



## Delayed hydride cracking assessment of PWR spent fuel during dry storage



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### HIGHLIGHTS

- The threshold SIF ( $K_{IH}$ ) as the assessment criterion of DHC is described.
- DHC evaluation was performed for PWR spent fuel during long-term dry storage.
- DHC does not largely contribute to cladding failure except in limiting cases.
- More information on spent fuel is required for a more accurate and reliable analysis.

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### ABSTRACT

One of the possible degradation mechanisms of PWR spent fuel cladding is delayed hydride cracking (DHC), which can activate within a relatively low temperature range. Due to the high crack growth rate, it is essential to prevent DHC crack growth, but most of the existing DHC studies have focused on the crack growth rate. In this regard, the values and characteristics of the threshold stress intensity factor (SIF) for PWR Zircaloy-4 cladding were described as assessment criteria of DHC. Based on the limited existing test results and analyses, threshold SIF and applied SIF were evaluated. In addition, the possibilities of degradation by DHC were discussed during long-term dry storage. From the analysis with conservative approaches, DHC does not largely contribute to cladding failure. For a more reliable analysis, additional information on spent fuel is needed.

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### 1. Introduction

In the operating PWRs, tens of millions of spent fuel rods have been discharged and will be discharged. The fraction of high burnup (>45 GWd/MTU) PWR fuel that has high rod internal pressures (RIP) and hydrogen contents with thick oxide significantly increased after 2000. After being discharged and stored in a wet storage (pool storage) for years, spent fuel is generally stored in the form of dry storage in a helium atmosphere after a vacuum drying process until disposal. During dry storage, the spent fuel claddings could degrade through degradation mechanisms such as oxidation, stress corrosion cracking, creep, hydride reorientation, and delayed hydride cracking (DHC). Among these, one of the possible failure mechanisms of PWR spent fuel cladding is DHC, which is a time dependent crack growth process resulting from stress assisted hydrogen diffusion to the crack tip (Zielinski and Sobieszczyk, 2011; U.S. Nuclear Regulatory Commission,

2014; Hanson et al., 2012; Rashid et al., 2000; Coleman, 2011; Tseng et al., 2014; Chao et al., 2008). Originally, DHC was considered one of the failure modes of CANDU pressure tubes, but DHC would be possible for Zr-alloy fuel cladding, triggered by the combination of stress through internal pressure of the fuel rod and accumulated hydrogen. In particular, DHC can be activated within a relatively low temperature range, in contrast to creep behavior, although cladding hoop stress also decreases as a consequence of decreasing temperature. Therefore, it is essential to assure the long-term integrity. In this regard, NRC (U.S. Nuclear Regulatory Commission, 2014), DOE (Hanson et al., 2012), and EPRI (Rashid et al., 2000; Coleman, 2011) also recognized that DHC is a potential cladding breach mechanism after ~100 years of dry storage, although regulatory guides have not yet been indicated. Meanwhile, relevant studies have been performed by an International Atomic Energy Agency (IAEA) coordinated research program (CRP) Coleman et al., 2009, 2010, 2014; IAEA, 2010; IAEA, 2015. However, most of the DHC data were obtained from crack growth rate (CGR) tests, which are irrelevant to a DHC integrity evaluation (Coleman et al., 2009, 2010; IAEA, 2010).

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In this paper, the threshold stress intensity factor, as the assessment criterion of DHC, is described. In an effort to assess the integrity of spent fuel during long-term dry storage, the application of threshold stress intensity factor to a PWR fuel evaluation was performed with the use of practical and reliable input parameters. In addition, the results were compared with estimations of existing studies.

**2. DHC assessment**

**2.1. Threshold stress intensity factor**

In spite of scatter in the published data, the maximum CGR by the DHC phenomenon is estimated to be within the range of  $10^{-7}$  m/s (Coleman, 2011; Coleman et al., 2009, 2010; IAEA, 2010) at which the cladding can rupture within a year at high temperature during the early stage of the dry storage period. Thus, to maintain the integrity of cladding during long-term storage, it is essential to suppress the crack growth of DHC. In this regard, the threshold stress intensity factor (SIF),  $K_{IH}$ , is generally used for a DHC evaluation in which a DHC crack grows when the applied SIF ( $K_I$ ) at the crack tip is larger than  $K_{IH}$ .

Despite a great deal of effort (IAEA, 2015; Coleman et al., 2014; CRWMS M&O, 2000a; Chan, 2013; Markelov et al., 2015; Pandey et al., 2010), it remains difficult to predict  $K_{IH}$  of PWR spent fuel cladding due to limited data. Most of the existing data of  $K_{IH}$  are limited to Zr-2.5Nb for the PHWR pressure tube, but only a small amount of data for BWR cladding (Zircaloy-2) and PWR cladding (Zircaloy-4) is available. It was reported that the  $K_{IH}$  value for Zr-2.5Nb ranges from 4.3 to 10 MPa√m and the value for Zircaloy-2 tubes ranges from 5.2 to 14.2 MPa√m (Chan, 2013). Although some experimental data for Zircaloy-4 fuel cladding exist, the experimentally measured data are widely scattered with large uncertainties and variations as well as different loading directions (IAEA, 2015; Coleman et al., 2014; CRWMS M&O, 2000a; Chan, 2013; Markelov et al., 2015). Also, could the data do not reflect the temperature effects or the detrimental effects of hydrogen content and neutron irradiation. In addition,  $K_{IH}$  values of Zircaloy-4 cladding are limited to data acquired at high temperature above 227 °C, although they are not very sensitive at low temperatures below 250 °C. Therefore, theoretical estimations from fracture mechanics approaches were conducted through Eqs. (1) and (2), as proposed by Shi and Puls (1994a,b), Kim et al. (2000) instead of using experimental data. Material properties of Zircaloy-4 accounting for spent fuel cladding were used as described in Table 1.

**Table 1**  
Parameters used in the estimation of  $K_{IH}$ .

Parameters	Values	Remarks
Young's modulus (E, MPa)	$95900 - 57.4[T(K) - 273]$	Matsuo (1987)
Yield strength ( $\sigma_y$ , MPa)	$584.89 - 0.578T(^{\circ}C)$	Correlation from IAEA (2010) (unirradiated)
Poisson's ratio ( $\nu$ )	$0.436 - 4.8 \times 10^{-4}[T(K) - 300]$	Shi and Puls (1994a)
Fracture strength of hydrides ( $\sigma_y^h$ , MPa)	710	Kubo et al. (2013)
Hydride thickness (t, m)	$0.1 - 2.0 \times 10^{-6}$	Shi and Puls (1994a), Kim et al. (2000), Kubo et al. (2013)
Stress free strain normal to hydride plate ( $\epsilon_{\perp}$ )	0.054	Shi and Puls (1994a)

$$K_{IH}^2 = \frac{E^2 \epsilon_{\perp} t}{8\pi(1 - \nu^2)^2 \left( \frac{1}{1-2\nu} - \frac{\sigma_y^h}{\sigma_y} \right)} \tag{1}$$

$$K_{IH}^2 = \frac{E\sigma_y\epsilon_{\perp}t}{2\kappa(1-\nu^2)(1-2\nu)^2 \left( \frac{1+\kappa}{1-2\nu} - \frac{\sigma_y^h}{\sigma_y} \right)}$$

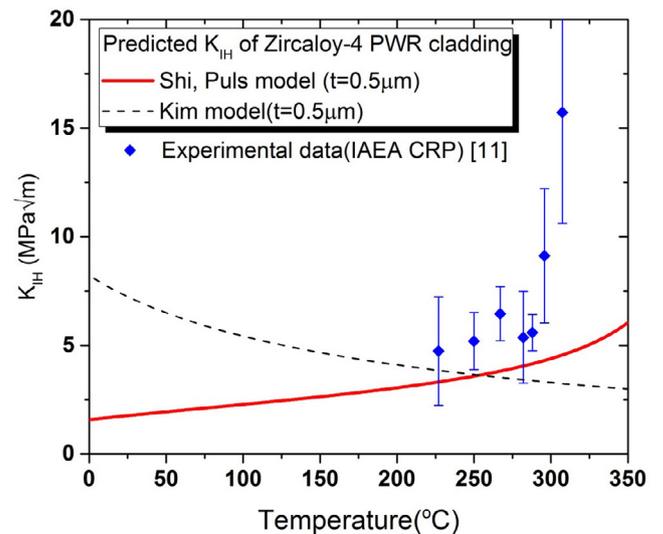
where  $\kappa = \left[ \frac{4}{\pi} \frac{(1-\nu^2)(1-2\nu)K_{IH}^2}{\sigma_y^2 t} - 1 \right] \frac{E}{E}$  (plane strain condition) (2)

The estimation by using Eq. (2) was numerically analyzed by the false position method. Fig. 1 shows the comparative results of the experimentally measured  $K_{IH}$  and the theoretically predicted model value by two groups. Although both models give underestimated results relative to the measured data, only Eq. (1) shows the same tendency as the experimental data that the  $K_{IH}$  increases as the temperature increases. Thus Eq. (1) is used for validating the theoretical estimation. Meanwhile,  $K_{IH}$  is highly dependent on the hydride thickness (t), as shown in Fig. 2. If 2 μm hydride thickness is assumed in the proposed model (Shi and Puls, 1994a; Kim et al., 2000), the predicted value shows good agreement with average values of measured data. However, in this study, a prediction model with the assumption of 0.5 μm hydride thickness was employed for conservatism, because it could give lower-bound values of measured data. In addition, the observed hydride thickness is in the range of 0.1–0.5 μm (Kubo et al., 2013).

**2.2. DHC assessment procedure**

The overall flow of the proposed DHC assessment procedure is shown in Fig. 3. The DHC assessment procedure consists of several modules to calculate or simulate each parameter. First, the rod internal pressure (RIP) is calculated considering the fuel burn-up and hoop stress is then subject to temperature and RIP. The applied SIF ( $K_I$ ) is dependent on the hoop stress and the initial crack morphology of the cladding. Meanwhile, the threshold stress intensity factor ( $K_{IH}$ ) is estimated by material properties and temperature, as mentioned in the previous section.

A comparison with  $K_I$  and  $K_{IH}$  was applied to determine whether the crack on the cladding grows. For the case where the  $K_I$  is smaller than  $K_{IH}$ , the cladding does not fail. In the case where the  $K_I$  is larger than the threshold value ( $K_{IH}$ ), the initial crack grows at the rates calculated in the crack growth module. The



**Fig. 1.** Comparative values of  $K_{IH}$  by prediction models and experimental results for Zircaloy-4.

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