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Understanding mechanical integrity Zircaloy with radial hydrides via image analysis of hydride morphology

Dahyeon Woo, Youho Lee*
Department of Nuclear Eng., Seoul National Univ., 1 Gwanak-ro, Gwanak-gu, Seoul 08826, Republic of Korea
*Corresponding author: leeyouho@snu.ac.kr

1. Introduction

Since the fuel rod is a closed system, the rod internal pressure increases when the temperature rises to 400℃ during vacuum drying when moving from wet storage to dry storage. As a result, hoop stress is applied to the cladding. When a stress exceeding the threshold stress is applied, radial hydride is generated during the subsequent temperature decrease. Radial hydride is a factor that degrades the integrity of the cladding.

The operation condition determines the internal pressure of the fuel rod, the hydrogen concentration, and the temperature. Modeling to predict Radial Hydride Fraction (RHF) and Radial Hydride Continuous Path (RHCP) values with hydrogen concentration and rod internal pressure was conducted by Kim et al. [1]. As the next step, in this study, modeling to predict the mechanical properties of cladding according to RHF and RHCP is carried out. Since both RHF and RHCP are factors that can be calculated using optical microscopy (OM) images, the mechanical strength can be predicted using only the image of the material. Also, from a broader perspective, if it is integrated with RHF and RHCP modeling, it is possible to predict the mechanical strength of the cladding finally through the operation history of the fuel rod.

2. Experiments

2.1 Material

Zr-Nb alloy was used in the experiment, and the detailed chemical composition is shown in Table 1. The outer diameter is 9.5 mm, the thickness of the cladding is 0.57 mm and the total length is 250 mm. The cladding was charged with hydrogen using a gaseous diffusion method. Specimens with various hydrogen concentrations from 0 to 1200 wppm were prepared.

Table 1. Chemical composition of Zr-Nb alloy cladding tube used as specimen

Materials	Zr-Nb alloy
Zr	Bal.
Sn	0.6~0.79 wt%
Fe	0.09~0.13 wt%
O	0.09~0.16 wt%
Nb	0.8~1.2 wt%

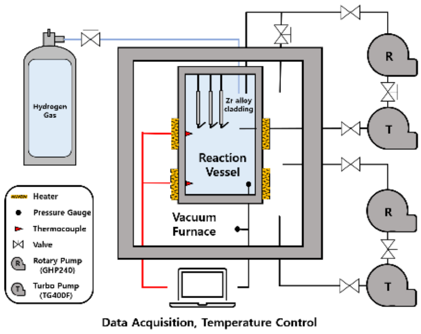

Fig 1. Illustration of hydrogen charging system

2.2 Hydride reorientation

Radial hydride was formed by the internal pressurization method. The pressure inside the rod was formed with Ar gas rather than hydraulic pressure. The chamber is filled with Ar 1 atm. Stress is generated by the pressure difference between the inside and outside of the cladding. Heat-up was performed for 2 hours to reach the dry storage start temperature of 400℃, and after 2 hours hold, cool down was performed for 10 hours to simulate cooling during dry storage. Depending on the magnitude of the stress, radial hydride is precipitated during the cool down process. After the hydride reorientation treatment, the samples were cut with height of 7 mm in the shape of rings, and then analyzed.

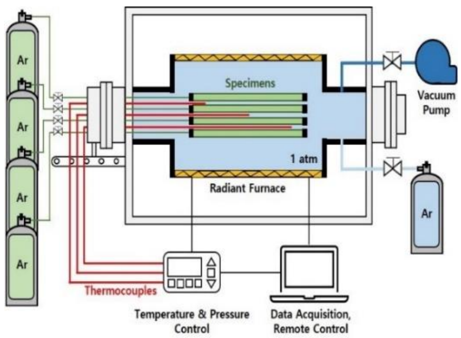

Fig 2. Illustration of hydride reorientation treatment system

2.3 Image analysis: PROPHET

There are two representative factors relevant to radial hydride: Radial Hydride Fraction (RHF) and Radial Hydride Continuous Path (RHCP). RHF represents the ratio of hydrides in the circumferential and radial directions. This is a widely used factor to explain the mechanical integrity of the hydride reoriented Zircaloy cladding. RHF definition of Eq.(1) was suggested by Raynaud et al. [2].

$$\mathrm{RHF} = \frac{\sum_i L_i f_i}{\sum_i L_i} \quad (1)$$

$$\mathrm{f_i} = \begin{cases} 0 : \ 0° \le \theta < 40° : circumferential \\ 0.5 : 40° \le \theta < 65° : mixed \\ 1 : 65° \le \theta < 90° : radial \end{cases} \quad (2)$$

RHCP provides a quantitative measure of hydrogen embrittlement by reflecting the geometrical characteristics of the circumferential and radial hydrides. This concept was developed by Simon et al. [3], recently. RHCP finds the minimum cost of crack propagation. It has a value between 0 and 1, and the closer to 1, the more brittle.

$$RHCP = \frac{Lw_{Zr} - (x_{Zr}w_{Zr} + x_{ZrH}w_{ZrH})}{L(w_{Zr} - w_{ZrH})} \quad (3)$$

$$w_{Zr} = 50MPa\sqrt{m}, \quad w_{ZrH} = 1MPa\sqrt{m} \quad (4)$$

Mechanical polishing and etching were conducted for each specimen. The cross section perpendicular to the axial direction was observed with Optical Microscope (OM). 200 magnification was used, and observations were made in four directions: east, west, north, and south.

Kim et al. [4] developed PROPEHT (PROPortion of radial Hydride Estimate Technique), an image processing code by MATLAB. When OM image is entered in PROPHET, RHF and RHCP values are calculated. PROPHET is available at the PROPET tab in https://fuel.snu.edu. The average value obtained by calculating both RHF and RHCP for 4 positions was used. Only one of the two sides of the cylinder was analyzed. It is assumed that the result represents a 7mm specimen.

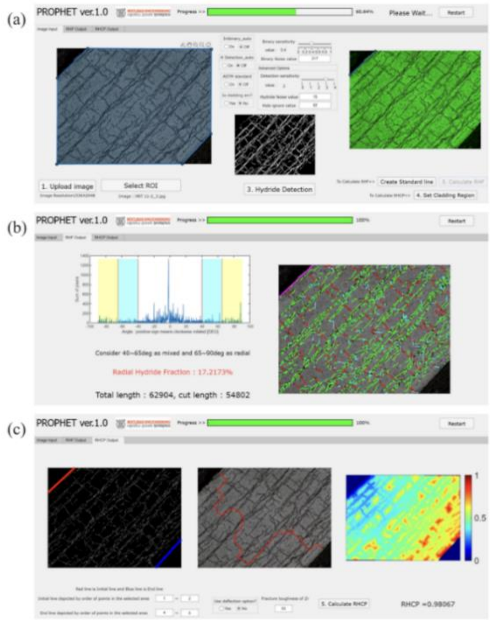

Fig 3. Run screen of PROPHET: (a) input screen, (b) RHF output screen, and (c) RHCP output screen

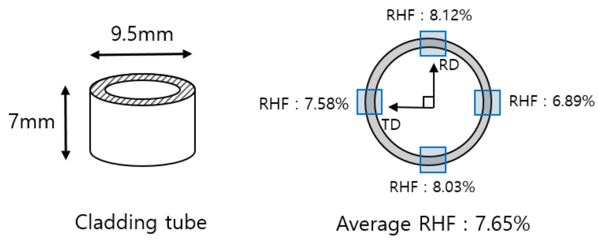

Fig 4. Geometry of specimen

2.4 Ring compression test

Ring compression test (RCT) was performed to measure the mechanical strength of the 7mm specimens. RCT was performed at room temperature, 25 ℃. As a result of RCT, Ultimate Strength and Strain Energy Density (SED) were calculated, and in this study, SED will be mainly dealt with. Lastly, the hydrogen concentration of 7mm specimen was confirmed with an ONH analyzer.

3. SED modeling

SED was shown for several factors that can explain the mechanical strength of cladding: hydrogen concentration, RHF, radial hydride concentration, and RHCP, respectively. Radial hydride concentration was obtained by multiplying hydrogen concentration by RHF. Overall, it was confirmed that a certain trend was shown for all factors. However, it was judged that hydrogen concentration, radial hydride concentration, and RHCP alone could not accurately explain SED, and only RHF alone seemed to clearly explain the tendency of SED.

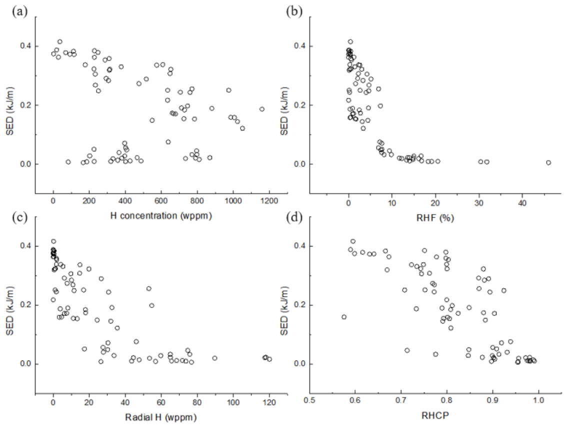

Fig 5. SED relationship: (a) with hydrogen concentration, (b) with RHF, (c) with radial hydride concentration, and (d) with RHCP

When RHF is greater than 5%, it is in exponential form. That is, in this area, the SED is the area of the physical model that can be explained only by the RHF. The fact that the prediction of SED is possible only with RHF has great significance in that the strength of the material can be predicted only with the OM image without fracture of the material.

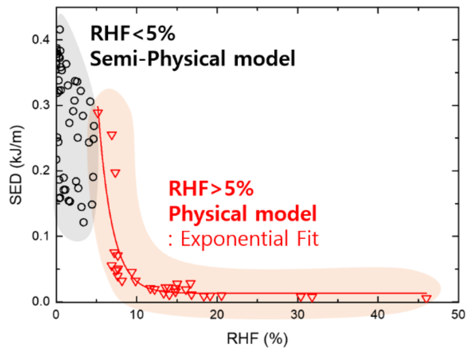

Fig 6. Relationship between RHF and SED

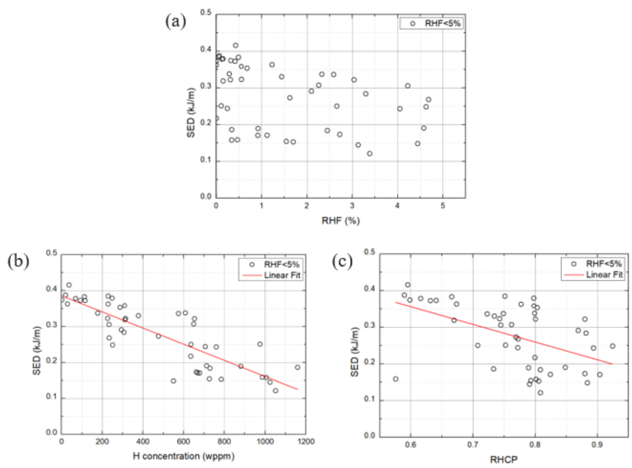

Fig 7. Relationship between SED and: (a) RHF, (b) hydrogen concentration, (c) RHCP

On the other hand, when RHF was less than 5%, the effect of radial hydride was insignificant, and there was no correlation between RHF and SED. As shown in Fig. 7, when RHF is 5% or less, the relationship between RHF and SED could not be confirmed. Hydrogen concentration and RHCP showed a linear relationship with SED although not perfect. Using machine learning, it is expected to investigate the relationship between RHF, hydrogen concentration, and RHCP and SED.

4. Advanced insight of the effect of hydrogen on SED

A total of 79 specimens were tested. The graph of hydrogen concentration and radial hydride concentration as a result of RHF controlled through rod internal pressure is as above. In order to check how the total amount of hydrogen and radial hydride affect the mechanical properties of the cladding, it was divided into four areas: region 1, region 2, region 3, region 4. Four data were selected to confirm the trend in each region: A, B, C, and D. These four data consist of a combination of low and high hydrogen concentration and radial hydride concentration, respectively. The results of RCT and OM, RHF, and RHCP of these four points are shown in Figs 9 and 10.

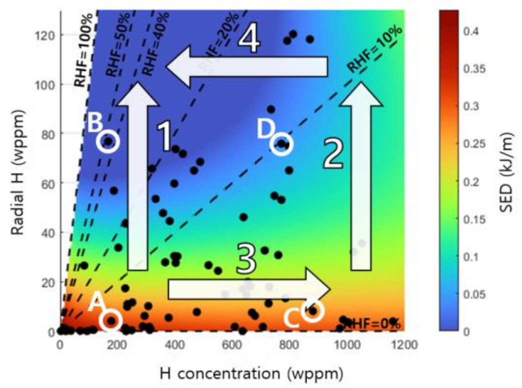

Fig 8. Colormap of SED with hydrogen concentration and radial hydride concentration

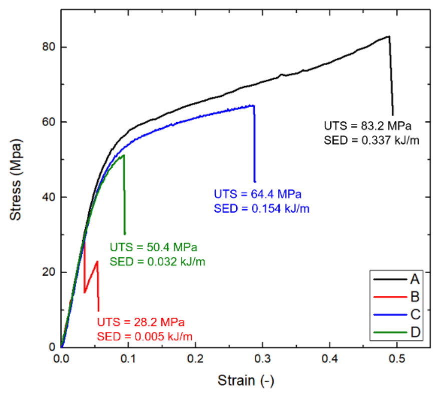

Fig 9. Stress-strain curve of four points, A, B, C, D in colormap

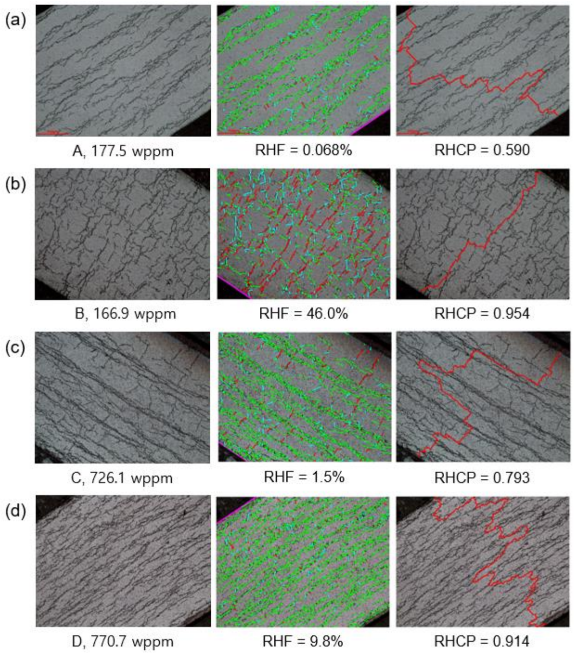

Fig 10. OM image and RHF, RHCP result: (a) case A, (b) case B, (c) case C, (d) case D

4.1 Radial Hydride effect (Region 1, 2)

A and B have similar total hydrogen concentration, but B has a higher radial hydride concentration. Since it is known that cladding becomes brittle by radial hydride, it can be seen that the SED decreases from A to B. Similarly, the total hydrogen concentration of C and D is similar, and it can be seen that the SED of D is lower than that of C due to the effect of radial hydride. And in the case of A and B with a lower total concentration, the difference in SED is larger than that of C and D with a higher total concentration. That is, if hydrogen concentration is lower, small amount of radial hydride result in the change in SED.

4.2 Total hydrogen concentration effect (Region 3)

In the absence of radial hydride, the effect of hydrogen concentration was investigated. Cases A and C have 4.1 wppm and 11.2 wppm of radial hydride, respectively, which is a level at which radial hydride does not affect the behavior of cladding at all. From the RCT result graph of Fig 9, it can be seen that the UTS and SED values decrease as the hydrogen concentration increases.

Since the fracture toughness of hydrogen is smaller than that of zirconium, the cladding becomes brittle when the concentration of hydrogen increases. In Fig 10, almost all hydrogen appeared in the circumferential direction, green lines, and the difference in hydrogen concentration between the two cases can be easily confirmed with the naked eye. RHCP also had a larger value in the case of (2) with a higher hydrogen concentration, which means that crack propagation is more likely when the hydrogen concentration is high. During the operation of a nuclear reactor, the hydrogen concentration is determined by the operating time, usually burnup. Through this, the mechanism by which high burnup nuclear fuel is weakened due to hydrogen embrittlement can be confirmed.

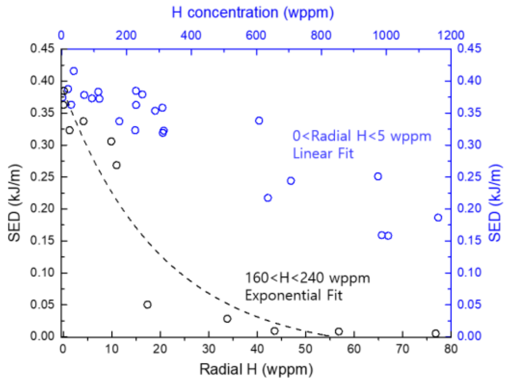

Fig 11. Comparison of region 1(black line) and 3(blue line) in colormap

Since radial hydride is known to be a factor that greatly degrades the soundness of spent nuclear fuel, it can be confirmed that the mechanical strength of the cladding is weakened even in region 1. Then, since both region 3 and region 1 degrade cladding, we compared the degree of cladding. Specimens in which the amount of radial hydride was 5 wppm or less were selected as the data of region 3, and specimens having a hydrogen concentration of 160 wppm and 240 wppm or less were selected as data of region 1.

Of course, in both cases, the SED decreased as each concentration increased, but the extent was greater with the radial hydride. When the hydrogen concentration increased from 0 to 1200 wppm, the SED decreased by about 0.25 J/m, but the radial hydride reduced the SED by almost 0.4 J/m with just 60 wppm.

4.3 Total hydrogen concentration effect: with radial hydride (Region 4)

According to the previous discussion, high hydrogen concentration results in low SED, so it is correct that the SED of D with a high total hydrogen concentration is lower when comparing the cases B and D with the same amount of radial hydride. However, as can be seen at Fig. 8 and Fig. 9, the result is the opposite. When the amount of radial hydride is large, it is more brittle when the concentration of total hydrogen is low. This is explained through the RHCP value. The RHCP of B is 0.954, and the RHCP of D is 0.914. Because hydrides are more brittle than the Zr matrix, they prefer to go through hydrides when cracks propagate. Therefore, in wall-to-wall crack propagation, if the total hydrogen concentration is low and most hydrogen exists as a radial hydride, it is propagated along the radial hydride. Conversely, if the amount of circumferential hydride is large, propagation is prevented along only the radial hydride, and the crack propagates in the circumferential direction, resulting in a lower RHCP value and higher SED.

5. Conclusion

If RHF was more than 5%, RHF alone could explain the mechanical properties of the cladding physically. Since RHF is an area of image processing, it is possible to predict material properties through images. In addition, in the case of RHF 5% or less, additional research is planned to explain the mechanical properties by introducing hydrogen concentration and RHCP. If this is connected with the prediction of RHF and RHCP values according to fuel rod operation history, it will be possible to predict the mechanical properties of the cladding in the period of spent fuel only with reactor data, not images.

NRC suggested 120 MPa as the threshold initiation stress for radial hydride generation, and proposed 90 MPa limit for higher short-term temperature of low burnup fuels [5]. This is a number that simply focuses on whether or not radial hydride has occurred. However, the degradation performance of fuel was different according to the total hydrogen concentration and the radial hydride concentration. Rather, it is conceivable that higher hydrogen concentrations, i.e. higher burnup fuel rods, can tolerate higher stresses, and lower burnup fuel rods require more accurate regulation. If this is also combined th fuel rod operation history, that is, RHF predictive modeling according to rod internal pressure, a more in-depth discussion will be possible.

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