The effect of peak cladding temperature occurring during interim-dry storage on transport-induced cladding embrittlement

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ABSTRACT

To evaluate transport-induced cladding embrittlement after interim-dry storage, ring compression tests were carried out at room temperature (RT) and 135 °C. The ring compression test specimens were prepared by simulating the interim-dry storage conditions that include four peak cladding temperatures of 250, 300, 350 and 400 °C, two tensile hoop stresses of 80 and 100 MPa, two hydrogen contents of 250 and 500 wt.ppm-H and a cooling rate of 0.3 °C/min. Radial hydride fractions of the ring specimens vary depending on those interim-dry storage conditions. The RT compression tests generated lower offset strains than the 135 °C ones. In addition, the RT and 135 °C compression tests indicate that a higher peak cladding temperature, a higher tensile hoop stress and the lower hydrogen content generated a lower offset strain. Based on the embrittlement criterion of 2.0% offset strain, an allowable peak temperature during the interim-dry storage may be proposed to be less than 350 °C under the tensile hoop stress of 80 MPa at the terminal cool-down temperature of 135 °C.

1. Introduction

The PWR nuclear fuel life may be schematically depicted in Fig. 1. Zirconium alloy cladding absorbs hydrogen during reactor operation through a waterside corrosion. It is required in the fuel rod design that the maximum hydrogen content in the cladding should be less than 600 wt.ppm and the maximum rod internal pressure should be less than the reactor coolant system pressure (15.5 MPa). Discharged nuclear fuels are usually stored in a wet storage pit for a few years at temperature less than 60 °C. During the vacuum dry heat-up period for interim-dry storage, existing hydrides in cladding tubes are dissolved, according to temperature-dependent hydrogen solubility. During the interim-dry storage, the spent fuel cladding temperatures may drop to about 120 °C, 60 °C and room temperature, respectively, depending on the storage periods of 20, 40 and 100 years [1]. It should be noted that during the interim dry storage a tensile hoop stress is generated on the spent nuclear fuel claddings due to internal gas-induced over-pressure of the spent nuclear fuel rods. Some dissolved hydrides can be precipitated in the radial direction of the cladding tube during the cool-down process only if a tensile hoop stress on the cladding is larger than a threshold stress [2–7]. The tensile hoop stresses occurring at high burnup fuel rods in PWRs are reported to be between 60 and 80 MPa [8]. It is also reported that hydrides precipitated in the radial direction may severely degrade cladding integrity and mechanical properties [9–13]. Hydride reorientation during interim dry storage may be related to cladding temperatures, hydrogen contents, tensile hoop stresses and cooling rates [14,15]. Regulation guidelines [16,17] of USNRC and Japan Nuclear Regulation Authority (NRA) are summarized in Table 1. As seen in this table, USNRC requires that maximum fuel cladding temperature should not exceed 400 °C for all fuel burn-ups under normal conditions of the interim dry storage, while NRA requires that allowable peak cladding temperatures should be less than 250 and 275 °C, depending on fuel burnup. In general, the ultimate goal of the interim dry storage is to prevent a gross failure of spent nuclear fuel cladding even under abnormal operations including transportation. A compressive load occurring during the spent fuel transportation after the interim dry storage is schematically depicted in Fig. 2. Considering compressive loads on the cladding occurring during the spent nuclear fuel transportation, extensive studies have been conducted on the hydride effect on the embrittlement and mechanical property degradation of zirconium alloy claddings. Previous investigations indicated that ductility can be reduced significantly when the hydride platelet normal is parallel to axis of the tensile direction [4,18,19], whereas when the platelet normal is perpendicular to the stress axis it has relatively little
In addition, the hydride orientation is known to affect the ductile to brittle transition temperature (DBTT) of zirconium alloys. In this study, compression tests were performed using ring specimens having radial hydrides precipitated during cool-down to simulate the impact of the transport-induced compressive loads on cladding embrittlement following the interim dry storage. To generate ring specimens having radial hydrides during cool-down, the interim dry storage conditions include four peak cladding temperatures of 250, 300, 350 and 400 °C, two tensile hoop stresses of 80 and 100 MPa, two hydrogen contents of 250 and 500 wt.ppm-H and a cooling rate of 0.3 °C/min from the four peak cladding temperatures. Then, using the ring specimens having various radial hydrides precipitated under the aforementioned interim-dry storage conditions, compression tests were formed at room temperature (RT) and 135 °C, respectively, simulating the spent nuclear fuel transport conditions. However, it should be noted that the aforementioned test conditions may not simulate exactly the behaviors of spent fuel claddings during the interim-dry storage, considering cladding oxide layer thickness, hydrogen content and hydrogen distribution in the cladding, cool-down rate during the interim dry storage, neutron irradiation, etc. Therefore, the results obtained in this study may be used qualitatively and restrictively to predict the amounts of radial hydrides precipitated, radial hydride-induced embrittlement and subsequently allowable peak cladding temperatures against spent fuel cladding integrity maintenance during transportation following

![Fig. 1. Cladding tube temperature variation during nuclear fuel.](image1)

<table>
<thead>
<tr>
<th>Organization</th>
<th>Peak cladding temperature (°C)</th>
<th>Allowable tensile hoop stress (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>US NRC</td>
<td>≤ 400</td>
<td>≤ 90</td>
</tr>
<tr>
<td>Japan NRA</td>
<td>≤ 275 for less than 48 GWD/MTU ≤ 90</td>
<td>≤ 100</td>
</tr>
<tr>
<td></td>
<td>≤ 250 for less than 55 GWD/MTU ≤ 100</td>
<td>≤ 90</td>
</tr>
</tbody>
</table>

![Fig. 2. A schematic diagram for compressive load occurrence during fuel assembly transportation.](image2)
Stress-relieved Zr–Nb alloy cladding tubes were used in this study. The material composition and dimensions of the Zr–Nb alloy cladding tubes are given in Table 2. Two hydrogen contents of 250 and 500 wt.ppm were selected to make test specimens, considering hydrogen contents of medium and high burnup spent fuels. Prior to hydrogen charging, cladding tubes were cut into 200 mm long pieces and cleaned with acetone and ethanol. The tubes were charged with hydrogen in a vacuum furnace at 400°C containing a mixture gas of hydrogen (0.020 MPa) and helium (0.026 MPa). Hydrogen charged specimens were sealed with quartz tube and heat-treated at 400°C for 24 h to attain a uniform hydrogen distribution [22]. Hydrogen contents were analyzed by an ELTRA ONH-2000. The two hydrogen contents were measured to be in the range of 250 ± 25 and 500 ± 25 wt.ppm, respectively. The hydrogen charged specimens were cut into 5 mm long rings. Fig. 3 shows a schematic diagram of the tensile and compressive tests using the ring specimens. Fig. 4 shows a schematic diagram of heat-up and cool-down processes simulating the interim dry storage, and the ring compression tests simulating the transportation after the interim dry storage.

As shown in Fig. 4, the specimens were heated up at a rate of 2.0°C/min to four respective peak temperatures of 250, 300, 350, and 400°C. After remaining for 2 hrs at those temperatures under no stress condition, the specimens were cooled down at a rate of 0.3°C/min under two tensile hoop stresses of 80 and 100 MPa generated by the upper and lower jigs shown in Fig. 3. It should be noted that a load applied by the upper and lower jigs causes much higher hoop stresses at the inner ring surface and much lower ones at the outer ring surface than a load applied by internal gas pressure. The heat-up and cool-down tests were performed using a KLES 500-Screep tester. It is noteworthy that the four heat-up temperatures and the two tensile hoop stress given in Table 1. It is reported that a slower cooling rate generates more radial hydride precipitation during cool-down [14,15]. The 0.3°C/min cooling rate was used in this study since it is the slowest cooling rate achievable in our experimental system. After cool-down with tensile hoop stresses, some specimens were used for metallography to investigate radial hydride fractions and others for ring compression tests to investigate radial embrittlement behaviors. Some specimens were cut at the A-A section in Fig. 3 and examined using optical microscopy to observe hydride distribution. An etchant used for metallographic examination was composed of HF, H2SO4, HNO3, and H2O at a volume ratio of 10:30:30:30.

The ring specimens used in the compression tests may contain radial hydrides precipitated during the aforementioned cool-down process, as shown in Fig. 4. The ring compression tests were carried out up to 2 mm displacement at RT and 135°C, respectively. As shown in Fig. 3, the cladding tube areas having radial hydrides are positioned at 6 and 12 o’clock directions. Based on load-displacement curves generated by the ring compression tests, offset strains were calculated for the respective specimens. The ring compression tests with a displacement rate of 2 mm/min were performed by Shimadzu Autograph AG-X plus (100 kN).

Table 2
Zirconium alloy cladding tubes used in this work.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Chemical composition (w/o)</th>
<th>Heat treatment</th>
<th>Texture (Kearns number)</th>
<th>Tube dimension (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zr–Nb alloy</td>
<td>Zr–1.0Nb–1.0Sn–0.1Fe–0.12O</td>
<td>Stress-relieved</td>
<td>f_r (radial) = 0.62</td>
<td>Outer dia. = 9.50</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>f_t (tangential) = 0.26</td>
<td>Thickness = 0.57</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>f_a (axial) = 0.12</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 3. Ring specimen configurations for tensile loading and ring compression tests.

Fig. 4. A schematic diagram for heat-up and cool-down simulating interim-dry storage and ring compression test simulating transportation.
3. Results and discussion

3.1. Radial hydride formation during the interim dry storage

Prior to the heat-up and cool-down tests (see Fig. 4), hydrogen-charged specimens were cut in the A-A section in Fig. 3 and the cut section was examined using optical microscope to reveal hydride distribution at room temperature. Fig. 5 [23] shows that most of the hydrides are uniformly distributed in the circumferential direction for the as-received 250 and 500 wt.ppm-H specimens.

As shown in Fig. 4, the as-received specimens were heated up to four respective peak temperatures, maintained for 2 h under zero stress condition at those peak temperatures for 2 h. Then, the specimens were cooled down at a rate of 0.3 °C/min to room temperature under two tensile hoop stresses of 80 and 100 MPa. These tensile hoop stresses represent average tensile hoop stresses on the A-A gage section shown in Fig. 3. The average tensile hoop stress was calculated by dividing a tensile load applied on the inner ring surface by the cross-sectional area of the gage section. After cool-down, the specimens were cut at the gage region in the radial direction and examined using an optical microscope to observe hydride distribution. The optical micrographs of the 250 and 500 wt.ppm-H specimens are shown in Fig. 6. This figure shows that the relatively larger amounts of radial hydrides are formed at the inner region. In order to explain this phenomenon, finite element calculations were performed to assess radial-dependent hoop stress levels at the gage sections caused by the upper and lower jigs (see Fig. 3). Fig. 7 shows tensile hoop stress variations along the cladding thickness caused by an average tensile hoop stress of 80 MPa. This figure indicates that the jig geometry generates a much higher stress gradient than the internal gas pressure. For the jig geometry, the finite element calculations indicate that the inner region generates a much higher hoop stress than the outer region. With an average tensile hoop stress of 80 MPa, the highest hoop stress of 122 MPa occurs at inner ring surface, while the lowest hoop stress of 40 MPa at outer ring surface. This is why the relatively larger amounts of radial hydrides are formed at the inner region.

![Fig. 6. Precipitated radial hydride during cool-down from four peak heat-up temperatures to room temperature as a function of tensile hoop stress and hydrogen content.](image)

<table>
<thead>
<tr>
<th>Hydrogen content (ppm)</th>
<th>Tensile hoop stress (MPa)</th>
<th>Peak cladding temperatures</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>250 °C</td>
<td>300 °C</td>
</tr>
<tr>
<td>250</td>
<td>80</td>
<td></td>
</tr>
<tr>
<td></td>
<td>100</td>
<td></td>
</tr>
<tr>
<td>500</td>
<td>80</td>
<td></td>
</tr>
<tr>
<td></td>
<td>100</td>
<td></td>
</tr>
</tbody>
</table>
From Fig. 6, radial hydride fractions were calculated as follows. Circumferential hydride is defined as the plates with the orientation within $0^\circ$ to $40^\circ$ to the circumferential axis, whereas radial hydride with the orientation within $50^\circ$ to $90^\circ$ to the circumferential axis was not counted as radial or circumferential hydrides. It is noteworthy that the uncertainty in the radial hydride fractions obtained in this study is less than 5% of the average values, which was estimated from the radial hydride fractions of the six portions of the optical micrographs at the inner regions of specimens. The radial hydride fractions shown in Figs. 8 and 9 were determined using the 500 magnified optical micrographs. From these figures, it can be seen that a higher peak temperature and a larger tensile hoop stress generated a larger radial hydride fraction, while the 250 wt.ppm-H specimens generated relatively a larger radial hydride fraction than did the 500 wt.ppm-H specimens. It is reported that radial hydrides will be precipitated during cool-down if a hoop stress on the cladding tube is larger than a critical stress $[2^e7]$. This indicates that even the lower tensile hoop stress of 80 MPa used in this study is larger than a critical stress for radial hydride precipitation.

The peak temperature-dependent radial hydride precipitation generated during cool-down may be explained by the terminal solid solubility for dissolution $[24]$. As explained by H. Cha et al. $[23]$, the amounts of hydrides precipitated during cool-down rather depend on TSSD (Terminal Solid Solubility for Dissolution) graph (see the lower graph in Fig. 10), while the TSSP (Terminal Solid Solubility for Precipitation) graph (see the upper graph in Fig. 10) provides the temperature at which an incipient precipitation will start during cool-down. As an example, when the specimens were cooled down from 400 °C (H1) to RT (C1), the dissolved hydrogen atoms at 400 °C did not form hydrides until about 340 °C (see the line from H1 to C0). It is noted that the hydrogen solubilities at the peak temperatures of 250, 300, 350 and 400 °C are 45, 80, 150 and 230 wt.ppm, respectively, and the hydrogen solubility at RT is about 10 wt.ppm. When the specimens were cooled down from the respective heat-up temperatures of 250(H4), 300(H3), 350(H2) and 400 °C(H1) to RT(C1), the respective amounts of hydrogen atoms precipitated may be 35, 70, 140 and 220 wt.ppm, indicating that a higher peak temperature will generate a larger amount of hydride precipitation during the cool-down process.

The difference in the radial hydride fractions between the 250 and 500 wt.ppm-H specimens shown in Figs. 8 and 9 may be explained by the difference in the remaining circumferential hydride fraction. For example, the hydrogen solubilities at 400 °C are 230 wt.ppm. The remaining circumferential fraction of the 250ppm-H specimens at 400 °C is negligible but that of the 500 wt.ppm-H specimens is about 50%. This considerable remaining circumferential hydrides of the 500 wt.ppm-H specimens may be partly used as circumferential hydride nucleation sites for the dissolved hydrogen atoms during the cool-down process and subsequently generate relatively larger circumferential hydride precipitation than the 250 wt.ppm-H specimens. On the other hand, the undissolved circumferential hydrides may block the growth of radial hydrides precipitated, resulting in smaller radial hydrides for the 500 wt.ppm-H specimens. Consistent with the results of many researchers $[8,14,15,23,25]$, the radial hydride fraction increases with decreasing hydrogen content for the same peak temperature and hoop stress.
3.2. Ring compression tests using the specimens with cool-down-induced radial hydrides

During the spent nuclear fuel transportation after the interim dry storage, the fuel may be susceptible to vibration or shock that will cause a compressive load on the fuel rods, as shown in Fig. 2. To evaluate the effect of the compressive load on the fuel rod failure, the ring compression tests were carried out at RT and 135 °C, respectively, using the ring specimens with radial hydrides (see Fig. 6). The load-displacement curves obtained from the ring compression tests are given in Figs. 11 and 12. The offset strains were calculated by the ratio of offset displacement (d) and ring outer diameter (9.50 mm). As shown in Fig. 13, the offset displacements were calculated by the dotted straight lines parallel to the solid line representing the elastic region (see Fig. 13-a). The first load drop shown in Figs. 11 and 12 may indicate an onset of crack initiation and propagation. The successive load drops in these figures may be caused by a zig-zag propagation of the crack. As shown in Fig. 14, the cracks were initiated at 6 and 12 o’clock directions during the ring compression tests. Kim et al.’s finite element analysis [25] showed that the maximum tensile bending stress occurs at the inner cladding surface located at 6 and 12 o’clock directions. A detailed crack initiation and propagation behavior at those directions is shown in Fig. 14. From this figure, it can be seen that a crack formed propagates along the radial hydride as the primary crack and then grows along the circumferential hydride as the secondary crack if the crack meets it, which generates a zig-zag propagation, e.g., propagation along radial hydride-circumferential.

![Ring compression tests using the specimens with cool-down-induced radial hydrides](image)

**Fig. 10.** Terminal solid solubility of hydrogen in the Zircaloy-4 claddings.

**Fig. 11.** Ring compression load-displacement curves at room temperature for the specimens experiencing the respective tensile hoop stresses of 80 and 100 MPa during the cool-down process.
hydride-radial hydride, generating the successive load drops in the load-displacement curves mentioned above.

The calculated offset strains are depicted in Figs. 15 and 16. In these figures, the ductile-to-brittle transition offset strain is given as 2.0%, which is proposed by Billone [8]. The load-displacement curves given in Figs. 11 and 12 show that no load drops occurred at the peak temperature of 250 °C, regardless of tensile hoop stress, indicating that no crack was initiated. Therefore, the actual offset strains at the peak temperature of 250 °C given in Figs. 15 and 16 are larger than the offset strains given in these figures. The offset strains for the 500 wt.ppm-H specimens are larger than those for the 250 wt.ppm-H ones since the radial hydride fractions of the former is less than those of the latter, as seen in Figs. 8 and 9. Similarly, the lower peak temperature also generates a larger offset strain due to the lower radial fraction. As expected, the offset strains measured at 135 °C are larger than those at RT. The offset strains at the peak temperature of 250 and 300 °C are much larger than the ductile-to-brittle transition offset strain of 2.0%, regardless of tensile hoop stress, hydrogen content and the ring compression test temperature.

For the ring compression test temperature of RT, the offset strains at the peak temperature of 300 °C are larger than 2.0% for the 500 wt.ppm-H specimens, while those are less than 2.0% for the 250 wt.ppm-H specimens. The offset strains at the peak temperature of 350 and 400 °C are less than 2.0% for all the specimens.

For the ring compression test temperature of 135 °C, the offset strains at the peak temperature of 250 and 300 °C are larger than...
2.0% for all the specimens. The offset strains at the peak temperature of 350 °C are less than 2.0% for the 250 wt.ppm-H and 100 MPa specimens, while those at the peak temperature of 400 °C are less than 2.0% for the 250 wt.ppm-H specimens.

It should be noted that both this paper and previous papers [15,25] used the tube specimens with various amounts of radial hydrides formed during cool-down under tensile hoop stresses. To investigate the effect of radial hydrides on the spent nuclear fuel embrittlement, the previous paper employed tensile loads on the inner ring surfaces of the specimens with radial hydrides, while this paper employed transport-induced compressive loads on the outer ring surfaces of the specimens with radial hydrides. However, it is found that there is a similar tendency in the impact of the radial hydride fractions on the cladding embrittlement, regardless of load directions, tensile or compressive load. It is important to derive allowable compressive loads or offset strains to maintain spent fuel cladding integrity during spent fuel transportation following the interim-dry storage. Therefore, the ring compression tests used in this work can produce direct results on transport-induced cladding embrittlement following the interim dry storage.

3.3. Estimation of allowable cladding peak temperatures against cladding integrity

Allowable cladding peak temperatures for maintaining cladding integrity can be estimated as follows. Assuming that the maximum tensile hoop stress on the cladding is 80 MPa, the respective allowable cladding peak temperatures for the terminal cool-down temperatures of RT and 135 °C may be estimated to be 250 and 350 °C, as seen in Figs. 15 and 16. On the other hand, assuming that the maximum tensile hoop stress on the cladding is 100 MPa, the respective allowable cladding peak temperatures for terminal cool-down temperatures of RT and 135 °C may be estimated to be 250 and 300 °C, as seen in Figs. 15 and 16.

It should be noted that allowable cladding temperatures for hoop stresses of 80 or 100 MPa provided below are only valid for hydrogen contents ranging between 250 and 500 wt.ppm-H.

Based on the ring compression test results mentioned above, a further detailed investigation on the allowable cladding peak temperatures during the spent nuclear fuel interim-dry storage and subsequent transportation is needed to verify the peak temperatures.
4. Conclusions

The effects of hydrogen contents, peak cladding temperatures and tensile hoop stresses prevailing at interim-dry storage spent nuclear fuel on the embrittlement of zirconium alloy cladding during transportation are investigated. The principal results obtained in this study may be summarized as follows:

- Higher peak cladding temperature and larger tensile hoop stress generate larger radial hydride fractions.
- Radial hydride fractions of 500 wt.ppm-H specimens are less than those of 250 wt.ppm-H ones. This is because relatively larger undissolved circumferential hydrides at the 500 wt.ppm-H specimens may be used as circumferential hydride precipitation sites on the one hand and may block the growth of radial hydrides precipitated on the other hand.
- Radial hydride fractions can explain the behaviors of offset strains measured by ring compression tests. A higher peak temperature, a larger tensile hoop stress and 250 wt.ppm-H generate a lower offset strain by a larger radial hydride fraction.
- If the maximum tensile hoop stress on the cladding is assumed to be 80 MPa, the respective allowable cladding peak temperatures may be estimated to be 250 and 350 °C for the terminal cool-down temperatures of RT and 135 °C during the interim dry storage. If the maximum tensile hoop stress on the cladding is assumed to be 100 MPa, the respective allowable cladding peak temperatures may be estimated to be 250 and 300 °C for the terminal cool-down temperatures of RT and 135 °C.
- Allowable cladding temperatures for hoop stresses of 80 or 100 MPa provided above are only valid for hydrogen contents ranging between 250 and 500 wt.ppm-H.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was supported by the Dongguk University Research Fund of 2019, Nuclear Energy Development Program (NO. 2017M2B2A9A02047) through the National Research Foundation (NRF) funded by the Ministry of Science and ICT as well as by the Global NPP Decommissioning Experts Cultivation Project funded by the Province of Gyoengsangbuk-do.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.net.2019.12.030.

References