

A Review of Delayed Hydride Cracking of Spent Fuel Rods in Dry Storage

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1. Introduction

Dry storage of spent fuel rods is being considered as an alternative to the interim storage method until a final decision on their final disposal is made [1]. Most studies on the integrity of spent fuel rods in dry storage have focused on cladding creep, which is known to be the most likely failure mechanism [2], and reorientation of hydrides to secure their retrievability after dry storage [3]. Little attention has been paid to delayed hydride cracking (DHC) because this phenomenon will not occur due to limited stresses and slow diffusion of hydrogen at low temperatures below 200 °C [4]. However, Simpson and Ells reported a failure of unirradiated Zr-2.5Nb fuel rods after their long-term storage at room temperature, the cause of which was recognized to be DHC [5]. Therefore, it is clear that in contrast to what is believed to be that no DHC occurs in the spent fuel rods in dry storage, they would fail as long as stress raisers such as surface flaws or the weld region are present inside the cladding tube. Especially, high burnup fuel rods may have incipient cracks on the inside cladding surface due to an interaction of the fuel and the cladding during reactor operation [4]. Three instances of spent fuel rod failures were also reported where a leakage occurred less than 2 months after the spent fuel rods were stored at rather low temperatures below 275 °C [2,6]. Although the cause of their failures is unknown, these failures have shown that the incipient cracks present inside them grow fast to through-wall cracks in the spent fuel rods even at low temperatures, because the leakage has occurred within 2 months after the start of their dry storage. Thus, we suggest that these failures of the spent fuel rods are due to DHC, the rationale of which is given in this study. The aim of this study is to demonstrate the DHC susceptibility of spent fuel rods by citing Simpson and Ells's results [5] and to provide a rationale for the failures of spent fuel rods due to DHC even at room temperature using our new DHC model [7-9], which remains unclear to date.

2. DHC Susceptibility of Spent Fuel Rods

Simpson and Ells [5] showed that for a Zr-2.5Nb cladding tube containing only 10 ppm of hydrogen the hydrides were precipitated at room temperature at the root of the end cap weld 4 weeks after welding and then a crack grew there to about 90% of the cladding wall thickness (equal to 0.41 mm) 2 years after that. A simple calculation assuming the growth of a crack to be 0.9 times the thickness over 2 years shows that the

crack growth rate was at the least 5.9×10^{-12} m/s, which is over 10^4 times faster than the predicted crack growth rate, 1.6×10^{-16} m/s [5], by the old DHC models. Then, they conducted another verification test using the cladding tube with 100 ppm H where a faster growing crack with time was also observed at the same place or the root of the end cap weld [5]. When a Vickers microhardness impression was made in a Zr-2.5Nb alloy sheet charged to 100 to 300 ppm H, the hydrides were seen after 6 months at each of the corners of the microhardness impression that did not exist a few minutes after that impression, causing a crack growth there by their cracking [5]. Given that the microhardness impression tests had been conducted at room temperature, it is obvious that DHC occurs in zirconium alloy with hydrogen even at room temperature only if a stress raiser is present, which cannot be satisfactorily understood by the old DHC models.

Precipitation of the hydrides and subsequent crack growth by their cracking in cladding tubes of zirconium alloys even at room temperature as described above is similar to the cases where DHC occurs at 250 °C for the water-quenched Zr-2.5Nb tube when approached by heating [9] or initiates near the TSSD temperature with little undercooling when cooled from 50 °C above the TSSD. A driving force for cracking of zirconium alloy cladding tubes at room temperature is the ΔC caused by the δ - to γ -hydride phase transformation, corresponding to the distance AA' or BB' shown in Fig. 1: the bulk region containing the δ - and γ -hydrides with a higher hydrogen solubility and the weld region containing δ -hydrides only with a lower hydrogen solubility. Considering that the γ hydrides are precipitated at low temperatures below 182 °C for the slow cooled zirconium alloys such as the spent fuel rods [10], it is not until the spent fuel rods are cooled to below 180 °C that DHC occurs due to stress-induced hydride phase transformation from γ to δ . We can estimate when the cladding temperatures of the spent fuel rods become lower than 180 °C using Einziger's method for predicting temperature of a fuel assembly [11]. Fig. 4 shows an example of the fuel assembly temperature predicted by Einziger's method. Assuming that the cladding temperature is conservatively similar to the average temperature of the fuel assembly, it takes at least 30 years for the cladding temperatures to completely fall to below 180 °C (Fig. 8). This estimation is in line with Sasahara's prediction [12] that more than 30 years of the cooling time is needed to cool

the cladding temperatures of spent fuel rods to below 180 °C. In other words, even though a visual

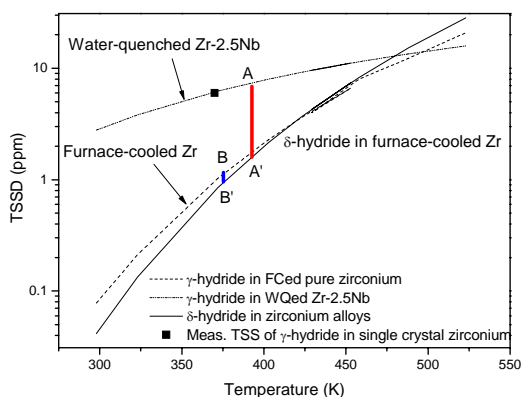


Fig. 2. Measured terminal solid solubility of the γ -hydride in the furnace-cooled zirconium and in the water-quenched Zr-2.5Nb alloy and of the δ -hydride determined from Kearns data along with the measured TSS of the γ -hydride in water-quenched zirconium single crystal.

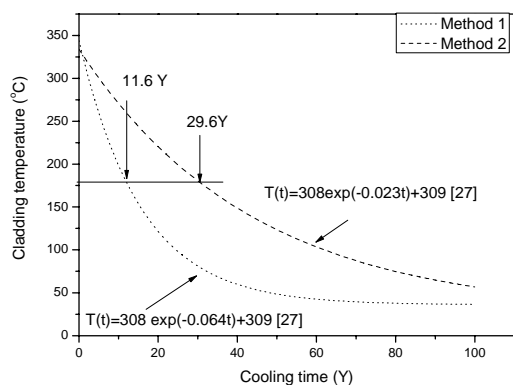


Fig. 3. Estimated temperatures of a fuel assembly during dry storage period depending on the methods to develop a temperature profile for calculating creep deformation for the storage period.

inspection of the spent fuel rods after dry storage of 15 to 20 years showed little evidence of DHC [11,12], it is too early to draw concrete conclusions about the DHC effect on the integrity of spent fuel rods. It is not until the storage time exceeds 30 years that DHC is activated, likely causing a leakage from the spent fuel rods. Other evidence for this hypothesis is provided by the failures of the spent fuel rods that had been stored at relatively low temperatures below 275 °C [6].

4. Conclusions

Failures of unirradiated Zr-2.5Nb cladding tubes occurred even at room temperature due to cracking of the hydrides precipitated at stress raisers such as the weld region. These facts demonstrate that DHC occurs even at room temperature in zirconium alloys with hydrogen, only if stress raisers are present. Consequently, it is concluded that the spent fuel rods to be stored in dry storage would fail due to this low

temperature DHC only if they have stress raisers inside the cladding surface. Given that this phenomenon occurs only after cooling the cladding temperatures of the spent fuel rods to below the γ - to δ -hydride phase transformation temperature, it is not until 30 years have passed that failures of the spent fuel rods by DHC would occur. The driving force for this low temperature DHC is a difference in hydrogen concentration between the bulk region and the crack tip arising from stress-induced hydride phase transformation from γ to δ , which is evidenced by a higher hydrogen solubility of the γ -hydrides when compared to that of the δ -hydrides. Further works are recommended to establish DHC failure criterion for the spent fuel rods that are being stored in dry storage.

ACKNOWLEDGEMENTS

This work has been carried out as part of the Nuclear R&D program supported by Ministry of Education, Science and Technology, Korea.

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