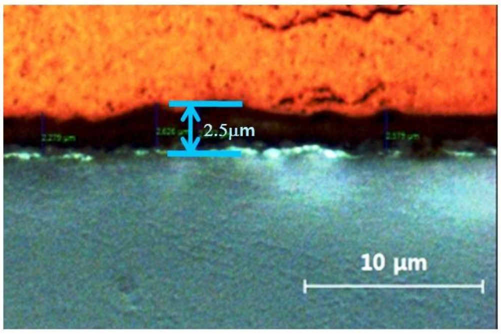

Fig.2. SEM micrographs of oxidized specimens

2.2 Experiment Procedure

As shown in Fig.3, the test specimens were heated up to 400^{o}C under the zero stress condition and hold for 2 hours at 400^{o}C using the KLES 500-S creep tester. Then the test specimens were cooled down with three cooling rates of 0.3, 2.0 and 8.0^{o}C/min under the 80, 100 and 150MPa stress condition. Using the Instron model 3366 mechanical testing machine, the tensile tests were carried out with 0.020mm/min. After the tensile tests, the gage regions and fracture surfaces of the tensile-tested specimens were examined by optical microscopy and scanning electron microscope, respectively.

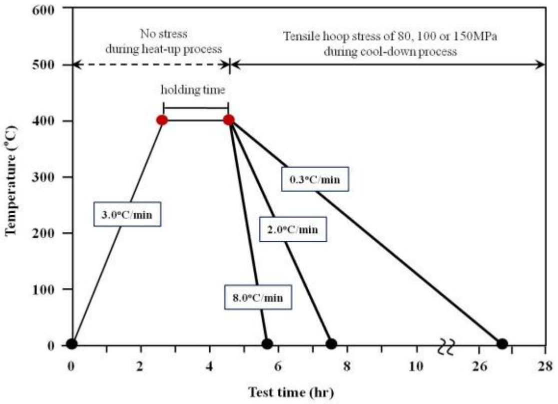

Fig.3. Heat-up and cool-down processes

3. Results and Discussion

Fig.4 shows the optical micrographs for 250ppm-H specimens after the cool-down process with the cooling rate of 8.0, 2.0 and 0.3^{o}C/min. It can be seen that the higher hoop tensile stress generated the larger radial hydride fraction. In addition, the slower cooling rate generated the longer radial hydride and larger radial hydride fraction. These phenomena are explained by a residence time at a relatively higher temperature during the cool-down. The slow cooling rate provides a sufficient time for searching out the radial hydride nucleation sites and growth of radial hydride.

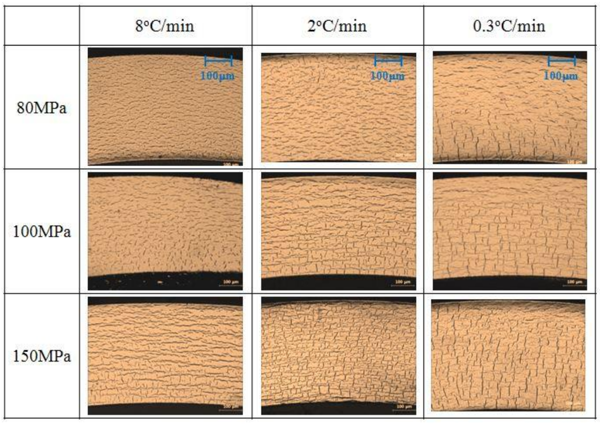

Fig.4. Optical micrographs of the 250ppm-H specimens after the cool-down processes.

Fig.5 shows the optical micrographs for 500ppm-H specimens after the cool-down process with the cooling rate of 8.0, 2.0 and 0.3^{o}C/min. In common with the Fig.4, the higher hoop tensile stress and the slower cooling rate generated the larger radial hydride fraction and the longer radial hydride length. In comparison with 250ppm-H specimens, 500ppm-H specimens generated smaller radial hydride fractions and shorter radial hydrides. It may be explained by the remaining circumferential hydrides for 500ppm-H specimens. The solid solubility for precipitation at 400^{o}C is 240ppm. Therefore, the remaining circumferential hydride of 250ppm-H specimens is negligible but that of 500ppm-H specimens is about 250ppm. The remaining circumferential hydrides may block up the growth of radial hydrides.

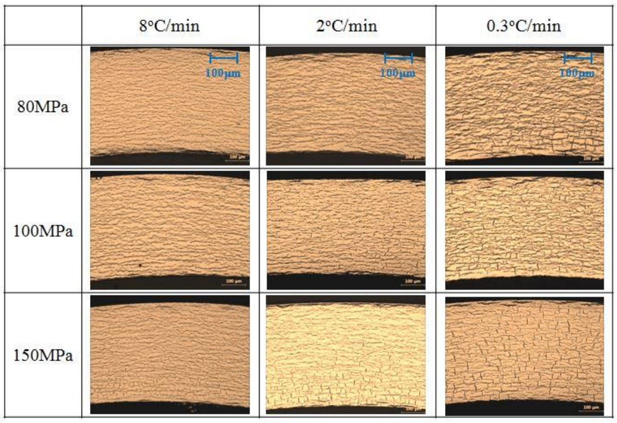

Fig.5. Optical micrographs of the 500ppm-H specimens after the cool-down processes.

Figs. 6 and 7 show the tensile test results of 250 and 500ppm-H specimens after the three cool-down tests under the 150MPa stress condition. It can be seen in these figures that the ultimate tensile strength and elongation were decreased with the slower cooling rate. However, it is noteworthy that the elongation of 500ppm-H specimens was larger than one of 250ppm-H specimens, which can be explained by the difference of radial hydride fraction generated in respective specimens. The crack propagation may continue through the interlinked radial hydrides, resulting in the reduction of the ultimate tensile strength and elongation.

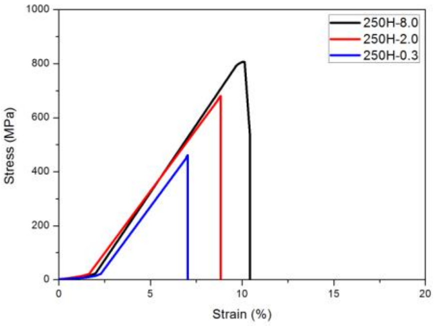

Fig.6. Tensile test results of 250ppm-H specimens after the cool-down process under 150MPa stress condition

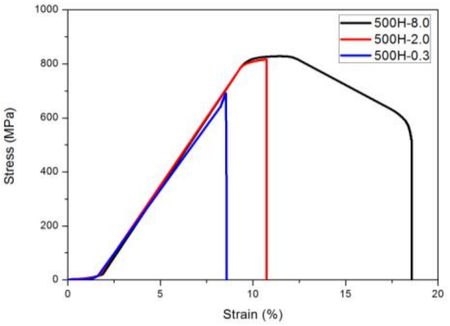

Fig.7. Tensile test results of 500ppm-H specimens after the cool-down process under 150MPa stress condition

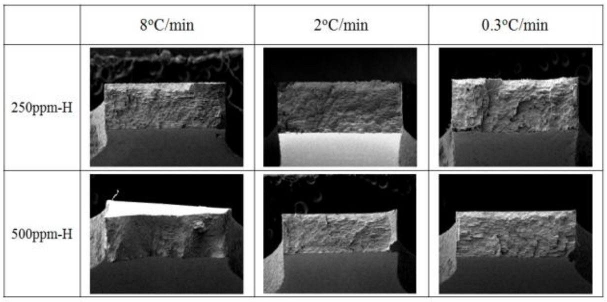

Fig.8. SEM micrographs of fracture surfaces on the tensile-tested specimens after the cool-down process under the 150MPa stress condition

Fig.8 shows SEM micrographs of the tensile-tested specimens after the cool-down process under the 150MPa stress condition. Based on the fracture surface examination shown in Fig. 8, the 500ppm-H specimen cooled down with the cooling rate of 8^{o}C/min shows a ductile fracture mode as predicted in Fig.7. On the other hand, the 250ppm-H specimens generated a typical brittle mode especially with the cooling rate of 0.3^{o}C/min.

3. Conclusions

The effect of hoop stress, hydrogen content, cooling rate on the hydride reorientation of oxidized Zr-Nb alloy cladding tubes was investigated using 250ppm-H and 500ppm-H Zr-Nb cladding tubes containing 2.5μm oxide layer at the surface of the cladding tubes. The 250ppm-H specimens generated larger radial hydride fractions and longer radial hydrides than the 500ppm-H ones because the latter contains the remaining circumferential hydrides that might block up the growth of the radial hydrides during the cool-down. On the other hand, the higher hoop tensile stress and the slower cooling rate generated the larger radial hydride fraction and longer radial hydride. It may be explained by the reduction in tensile stress-induced radial hydride nucleation energy as well as a residence time at a relatively higher temperature during the cool-down process. Tensile tests indicate that ultimate tensile strength and elongation were decreased with the slower cooling rate because the crack propagation may continue through the interlinked radial hydrides prevailing in the slowly cool-downed specimens.

REFERENCES

[1] H. Lee, Effect of Hydride Precipitation on Mechanical Property Degradation of Zirconium Alloy Cladding, Hanyang university, 2013.

[2] S.Y Kim, The study of Fuel Cladding Degradation on Interim Dry Storage System, p. 7~8, Kyunghee University, 2012.

[3] Y.S. Kim, Development of Spent Fuel Integrity Evaluation Technology for the Long-term Dry Storage, Hanyang university, 2010.

[4] S.S. Kim, The Observation of Hydride Precipitates in Zr-2.5Nb Pressure Tube, KSME 02MF009, p55-60, 2002

[5] H.M. Chung, Proceedings of the International Meeting on LWR Fuel Performance, Orlando, Florida, September 19–22, 2004

[6] Bai, J.B., Ji, N., Gilbon, D., Prioul, C., and Francois, D., “Hydride Embrittlement in Zircaloy-4 Plate: Part II. Interaction between the Tensile Stress and the Hydride Morphology, Metallurgical and Materials Transactions, Vol. 25A, June 1994

[7] Kammenzind, B. E, Berquist, B. M., Bajaj, R., Kreyns, E H., and Franklin, D. G., "The Long-Range Migration of Hydrogen Through Zircaloy in Response to Tensile and Compressive Stress Gradients," Zirconium in the Nuclear Industry: Twelfth International Symposium, ASTM STP 1354, pp. 196-233, 2000

[8] NRC,2001. Preliminary NRC Staff Guidance for 10 CFR Part 72 License Renewal, U.S. Nuclear Regulatory Commision, March, 29, 2001

[9] V. Jain, G. Cragnolino, L. Howard, A REVIEW REPORT ON HIGH BURNUP SPENT NUCLEAR FUEL-DISPOSAL ISSUES, NRC, CNWRA 2004-08, 2004

[10]H. C. Chu at al. Hydride reorientation in Zircaloy-4 cladding, Journal of Nuclear materials 373 (2008) 319-327

[11]H. Chung, R. Daum, J.Miller, and M.Billone, Zirconium in the Nuclear Industry, p. 561, ASTM STP 1423 (2002).

[12] H.A. Wriedt and R.A. Oriani, Effect of Tensle and Compressive Elastic Stress on Equilibrium Hydrogen Solubility in a Solid, Acta Metallurgica, Vol. 18, 1970.