Hydrogen Migration under Stress in Zirconium Alloy

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1. Introduction

To prepare for the decommissioning of Kori Unit 1, the spent nuclear fuel (SNF) in the nuclear power plant should be transferred. Even if SNFs are temporarily transferred to other units, due to insufficient wet storage capacities, SNFs will be needed to be stored in independent interim storage. In Korea, dry storage has been considered as the realistic solution for this issue.

It is important to establish the plans for managing the SNF dry storage system. Especially, because the integrity of SNF is related to the safety requirements, the studies on degradation mechanism are necessary. According to NUREG-2214, hydride reorientation is listed first among all degradation issues of SNF in dry storage system. Fuel claddings are hydrided during reactor operation and zirconium hydrides precipitate circumferentially when fuel is cooled in interim spent fuel pool. However, during dry process of spent fuel, precipitated hydrides dissolve again and re-precipitate radially because of hoop stress caused by fission gas. This phenomenon is names as hydride reorientation, and it makes the mechanical properties of the cladding reduced drastically. Though, mechanism of hydrogen migration in zirconium alloy is not clearly defined yet, factors affecting hydrogen diffusion in zirconium are known as concentration, temperature and stress[1, 2].

In this study, the proof of stress-induced hydrogen migration is shown experimentally. The tendency of hydrogen migration was observed through morphology based on three variables: hydrogen concentration, temperature, and stress. If the degree of migration is quantified by concentration, temperature and stress, it is expected to support the basis of the criteria of dry storage conditions.

2. Materials and Methods

2.1 Specimen preparation

The test specimens used in this study were un-irradiated Zirclaoy-4 Plate (25mm×25mm×0.9mm). The specimens were pickled in an acid solution to remove pre-filmed surface oxide. Prior to the tests, the specimens were charged with hydrogen (50 - 300 wppm) using gaseous hydrogen charging apparatus at 400 ℃ and cooled down to room temperature with cooling rate of 0.5 ℃/min.

2.2 Point load test

To develop the stress distribution in plate specimen, the point load was applied constantly in high-temperature condition. Fig. 1 shows a schematic diagram of thermal and loading history used in this study. Once specimen was thermally stabilized at peak temperature, constant load was applied to the specimen for 6 hours. The constant load was maintained at peak temperature for 1 hour and at cooling for 5 hours. The experiments are being carried out and Table I shows combination of peak temperature and load cases.

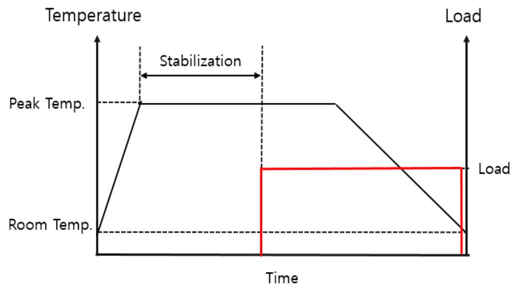

Fig. 1. Schematic diagram of thermal and loading history

Table I: Test matrix of point load test

Hydrogen Concentration (wppm)	Peak Temperature (℃)	Load (N)
50 - 300	290	150
	290	300
	350	150
	350	300
	400	150
	400	300
	420	250

2.3 Finite Element Method (FEM) analysis

3D finite element analysis was conducted to figure out stress distribution of the specimens under point load. 1/4 segment was modeled because of geometrical symmetry and material properties as a function of temperature were taken from MATPRO database. Fig. 2 shows stress distribution of point load test simulation.

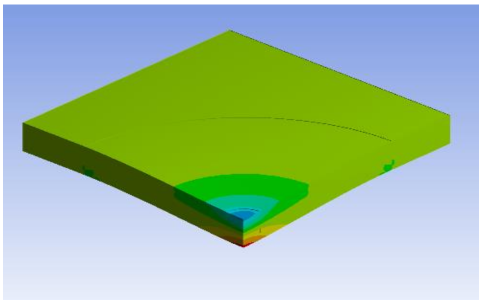

Fig. 2. Example result image of 3D finite element analysis of 1/4 segment of a specimen

3. Current Results

3.1 Hydride morphology and FEM results

To confirm the hydrogen behavior consequences of the preliminary test, hydride morphologies were observed. Fig. 3. shows cross-sectional morphologies of before and after the test. A specimen was charged with hydrogen about 150 – 200 wppm uniformly (left). After the test (420 ℃, 250 N), hydride reorientation was observed.

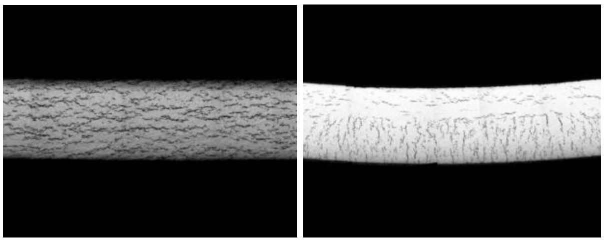

Fig. 3. Hydride morphology before (left) and after test (right)

FEM results and morphology images were merged to see the effect of stress-gradient on hydrogen behavior. The maximum tensile stress was calculated as 414 MPa when 250 N of load was applied. Also, maximum tensile stress was observed at the opposite side of the load contact surface and hydride reorientation occurred only in the region under tensile stress.

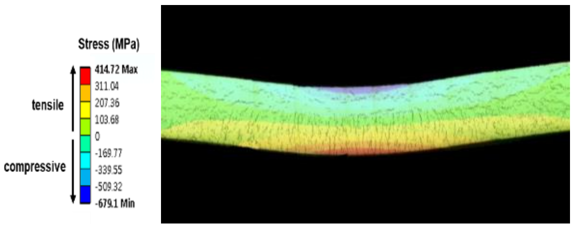

Fig. 4. Merged image of FEM results and morphology: 150wppm (expectation), 420 ℃, 250 N

3.2 Peak temperature effect on hydrogen migration

It is well known that hydrogen dissolves in zirconium matrix following the terminal solid solubility (TSS) system. Under the same temperature, concentration of dissolved hydrogen is same by the TSS system. It means hydrogen amount which can migrate and re-precipitate is equal under the same temperature. Therefore, as the hydrogen concentration increases, the fraction of hydride reorientation decreases. Fig. 5 shows morphologies of each hydrogen concentration (50, 100, 150, 300 wppm) and tendency of fraction of hydride reorientation.

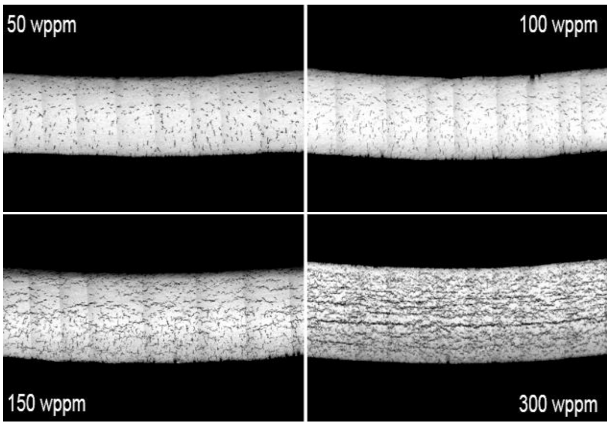

Fig. 5. Morphology by hydrogen concentration under same peak temperature and load (350 ℃, 300 N)

4. Conclusions and Future Plans

In this study, hydrogen migration under stress was experimentally shown through the three factors: hydrogen concentration, peak temperature and stress. First, if certain conditions are met, uniformly existed hydrogen can migrate under stress in zirconium alloy. However, if the temperature is not sufficient for hydride to be re-dissolved, hydrogen migration is disturbed. In other words, the fraction of hydride reorientation decreases when peak temperature decreases or hydrogen concentration increases.

In the future, additional experiments with various combination of the three factors will be conducted. And then, to fulfill the effect of hydrogen migration, micro-hardness tests will be conducted in the direction of stress-gradient. Also, to confirm the migration of hydrogen quantitatively, hydrogen concentration will be measured using an element analyzer (ONH-2000, ELTRA)

REFERENCES

[1] B. F. Kammenzind, ASTM STP 1354 (2000) 196-233.
[2] J. M. Lee, H. A. Kim, D. H. Kook, Y. S. Kim, J. Nucl. Mater., 509 (2018) 285-294