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RADIAL HYDRIDE PRECIPITATION IN FUEL CLADDING DURING BACK-END COOLING TRANSIENT UNDER DECREASING PRESSURE

J. Desquines¹, C. Sartoris¹, M. Guémas¹, and A. Gérard²

¹ IRSN, Cadarache, France

² IRSN, Les Angles, France

jean.desquines@irsn.fr, christine.sartoris@irsn.fr, marine.guemas@irsn.fr, alain.gerard@irsn.fr

Abstract. During spent fuel assembly transportation or dry cask storage, filling gas and fission gas release pressurize the fuel rods contained in the cask. These rods are heated by fuel assembly residual power resulting in internal pressurization stress increase in the rods. During the subsequent cooling associated to removal of rods from the transportation cask or during the decrease of residual power after a long storage period, the cladding hoop stress is expected to decrease. Concomitant cladding temperature and stress decrease may influence the morphology of precipitated hydrides in corroded claddings. Radial hydride precipitation has a detrimental role on the mechanical resistance of the cladding and is promoted by large hoop stresses during the cooling of the cladding. There is, consequently, a need to predict the radial hydride precipitation in corroded cladding tubes during dry storage or transportation cooling transients. Existing models usually address the radial hydride precipitation conditions under constant stress leading to over-estimated radial hydride content within the cladding.

Keywords: fuel transportation, back-end, radial hydrides, Zircaloy-4.

1 Introduction

In the present study a model describing the radial hydride precipitation in a pre-hydrated unirradiated (SRA Zircaloy-4) cladding alloy under time varying stress and temperature transient. The model is first described and, afterwards validated using pressurized tests with constant depressurization rate during the cool down at 24°C/h. This model is implemented in IRSN SHOWBIZ [1] simulation code including back-end cycle modeling capabilities.

2 Modeling the influence of decreasing stress on radial hydride precipitation

2.1 Constant stress model

IRSN already developed a model to predict the radial hydride precipitation under constant stress at 24°C/h cooling rate [2]. Based on metallographs performed at room

temperature, two tangential stress thresholds have been identified, $\sigma_{t,0\%}$ below which only circumferential hydrides can precipitate and $\sigma_{t,100\%}$ above which 100% of newly precipitated hydrides are formed in the radial direction.

$$\sigma_{t,0\%}(\text{MPa}) = 0.02[H](\text{wppm}) + 186.9 - 0.3862\text{Min}(T_{\text{max}}(\text{°C}); T_{\text{SSD}}(\text{°C})) \quad (1)$$

$$\sigma_{t,100\%}(\text{MPa}) = 110 + 65 \left(1 - e^{-\frac{[H](\text{wppm})}{65}} \right) \pm 20 \quad (2)$$

T_{SSD} is, the temperature required to dissolve $[H](\text{wppm})$ hydrogen while heating from room temperature, derived from the Une and Ishimoto correlation [3]:

$$T_{\text{SSD}}(\text{°C}) = -\frac{4395}{\ln\left(\frac{[H](\text{wppm})}{12.8 \times 10^4}\right)} - 273.15 \quad (3)$$

The data from Slattery [4] on Zircaloy-4 was used to adjust the terminal solubility while cooling:

$$C_{\text{SP}}(\text{wppm}) = 69100. e^{-\frac{3596}{T(\text{K})}} \quad (4)$$

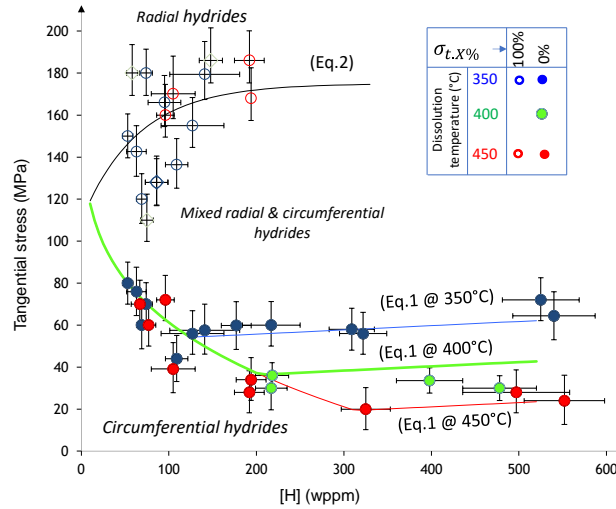


Fig. 1. Hydride orientation/precipitation map between 350 and 450°C maximum temperature.

The fraction of radial hydrides precipitated at room temperature under constant hoop stress, σ_t , is modeled using a simple lever rule [2]:

$$F_R([H], \sigma_t, T_{max}, T_{SSD}) = \begin{cases} 0 & \text{for } \sigma_t \leq \sigma_{t,0\%} \\ \frac{\sigma_t - \sigma_{t,0\%}}{\sigma_{t,100\%} - \sigma_{t,0\%}} & \text{for } \sigma_{t,0\%} \leq \sigma_t \leq \sigma_{t,100\%} \\ 1 & \text{for } \sigma_{t,100\%} \leq \sigma_t \end{cases} \quad (5)$$

2.2 Decreasing stress model

The above-described model was interpreted and transformed to extend its applicability to the instantaneous fraction of radial hydride precipitated in the cladding at each cooling step under applied hoop stress [5, 6]. A very close approach was developed by Rashid and Machiels [7]. The fraction of hydrogen precipitated in the radial direction ($d[H]_R$) formed when precipitating $d[H]_P$, at each time step is defined as: $f_R = \frac{d[H]_R}{d[H]_P}$. This parameter can be easily derived from the constant stress model by comparing the radial hydride fraction at room temperature between two very close precipitation temperatures ($\sim T$):

$$f_R([H], \sigma_t, T, T_{max}) = \frac{\partial(F_R([H], \sigma_t, T_{max}, T_{SSD})C_{SP}(T))}{\partial C_{SP}} \quad (6)$$

In the above equation, the T_{SSD} temperature is deduced from $T_{SSD}(C_{SP}(T))$. To illustrate the model, the obtained function is represented in Fig.2. The plots show that under constant stress, radial hydrides rather form during early stage of precipitation below $\sigma_{t,100\%}$. And above this value, radial hydride precipitation is expected all along the cooling transient.

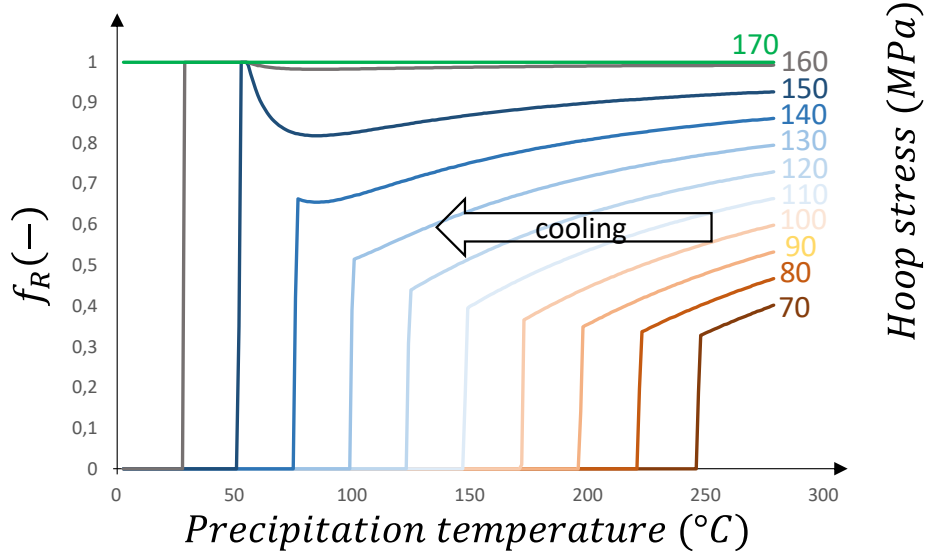


Fig. 2. Predicted fraction of radial hydrides formed during cooling from 350°C modeling 100 wppm hydrogen content.

3 Pressurization tests performed on Zircaloy-4 tubes

3.1 Tested material

Six 90 mm long tubes constituted of low tin SRA Zircaloy-4 17x17 cladding tubes were uniformly pre-hydrated under pure vacuum at about 100 and 150 wppm.

Table 1. Zircaloy-4 composition.

Sn (wt%)	Fe (wt%)	Cr(wt%)	O(wt%)	H(wtppm)
1.30	0.21	0.11	0.14	7

3.2 Testing conditions

The tubes were sealed with Swagelok and pressurized with argon using a GE Druck PACE 5000 for pressure control. The rodlets were inserted in a Instron heating furnace set-up with a Eurotherm controller. The transient is illustrated in Fig.3. The sample was heated during 1h to 350°C, a 2h dwell time was imposed to achieve homogeneous temperature within the furnace and then slowly cooled at 24°C/h. When reaching 315°C, about 25°C above precipitation solvus, the internal pressurization was established and controlled at different cooling rates.

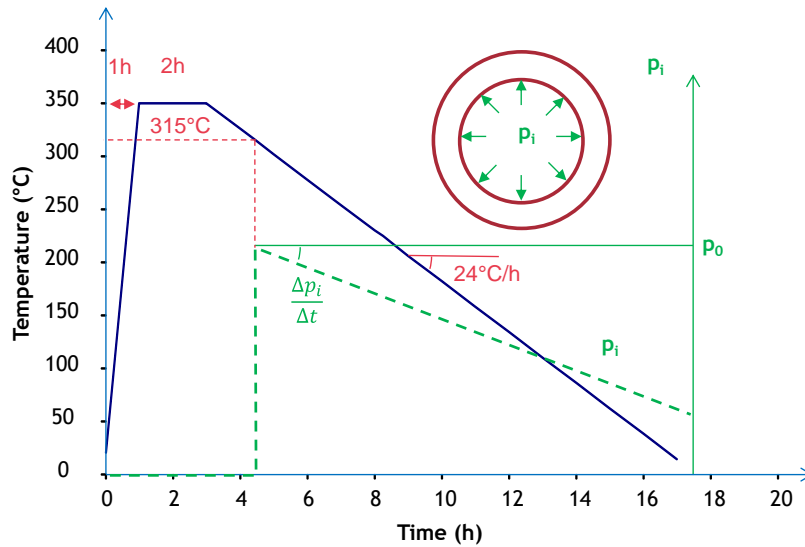


Fig. 3. Heating and pressurization transients imposed to the samples.

3.3 Post-test examinations

After testing, a cross sectional metallographic examination with hydride etching was performed using a Keyence 5000 numerical microscope. The entire cross-section was

analyzed, using ImageJ image analysis software, to determine the radial hydride fraction and associated uncertainties. Hot vacuum measurements of hydrogen content were obtained with a LECO OH836 using two rings cut below and above the metallographic cross section position.

3.4 Test matrix and results

Table 2, provides a summary of the test parameters and main results obtained after material testing. Tests 1 to 4 had hydrogen contents close to 100 wppm and tests 5 to 7 close to 150 wppm. Four depressurization rates were evaluated form 0, 1, 5 and 10 bar/h. The measured radial hydride fraction and uncertainties appear in Table 2. The test #7 at constant pressure stands as a reference test.

Table 2. test matrix and main results.

Test (#)	[H] (wppm)	$p_{i,max}$ (bar)	$p_{i,min}$ (bar)	dp_i/dt (bar/h)	$\sigma_{tt,max}$ (MPa)	$\sigma_{tt,min}$ (MPa)	F_R (%)
1	82	150.3	138.3	1.0	110.2	101.4	31.9±4.6
2	96	150.4	89.8	5.0	110.3	65.9	26.4±4.7
3	98	150.3	25.4	10.0	110.2	18.6	19.9±2.4
4	101	150.1	137.6	1.0	110.0	100.9	30.1±4.6
5	148	150.4	29.5	10.0	110.3	21.7	14.1±4.7
6	155	150.5	88.9	5.1	110.4	65.2	18.6±4.3
7	156	150.2	150.2	0.0	110.1	110.1	25.0±4.7

4 Test analysis and interpretation

The experimental post-test radial hydride fractions (F_R) are plotted in Fig.4, separating samples containing about 100 wppm and the one containing 150 wppm. The samples containing 150 wppm have hydrogen content slightly above the dissolution limit at 350°C (there are consequently remaining circumferential hydrides at maximum temperature) whereas the samples containing 100 wppm experience full dissolution at the same maximum temperature. As expected, the samples with remaining circumferential hydrides lead to lower radial hydride fraction than the samples experiencing full dissolution (100wppm). For both hydrogen contents, a depressurization during the cooling transient has a limiting effect on radial hydride precipitation. This is consistent with the modeling results of Fig.2 showing that the instantaneous radial hydride fraction precipitated decreases with decreasing stress.

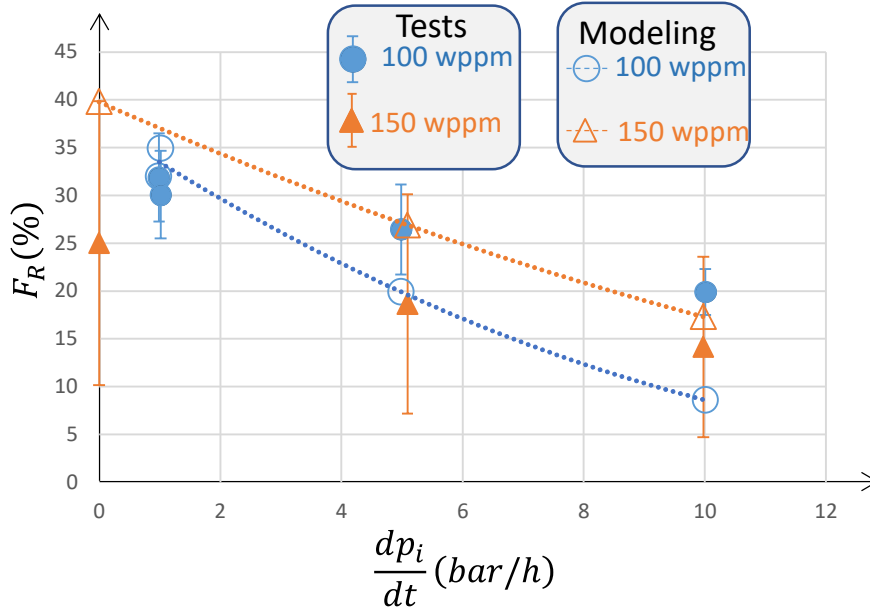


Fig. 4. Test and modeling results of moderate cooling transients at various depressurization rates.

The tests have been modeled using the above-described modeling and the results are compared to experimental data in Fig.4. The models acceptably predict the influence of depressurization however the prediction accuracy has some sensitivity to hydrogen content.

There is a clear benefit in modeling the influence of decreasing stress on radial hydride precipitation and simple models provide acceptably accurate predictions.

5 Conclusion

This paper summarizes long-term objectives of the testing campaign performed at IRSN on Zircaloy-4. Past results [2] showed that there was a complex influence of maximum temperature and hydrogen content on radial hydride precipitation under constant stress.

These past results were used to build a model to assess the influence of decreasing stress on radial hydride precipitation. The model provides a reasonably good assessment of the influence of a decreasing pressure. This model is now implemented in the SHOWBIZ [1] multiphysics code to add a modeling capability dedicated to back-end cycle to the already existing modeling developed for oxygen diffusion under LOCA conditions.

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