

A Study on the Effect of Oxygen Content and Cooling rates on Hydride reorientations of Zirconium alloy cladding tubes

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1. Introduction

The fuel corrosion accelerated because of the extended fuel cycle. After the burning task, spent fuel rods are transferred to wet storage pit and cooled down. After the cool-down process in the fuel pit, spent fuel rods are moved to the interim dry storage. During the dry process, hoop tensile stresses and heat-up temperatures may occur on the cladding tubes because cladding temperature increased from 60°C to 400°C when the spent fuel rods are transferred from the wet storage pit to the interim dry storage cask. The existing circumferential hydrides in the spent fuel claddings may be dissolved during heat-up process and then the dissolved hydrogen atoms may be precipitated mainly in the radial direction during the cool-down process if the hoop tensile stress on the cladding is larger than a certain threshold stress. The threshold stress of radial hydride formation was reported to be about 90MPa in the temperature range of between 250 and 550°C [1,2]. In this study, therefore, the effect of cooling rate, oxygen and hydrogen content on hydride reorientation of oxidized and non-oxidized Zirconium alloy cladding tubes was examined.

2. Experimental Setup

2.1 Specimen Preparation

The Zr-Nb alloy cladding tubes that were supplied by KEPSCO Nuclear Fuel Company were used in this study. The chemical compositions of the Zr-Nb alloy cladding are given in Table I.

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

Nb	Sn	Cr	Fe	Zr
1.0	1.0	-	0.1	Bal.

Prior to hydrogen charging, the tubes were cut into 100mm length. Two parts of the tubes were charged with hydrogen for 24 hours in a vacuum furnace at 420°C containing a mixture gas of hydrogen (150torr) and helium (200torr) to generate a uniform distribution of hydrogen atoms through the tubes. The hydrogen contents of the test specimens were analyzed by the LECO hydrogen analyzer RH600. The measured hydrogen contents were 250ppm and 500ppm,

respectively. Fig.1 shows a schematic configuration of the test specimen. The external diameter and thickness of the ring test specimen are 9.5mm and 0.57mm, respectively. After the hydrogen charging, some of ring test specimens were oxidized for 60days at 360°C and 17.5MPa in autoclave under the water atmosphere containing 70ppm LiOH. As shown in Fig.2, 3.6µm oxide was generated at the inner and outer surface of the specimens after the oxidation test.

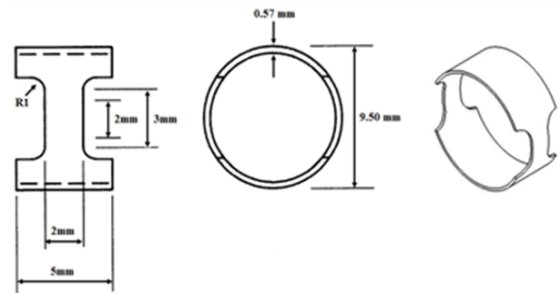


Fig.1. Dimensions of the test specimen

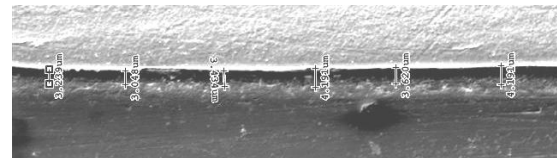


Fig.2. SEM micrographs of oxidized specimens

2.2 Experiment Procedure

As seen in Fig.3, the oxidized and non-oxidized specimens were heated up to 400°C under the zero stress condition and hold for 2 hours at that temperature using the KLES 500-S creep tester. Then the test specimens were cooled down with two cooling rates of 0.3 and 2.0°C/min under a tensile hoop stresses of 80 and 150MPa. After the hydride orientation tests, the tensile tests were carried out with 0.020mm/min using the Instron model 3366 mechanical testing machine. The fracture surfaces of the tensile-tested specimens were examined by scanning electron microscope, and the gage regions were examined by optical microscopy.

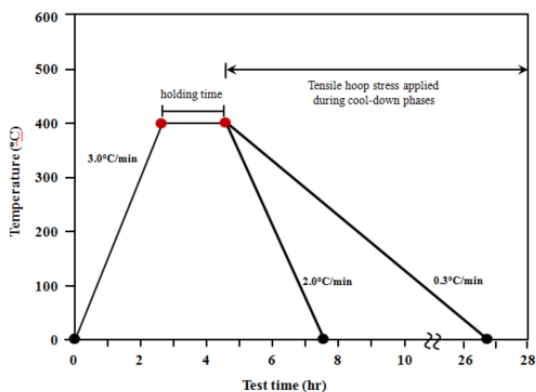


Fig.3 Heat-up and cool-down processes

3. Results and discussion

Fig.4 and Fig.5 show the optical micrographs for non-oxidized and oxidized specimens after the cool-down process with the cooling rate of 2.0 and 0.3°C/min under the tensile hoop stress of 80MPa and 150MPa, respectively. 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.

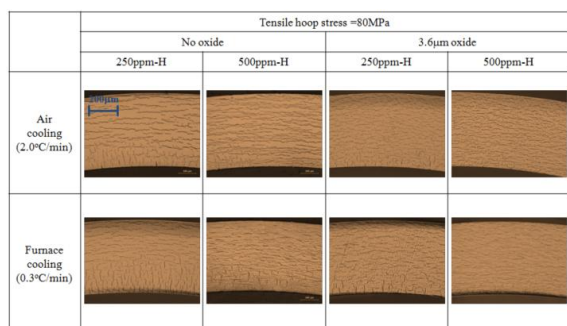


Fig.4 Optical micrographs of the non-oxidized and oxidized specimens after the cool-down processes under 80MPa

On the other hands, the 250ppm-H specimens generated larger radial hydride fractions and longer radial hydrides than the 500ppm-H specimens. It may be explained by solid solubility of hydrogen in the zirconium. There are larger amount of the undissolved circumferential hydrides of the 500ppm-H specimens at the heat-up temperature of 400°C. The undissolved circumferential hydrides may block up the growth of the radial hydrides.

As shown in Fig.4 and 5, non-oxidized specimens generated larger radial hydride fraction and longer hydride length than the oxidized specimens. It may be expected that oxygen and hydrogen atoms may occupy available radial hydride nucleation sites. As a result, the radial hydride nucleation energy may increase in the

oxidized specimens, and the radial hydride nucleation may be interrupted.

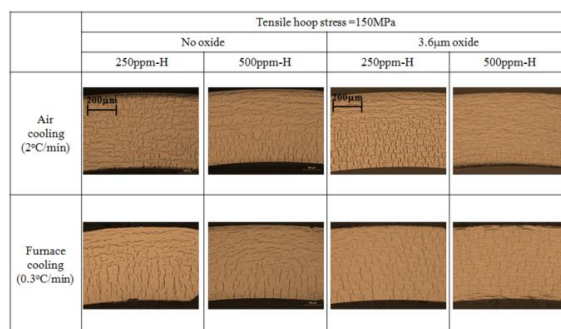


Fig.5 Optical micrographs of the non-oxidized and oxidized specimens after the cool-down processes under 150MPa

Fig.6 shows the scanning electron micrographs of fracture surfaces on the tensile-tested specimens after the hydride reorientation tests. Based on the fracture surface examination shown in Fig.6, the cleavage facet size becomes longer and wider with the decrease in the cooling rate. The crack propagation mechanism through the radial and circumferential hydrides may be described as follows. When the tensile hoop stress exceeds the critical stress for the micro-crack formation, micro-cracks may be generated at the radial hydrides and propagate along the radial hydrides.

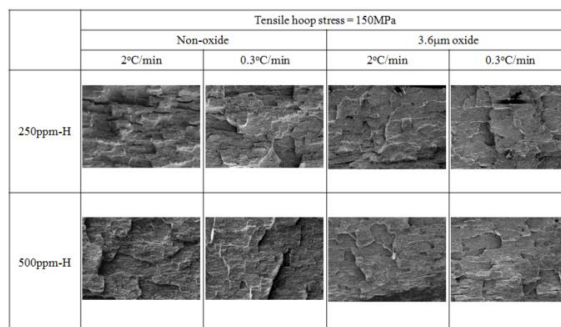


Fig.6 Scanning electron micrographs of fracture surfaces on the tensile-tested specimens (x500)

4. Conclusions

The effect of cooling rates, oxygen and hydrogen contents on the hydride reorientation of Zr-Nb alloy cladding tubes was studied using oxidized and non-oxidized specimens. It was shown that 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 slower cooling rate generated the larger radial hydride fraction and longer radial hydride. It may be explained by a residence time at a relatively higher temperature during the cool-down process. In addition, radial hydride fractions were reduced for oxidized specimens. It may be explained by the increase of radial hydride nucleation energy. The fracture surfaces on the tensile-

tested specimens were analyzed by the scanning electron microscope. The cleavage facet size becomes longer and wider with the decrease in the cooling rate.

REFERENCES

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- [2] A. Colas et al., "In-situ Study of Hydride Precipitation Kinetics and Re-orientation in Zircaloy Using Synchrotron Radiation," *Acta Mater.* 58, 6575 (2010).