A New Perspective of Hydride Re-orientation in SNF

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1. Introduction

In recent years, hydride re-orientation in spent nuclear fuels (SNF) and its effects on cladding properties have become important considerations in storage, handling and transportation of SNFs. In particular, current high-burnup fuels would have more severe potential for damaging re-orientation of hydrides. The mechanism of hydride re-orientation involves dissolution of circumferential hydrides and formation of radial hydrides. Past studies have already illustrated the mechanism of hydride re-orientation and the factors that affect it, including stress, hydrogen concentration, temperature, cooling rate etc. [1~6]. In this paper, we review those studies and suggest a new perspective for additional research on hydride re-orientation.

2. Studies on Hydride Re-orientation in SNF

2.1 General

Most studies to-date on hydride re-orientation have focused on hydride re-orientation during stylized conditions pertinent to dry storage and to transportation of SNFs. Based on results from those studies [1], the U.S. Nuclear Regulatory Commission (NRC) has set some design limits to inhibit hydride re-orientation as follows [2]:

- For all fuel burnups (low and high), the maximum fuel cladding temperature should not exceed 400˚C. However, for low burnup fuel, a higher short-term temperature limit may be used when cladding hoop stress is equal to or less than 90 MPa for the temperature limit proposed.
- During loading operations, repeated thermal cycling should be limited to less than 10 cycles, with cladding temperature variations less than 65˚C each.

As can be seen in the NRC requirements, temperature and stress are recognized as the most important factors affecting hydride re-orientation in SNF cladding. Many investigations have already quantified the impacts of these two parameters on radial hydride re-orientation. Also, the effects of thermal cycles and of pre-heating on hydride re-orientation have also been investigated [3, 4].

2.2 Formation of Hydrides

Oxidation of Zircaloy cladding during reactor operation produces hydrogen. Some portion, estimated to be 5% to 20%, of the hydrogen released by this reaction is absorbed by the cladding and diffuses into the clad [4]. The absorbed hydrogen may remain in solid solution in the Zr lattice or precipitate as hydride depending on the temperature and the amount of hydrogen absorbed. When hydrogen concentration exceeds the solubility limit of hydrogen, hydrides form. The solubility of hydrogen decreases drastically with decreasing temperature in zirconium alloys. Preferred sites for hydride formation are either grain boundaries or inside the matrix, in which basal plane (0002) is the dominant habit plane [4]. Since the density of the hydride is less than that of zirconium, hydride nucleation leads to a local lattice expansion, which should be accommodated in the zirconium matrix [5].

Hydride precipitates can be seen with an optical microscope. They appear as single platelets or as a cluster of platelets, as shown in Fig. 1[6].

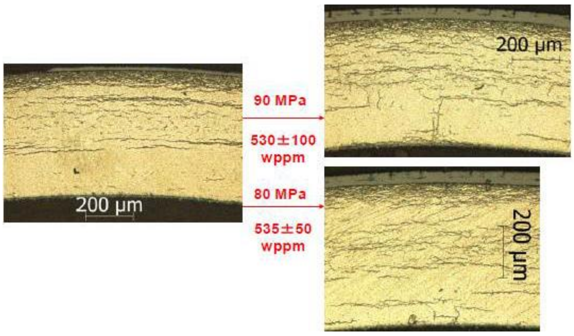

Fig. 1. Hydride Re-orientation in Irradiated ZIRLO Cladding Heated to 673˚K and Cooled under Stress[6].

2.3 Factors affecting hydride re-orientation

Threshold Stress

Radial hydride formation is believed to be driven by tensile hoop stress (along the circumferential direction). As hydrides form perpendicular to the tensile stress and parallel to the compressive stress, stress plays a major role in hydride re-orientation. Therefore, a critical stress also known as the threshold stress, under which the re-orientation takes place, has been defined. It is clear that the stress limits for hydride re-orientation appear to decrease with an increase of peak temperature.

NRC's temperature and stress limits were determined to prevent hydride re-orientation from occurring[2]. However, further research has shown that hydrides may still reorient radially even within these limits as seen in Fig. 2 [1].

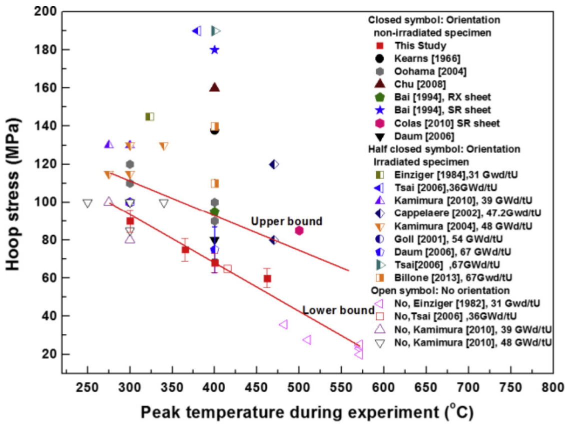

Fig. 2. Collected data on threshold stress triggering hydride reorientation in Zircaloy-4 as a function of peak temperature[1].

Temperature

As the level of re-oriented hydrides depends on the amount of dissolved hydrogen at the peak temperature of the cladding, the temperature history experienced by the cladding is of interest for hydride re-orientation in SNF. The re-orientation process is of particular concern during the drying process right after fuel is removed from the pool, at which time the cladding temperature is assumed to reach around 400˚C. This would cause a significant portion of hydrogen, about 200 wppm in Zircaloy, to dissolve back into solution. Hydrogen dissolved upon heating is available to re-precipitate as radial hydrides upon subsequent cooling. The temperature affects internal pressure of SNF: the internal pressure increases with increasing temperature. Considering the measured peak temperatures of SNF, which ranges over 400°C, the temperature of cladding should be monitored closely, especially during drying process [4].

Thermal Cycle

The percentage of radial hydrides increases as the number of thermal cycles increases until it saturates. Hence, it is believed that the effect of thermal cycling on hydride re-orientation is more significant than that of isothermal treatment [3, 4].

3. A New Perspective

Simulating the realistic conditions of SNF

Hoop Stress

Studies performed so far mostly used constant hoop stresses by using ring compression, ring tension, tapered specimen or internal gas pressure. In reality, the hoop stress decreases continually during the storage period, due to drop in temperature and cladding creep.

Temperature

Cladding temperature also decreases over time. Thus hydride re-orientation is better assessed using more realistic cladding temperatures calculated from decay heat and conduction.

Hydride Distribution

Non-irradiated cladding tested for hydride re-orientation often contained relatively uniform hydride distributions compared to the non-uniform distributions generally found in irradiated claddings. This potential difference in hydride distribution and morphologies over the cross section could be one reason for the discrepancy in test results between un-irradiated alloys and irradiated alloys.

Stress State

Stress state also impacts the threshold stress for hydride reorientation. Most results of threshold stresses for hydride re-orientation have been obtained under nearly uniaxial test conditions, thus, may not be directly applicable to re-orientation in a closed fuel rod, in which stress state would be multi-axial.

CANDU Fuel

Some differences between CANDU and PWR fuels impact Delayed Hydride Cracking (DHC) in the former. As illustrative examples: (a) Stresses near the junction between the sheath and the endcap are highly multi-axial; also much higher than in the clad; therefore hydride assessments at this location need quite detailed considerations of highly multi-axial stresses; (b) Endplates have the lowest temperature in a CANDU fuel assembly, therefore highest concentration of hydrides usually occurs in endplates and in assembly welds; (c) Variety of microstructures occur in CANDU fuel's Zircaloy. Thus DHC characteristics need to be established and analyzed for CANDU fuel differently from PWR fuels.

4. Conclusion

Considering the gaps between the conditions of laboratory hydriding tests and realistic conditions of SNF currently in dry storage, more realistic test conditions would be desirable to better understand the behaviors of hydride re-orientation in SNF.

Therefore further research is needed to better quantify the relationship between hydride re-orientation and ductility; this will improve the hydride criterion. Also, to establish the requirements for CANDU fuel, additional research is needed to reflect the several unique aspects of that fuel.

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