The Effect of Cooling Rates on the Post-LOCA Ductility of Zircaloy Cladding

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1. Introduction

The current Emergency Core Cooling System (ECCS) criteria (LOCA criteria, 10 CFR 50.46) are set to guarantee an adequate level residual cladding ductility following high temperature steam oxidation and subsequent water quenching. The current U.S NRC's experimental protocol uses the following conditions for standardized experiments:

-Quenching of cladding specimen by dropping into water bath (temperature unreported)

-Ring Compression Test (RCT) for ductility assessment

-strain rate: 0.033mm/s

-offset strain criteria: 2%

-temperature: 135℃

Some of these values can be readily rationalized based on the realistic depiction of conditions to which the fuel materials are subjected. For example, the temperature condition 135℃ should be relevant to the saturated temperature of water at ~3 atm which may represent a substantially depressurized condition upon the occurrence of LBLOCA.

Efforts are being made to assess the rationales of these experimental protocols for potential application to high burn fuel and accident tolerant fuel cladding ECCS criteria. In the experimental protocols, a noticeable knowledge gap exists in the quenching water temperature. The work that laid a foundation on 10 CFR 50.46 unreported the water bath temperature [Hobson]. It is presumable that the consideration for the potential effect of water temperature was neglected. Partly due to the lack of attention in the founding work, no attention has been made to the water bath temperature effect in subsequent investigations to date; no technical consensus or standardization has been made as various (i.e., oil) or water bath temperature are used to quench the cladding in regulation supporting studies.

Recent studies on thermal shock fracture revealed the importance of heat transfer rate on material's thermal shock fracture upon water quenching. It can be inferred from these past studies that the quenching water temperature can significantly affect the boiling heat transfer rates on cladding surface, thereby changing the stress levels. Different cooling rates and resulting stress levels may imply different states of oxide scale integrity and resulting frozen phase, all of which may affect the cladding's post-LOCA ductility.

In such a context, this study is designed to experimentally explore the effect of water bath temperature on post-LOCA zircaloy cladding ductility. As-received Zircaloy-4 cladding and pre-hydrided Zircaloy cladding were oxidized to ~17% CP ECR at 1204℃, and subsequently water quenched at room temperature water (~20℃), boiling water (~100℃), and air-cooled, respectively. Their residual ductility were investigated by RCT with Digital Image Correlation (DIC), and analyzed using Optical Microscopy (OM), Scanning Electron Microscopy (SEM), and Energy Dispersive Spectrometry (EDS).

2. Experimental setup

2.1. Sample preparation

Samples used in this experiment were the Zircaloy-4 cladding tube specimen (OD:9.5mm, thickness = 0.57mm).

To simulate high-burnup effect on Zircaloy cladding, some samples were cut into 10cm tube, and charged with 100% H_2 gas in 400℃ for 4 hours, and heat-treated at 400℃ for 24 hours to make homogeneous distribution of hydrogen in cladding tube. The resulting hydrogen contents in those samples were 283 ± 12wppm.

2.2. LOCA experiment

All samples, including hydrogen-charged samples and as-received Zr-4 cladding samples were cut into 1cm in length, and located on alumina-coated specimen holders as shown in Fig. 1.

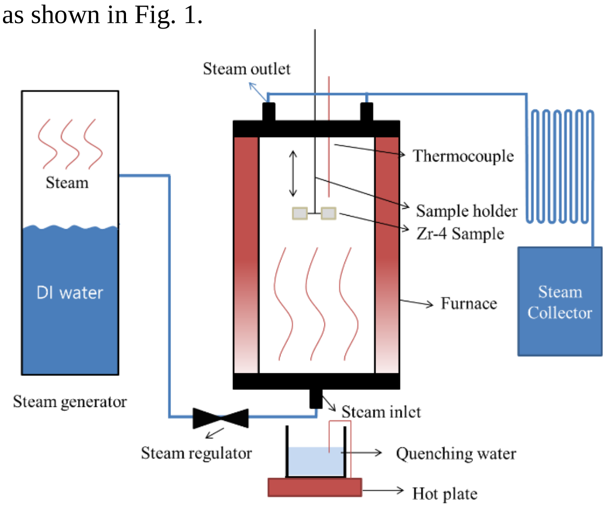

Fig. 1. Schematic of LOCA experimental set up

The specimens were oxidized with high temperature steam at 1204℃ ± 5℃ to 17% Equivalent Cladding Reacted (ECR). The oxidized specimens were then quenched into water bath (either 100℃ or room temperature ~20℃) or air-cooled.

2.3. Ring Compression Test

Cladding specimens were cut into 0.625cm to match the length used for RCT test of U.S NRC. Ambient temperature during the test were either at 20℃ or 135℃, and strain rate was 0.033mm/s. DIC measurements were conducted to obtain the strain fields of tested specimens during RCT. The ductility assessment was conducted using the obtained stress-strain curve with 2% offset criteria.

3. Results and discussion

3.1. Oxidation rate

Steam oxidation of zircaloy specimens were conducted at 1204±5℃, and the oxidation rates were compared with Cathcart-Pawel (CP) and Leistikow correlation, in terms of ECR, as shown in Fig. 2. Experimental ECRs were obtained by measuring the mass gain after oxidation. The conducted experiments give a good agreement with both CP and Leistikow correlation.

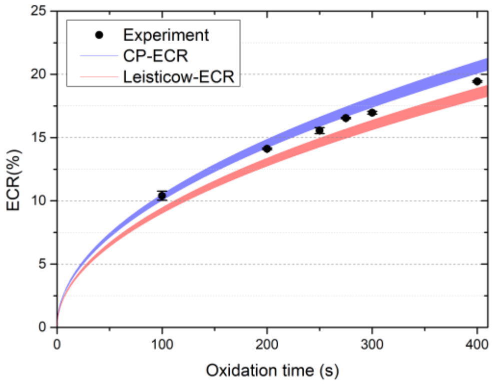

Fig. 2. Comparison between CP-ECR (1204±5℃) and mass-calculated ECR in this experiment

Oxygen distributions of specimens were analyzed by Energy Dispersive Spectrometry (EDS) as shown in Fig. 3. The oxygen distribution and amounts are in agreement with the past experiments, demonstrating the sanity of the conducted experiments and facility. The oxidized specimens were used for metallographic analysis and ring compression tests with Digital Image Correlation (DIC) technique.

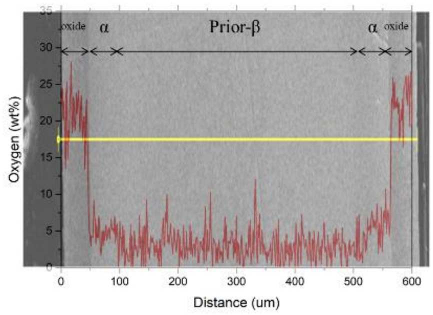

Fig. 3. Energy Dispersive Spectrometry (EDS) of phase layer. Oxide, α, and prior-β layer are shown.

3.2. Metallographic analysis for phase thicknesses

Thicknesses of each phase (α, prior-β layer, and oxide scale) for three different cooling rates were measured using optical microscopy, as shown in Fig. 4.

Fig. 4. Optical microscopy snapshot of phase layer in (a) air-cooled sample, (b) 100℃ water-quenched sample, (c) 25℃ water-quenched sample

For each specimen total 8 measurements were made for every 45° angular positions along the azimuthal direction of the cladding. The measured thickness for each phase is shown in Fig. 5.

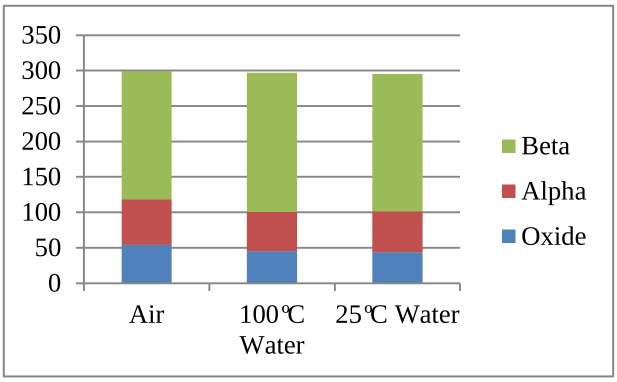

Fig. 5. Thicknesses of each phase layer. Air-cooled specimen had thickest oxide and alpha layer.

The thinner oxide layers found in the water quenched specimens are considered due to breakage of some oxide layers upon thermal shock induced by quenching. Slightly thicker alpha-layer of the air cooled specimens is considered to pertain to the slow cooling rates which allow a longer time period for the transition of β to α. It is noteworthy that the phase thickness difference for the different cooling rates is considerably small. This may imply limited sensitivity of the post-LOCA cladding ductility on quenching process.

3.3. DIC supported Ring Compression Test analysis

RCT was conducted to assess post-LOCA ductility of different cooling rates. The applied strain rate (0.033mm/s) was used in compliance with the NRC's test protocol. The measured off-set strains with the first load-drop was compared against the U.S NRC's 2% offset criteria. In the room temperature testing, no appreciable difference among the tested specimens was observed. This is considered due to the considerably small difference in the resulting phase of three cooling rates (Fig.5).

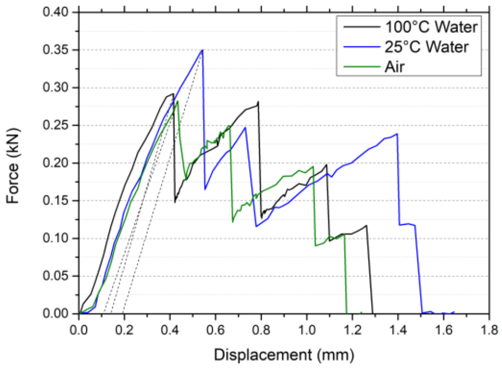

Fig. 6. Displacement-Force graph of ring compression test (RCT) in room temperature (20℃). Each claddings had 1.5%, 2.0%, 1.1% offset strain.

DIC was performed to explore potential difference in local strain fields during RCTs (Fig.7). All cooling rates exhibited similar strain fields during RCT deformation, demonstrating no appreciable difference in mechanical behavior of post-LOCA cladding materials with respect to cooling rates in the quenching stage.

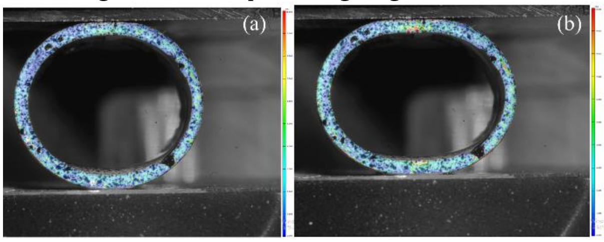

Fig. 7. Digital image correlation(DIC) analysis of cladding sample. (a) before failure (crack opening), (b) after failure of the sample.

5. Conclusion

No appreciable effect of cooling rates in quenching state on cladding's post LOCA ductility is yet identified with the RCTs conducted at room temperature. The observed cooling rate insensitivity is primarily due to similar phase thicknesses. This result provides a technical basis that may support the sanity of past regulatory experiments which used various quenching environments without considering its potential effects. Yet, the insensitivity of the cooling rates is an evidence that the current ductility based approach may inappropriately weigh on the phase thicknesses, thereby neglecting the effect of water side in the quenching stage.

6. Acknowledgement

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REFERENCES

[1] Y. H. Lee, B. K. Kim, H. C. No, Improving safety margin of LWRs by rethinking the emergency core cooling system criteria and safety system capacity, Nuclear Engineering and Design, Vol. 307, p.402, 2016

[2] D. O. Hobson, P. L. Rittenhouse, Embrittlement of Zircaloy Clad Fuel Rods by Steam during LOCA Transients, ORNL-4758, 1972

[3] F. Nagase, T. Fuketa, Fracture behavior of irradiated Zircaloy-4 cladding under simulated LOCA conditions, Journal of Nuclear Science and Technology, Vol. 43, p.1114,2016

[4] L. Maolong, J. Saetbyul, K. Hyungdae, L. Youho Experimental and Analytical Investigation into Boiling Induced Thermal Stress: Its Impact on the Stress State of Oxide Scales of Nuclear Components. Nuclear Engineering and Design, 341, 66-72, 2019.

[5] R. E. Pawel, J. V. Cathcart, R. A. McKee, The kinetics of oxidation of zircaloy-4 in steam at high temperatures, Journal of The Electrochemical Society, 1979