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Post-Quench Ductility Test on Pre-hydrided Zr-Nb Cladding and Review on ECCS Criteria

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1. Introduction

Emergency core cooling system (ECCS) acceptance criteria was established to assure cladding integrity in situation of loss of coolant accident (LOCA). Two different assessment methods exist in establishing ECCS criteria. Widely used ductility-based criteria utilize ring compression test (RCT) at 135℃, which is water saturation temperature after reflood. Therefore, the rationale for selecting this temperature is to assess cladding ductility after LOCA. On the other hand, fracture-based criteria utilizes rod integral test to determine whether cladding will fracture due to thermal shock during reflood quenching. Notable difference in two methods is that ductility-based criteria focus on cladding integrity after LOCA, while fracture-based criteria focus on thermal shock during LOCA.

When the test was conducted with fresh fuel claddings, both methods draw similar equivalent cladding reacted (ECR) and peak cladding temperature (PCT) limit, which is ECR 17%, 15% and 1200℃. However, when it comes to high-burnup claddings, two methods draw different results. Ductility-based criteria lowered its ECR and PCT limit with burnup increase, while fracture-based criteria maintained ECR and PCT limit regardless of burnup increase (Fig. 1) [1-2]. To find out the reason of the difference between two methods, each method will be thoroughly discussed in this paper.

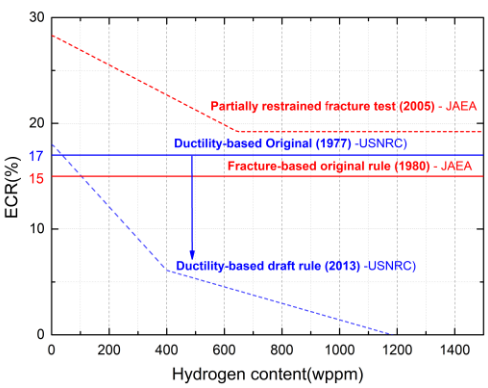

Fig.1. Ductility-based ad Fracture-based criteria for high-burnup fuel

2. Research methods

2.1. Materials

Zr-Nb cladding tube with outer diameter 9.5mm, thickness 0.57mm was used in this study. Composition of the cladding is shown in Table 1.

Table. 1. Composition of Zr-Nb cladding tube

	O	Nb	Sn	Zr
Zr-Nb (wt.%)	0.1-0.16	0.8-1.4	0.9-1.3	Bal.

2.2. Post-quench ductility measurement of pre-hydrided cladding

Cladding was hydrogen gaseously charged with hydrogen charging apparatus shown in Fig.2, to surrogate high-burnup effect. Hydrogen charging was done in range from 200wppm to 800wppm.

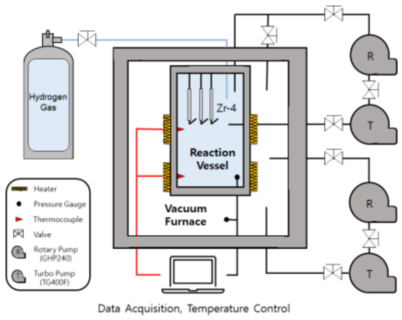

Fig.2. Gaseous hydrogen charging apparatus

High-temperature oxidation was performed with LOCA facility shown in Fig.3. Oxidation at 1200℃ was done until specimen reaches to purposed oxidation level, and specimen was quenched at 1200℃ and 800℃ with two different types of quenching water temperature (100℃, 25℃).

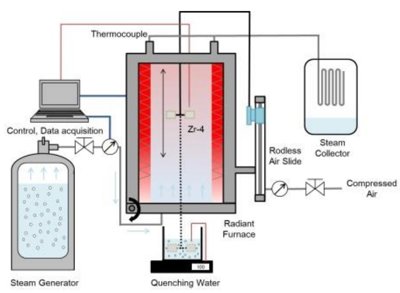

Fig.3. LOCA apparatus

Typical temperature curve of 1204℃ oxidation and quench test is shown in Fig. 4.

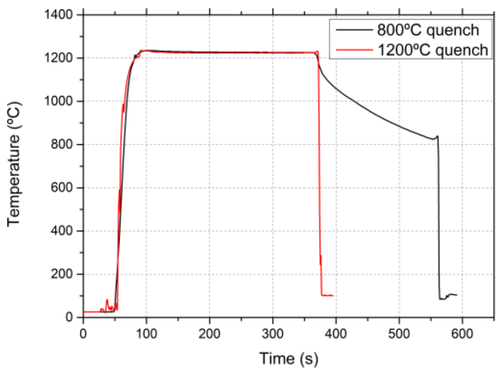

Fig.4. Temperautre during oxidation test

Effect of different quenching temperature, cooling history, quenching water temperature on Post-quench ductility (PQD) is explored in this study.

Ring compression tests were performed with various pre-hydrided, oxidized Zr-Nb specimens. RCT at two different temperatures were performed in this study. 135℃ RCT was performed to reproduce U.S.NRC PQD results of ductile-to-brittle transition ECR level for different hydrogen contents. RCT at peak thermal shock stress temperature, derived from code-integrated study at 2.2, was also performed to test cladding ductility when it is at highest stress during LOCA.

Residual ductility was determined with two different criteria. Offset strain criteria, suggested in U.S.NRC draft rule [1], and stored-energy criteria, which is suggested in this study, were both used.

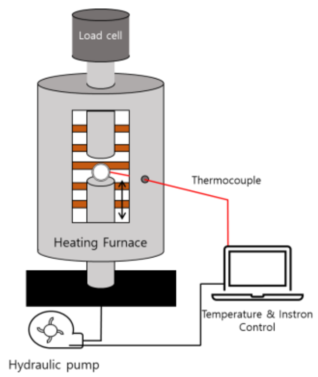

Fig.5. Ring compression test apparatus

2.2. Code-integrated LOCA analysis

For high-burnup fuels, hydrogen inside the cladding plays important role on determining cladding ductility. If hydrogen is in precipitated state, it forms hydride which significantly lowers cladding integrity, while dissolved hydrogen has little influence on cladding integrity. During LOCA transient, hydrogen is either precipitated or dissolved depending on the changing of cladding temperature.

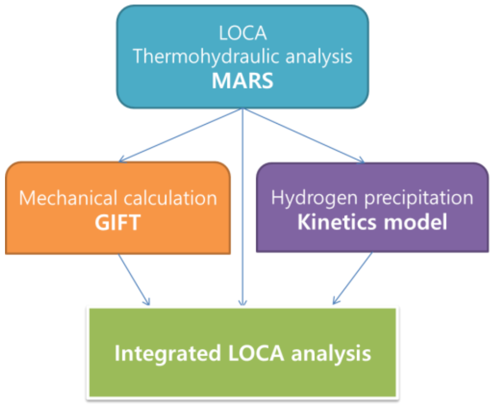

Fig.6. Schematic for Code-integrated LOCA analysis

In this study, MARS LOCA simulation, hydrogen precipitation kinetics module, structure analysis module was used to analyze cladding integrity during LOCA.

MARS-KS code with KINS-realistic estimate method (KINS-REM) was used to estimate temperature during LOCA. Code input was based on Ulchin(UCN) 3&4(OPR-1000). Since the exact and high resolution temperature profile is needed for structure analysis, cladding heat structure was divided to 99 axial mesh, and 21 radial mesh.

Hydrogen precipitation model is based on Kammenzind's study [3], and hydrogen precipitation parameter K_p=0.012s^{-1} was acquired from Zanellato's study [4]. Hydrogen kinetics correlation, and terminal solid solution for precipitation (TSSP) correlation is shown below.

$$\frac{dC_{ss}}{dt} = K_p(C_{ss} - TSSp) \quad (1)$$

$$TSSp = 1.39 * 10^5 \exp\left(-\frac{34470}{RT}\right) \quad (2)$$

(C_{ss}: Solid solution hydrogen content
T: Temperature, R: Gas constant)

Structure analysis code is SNU in-house code using finite difference method (FDM). Calculation accuracy of this code was confirmed by comparing code calculation results and ANSYS calculated results [5].

3. Results and discussion

3.1. Post quench ductility(PQD) test results

RCT test results in fig.7 shows that RCT at 135℃ successfully reproduced PQD test results of U.S. NRC [1]. RCT at peak cladding stress temperature (200℃) showed slightly higher ECR limit than 135℃ RCT. However, ECR limit of 200℃ RCT was still lower than ECR limit of fracture-based criteria (15% ECR). This is because hydrogen is precipitated in 200℃ RCT test condition, while almost all of hydrogen is dissolved at the peak stress time of refload quench.

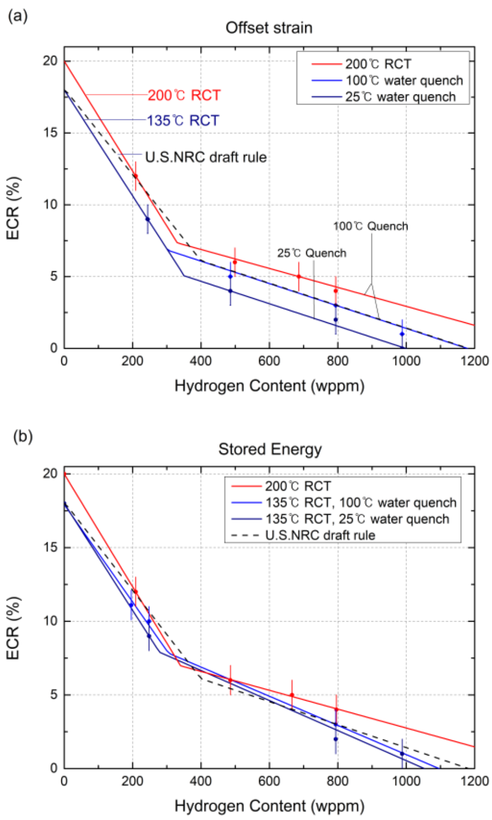

Fig.7. RCT results of Pre-hydrided and oxidized specimen at 135℃-100℃ water quench, 135℃-25℃ water quench, 200℃-100℃ water quench. (a) Offset strain criteria, (b) stored energy criteria

For offset strain criteria, 100℃ water quench had slightly higher ECR limit than 25℃ water quench result. However, for stored energy criteria, difference between two quenching conditions has been diminished. In fig. 8, two different RCT results- H 0wppm, ECR17% and H 908wppm-ECR 1% is shown. H 908wppm specimen had offset strain which is lower than ductile limit (1.6% offset strain) so it was classified as brittle. However, it had higher fracture stress, stored energy than ECR 17% specimen. This implies that offset strain criteria can be sometimes excessively conservative.

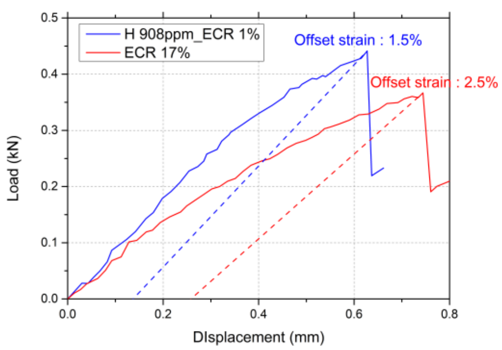

Fig.8. RCT result of H 0wppm, ECR 17% specimen and H 908wppm-ECR 1% specimen

3.2. Effect of cooling history

Experiment on two quenching condition, 800℃ quench and 1200℃ direct quenching condition was conducted.

Past studies on cooling rate effect on PQD showed that mechanical properties of the cladding do not change due to cooling rate for as received cladding.

Different microstructures of each cooling condition is shown in Fig.9. 1200℃ qucnch showed thinner lamellar width than 800℃ qucnch. However, this difference in microstructure had little influence on RCT result (Fig.10).

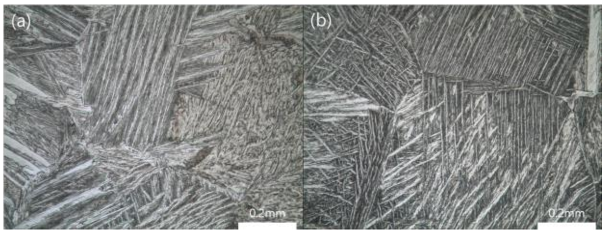

Fig.9. Microstructure of Prior-β phase layer. (a) 800℃ quench, (b) 1200℃ quench.

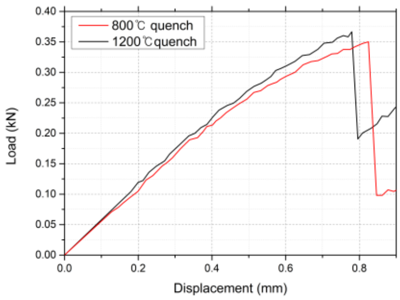

Fig.10. RCT results of ECR 17%, 1200℃ quench and 800℃ quench.

But for high-burnup cladding, slow cooling made specimen more ductile. This effect was explained by the hydrogen diffusion from β phase to α phase. Hydrogen diffusion made prior- β phase more ductile, and made cladding ductile as a whole. Accordingly, 800℃ quenching made H 427wppm-ECR 4.6% specimen more ductile, as shown in Fig. 11.

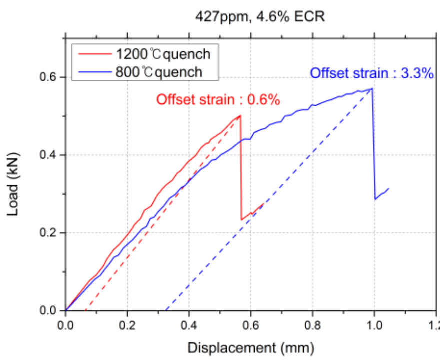

Fig.11. RCT results for 1200℃ and 800℃ quenched H 427wppm-ECR 4.6% specimen

3.3. Interpretation on difference between ductility and fracture-based criteria

LOCA analysis in fig. 12 shows that peak cladding stress occurred during reflood quenching. And at the timing of peak cladding stress occurrence, (around 200℃) hydrogen is still at dissolved state, because of slow precipitation kinetics. This discordance of peak stress and hydrogen precipitation time occurred generally for all axial position of the cladding. High stress imposed to the cladding at the early moment (~1 sec) was due to the sharp temperature rise and resulting temperature gradient at the beginning of the LOCA.

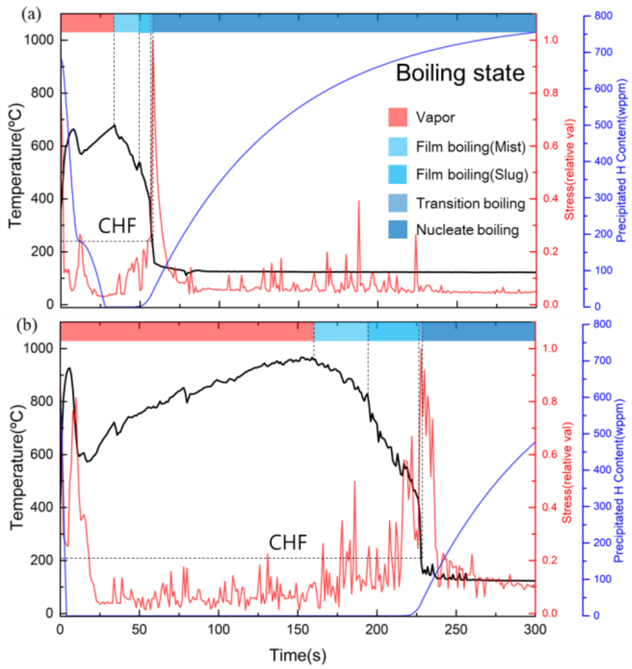

Fig.12. LOCA integral analysis of UCN 3&4. (a) Axial location 1.17m, (b) axial location 2.99m

This result explains how fracture-based criteria maintained its ECR limit in high-burnup fuel claddings. It is because of the slow hydrogen kinetics, compared to quick quenching of the cladding during reflood. Since dissolved hydrogen has little influence on the integrity of the cladding, fracture-based criteria could have maintained same ECR limit independent to burnup effect. Nevertheless, for ductility-based criteria, almost all of the hydrogen is in precipitated state (TSSp = 5.3wppm for 135℃). Precipitated hydrogen forms hydride, which severely deteriorates cladding integrity. This resulted in lowering ECR limit for ductility-based criteria.

Cladding located at 1.17m gets thermal shock at ~50s, while cladding at 2.99m gets thermal shock at ~230s. And at the time of ~230s, stress is imposed to the cladding at 1.17m due to the thermal shock of upper part. And at that moment, most of hydrogen is in precipitated at 1.17m. This implies that fracture can occur even after the time cladding get thermal shock, due to the stress propagation from different part of the cladding.

4. Conclusion

High temperature steam oxidation and RCT was conducted with pre-hydrided and oxidized Zr-Nb cladding. 135℃ RCT reproduced similar ductility boundary with U.S.NRC test results. RCT at peak cladding thermal shock temperature (200℃) showed slightly increased ECR boundary. But even in 200℃ RCT, ductile ECR boundary was still lower than that of fracture-based criteria. This discrepancy was also due to the difference between precipitated and dissolved hydrogen.

Fast cooling of the specimen using 25℃ water quench, or 1200℃ direct quenching made pre-hydrided and oxidized specimen more brittle than slow-cooled specimen. Since the cooling rate can be different for different locations in LOCA conditions, this implies that scenario-dependant LOCA experiment can make PQD results less conservative.

Discrepancy between ductility-based and fracture-based ECCS criteria was explained. During LOCA trainsient, reflood quenching and thermal shock occurs so rapidly that slow hydrogen precipitation does not occur until the end of quenching. As a result, fracture-based ECCS criteria maintained same ECR level for high-burnup claddings.

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