Effect of non-Isothermal Transient Zircaloy Oxidation on Emergency Core Cooling System Criteria

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1. Introduction

The current Emergency Core Cooling System (ECCS) criteria (LOCA criteria) are set to assure an adequate level of post-LOCA cladding ductility. The criteria limit the Peak Cladding Temperature (PCT) and Equivalent Cladding Reacted (ECR) to 1204°C and 17% (calculated by the Baker-Just correlation), respectively.

These criteria were developed by quantifying residual ductility of isothermally steam-oxidized and subsequently water quenched Zircaloy cladding specimens in Ring Compression Test.

In reality, LOCA accompanies a significant time-varying temperature change of fuel cladding. The current experimental protocols for the aforementioned post-LOCA cladding ductility assessment neglect the non-isothermal nature of cladding oxidation.

The key knowledge gap of using the isothermal experimental data is twofold; 1. The isothermal ECR prediction is believed to have limited accuracy for non-isothermal oxidation transience, and 2. No assurance is given to the agreement of post-LOCA ductility between isothermal and non-isothermal oxidation. That is, even if ECR prediction is accurate, question remains if post-LOCA ductility of non-isothermally oxidized cladding and isothermally oxidized cladding would be acceptably identical.

Hence, from the perspective of post-LOCA ductility assurance, the effect of non-isothermal oxidation on both ECR prediction and cladding mechanical behavior needs to be systematically quantified.

This study aims at assessing the effect of non-isothermal transience on the predictability of existing ECR correlations, and cladding's post-LOCA residual ductility. To verify it, cladding steam oxidation experiments were conducted to compare the ECR calculated by an existing isothermal correlation and ECR experimentally obtained in rapidly varying temperature. Ring compression tests are then followed to measure stress-strain curve of both cases from which the residual ductility is assessed.

2. Experiments

2.1. LOCA Experiments

The experimental facility has a steam boiler which boils and introduces atmospheric steam into the test section. The steam is further heated by radiant heaters surrounding the test section up to ~1450°C. The steam leaving the test section is collected by a condenser and the entire system is closed to prevent air ingress.

Experiments were performed on Zircaloy-4 cladding tube specimens. The length, outer diameter, and thickness of tested specimens were 10 mm, 9.5 mm, and 0.57 mm, respectively. Specimens were oxidized by flowing steam in a radiant heating furnace shown in Fig. 1. The condition within the furnace was maintained steady by flowing steam for 15 minutes before starting the test. The steam temperature right adjacent to the specimen was measured using a K-type thermocouple inserted in the furnace (Fig. 1). For isothermal tests, a steady-flow of steam is introduced in the test section. For non-isothermal tests, temperature drop was achieved by increasing the steam flow rate.

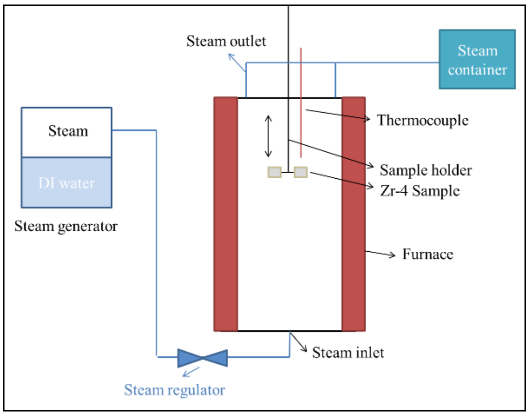

Fig. 1. Schematic diagram of experiment device

ECR was quantified by measuring weights with a digital balance that can make a measurement to 5 decimal-point. Ring compression tests were conducted on as-received, and oxidized cladding at room temperature in compliance with the US. NRC's post LOCA cladding ductility assessment protocols (strain rate = 0.033mm/s). Metallographic analyses were conducted using Optical Microscopy (OM) and Digital Image Correlation(DIC).

3. Results

3.1 Isothermal ECR

The experimental results were compared with CP correlation. As can be seen in Fig. 2, the obtained results are in a good agreement with the CP correlation. CP correlation for ECR is shown in Eq. (1) and (2).

$$(w_0)^2 = 0.3622 * t * \exp(-39940/RT) \qquad (1)$$

$$\mathrm{ECR} = 2.85 * w_0/(\rho_z h_r) \qquad (2)$$

This result provides another validation for CP correlation in an isothermal condition. The ECR region of temperature range(1194 °C~1214 °C) is colored at Fig. 2 and two specimens were oxidized at each oxidation time cases.

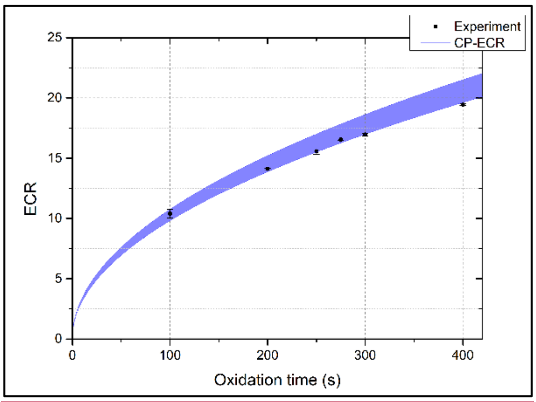

Fig. 2. Experimentally obtained ECR and CP correlation prediction at 1204 °C

3.2 Non-isothermal transient ECR

Steam flow rate increased to introduce changes in temperature. The tested transient history is shown in Fig. 3(a). The peak temperature for all tested five cases was ~1210°C. The temperature decreased to roughly ~1000 °C at various rates, as can be seen in Fig. 3(a). Such temperature transience is considered relevant to postulated LBLOCA scenarios.

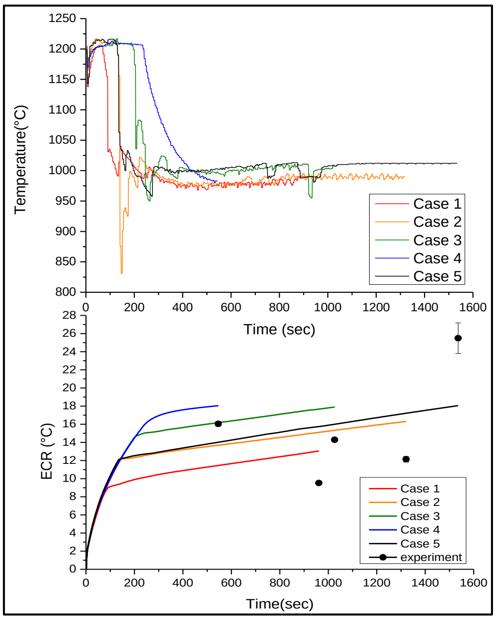

Fig.3. (a) tested temperature transience (b) calculated ECR with Cp correlation

The isothermal Cp correlation was used for ECR calculation. The Cp correlation can be forced to change temperature from T_i to T_{i+1} during the time step Δt using Eq.(3).

$$(w_{i+1})^2 = (w_i)^2 + 0.3622 * \exp(-39940/RT_i) \quad (3)$$

This method that enables an isothermal correlation to be applied for non-isothermal case only conserves the ECR without conserving individual phase thicknesses (α, α + β, β, and ZrO_2), as illustrated in Fig. 4.

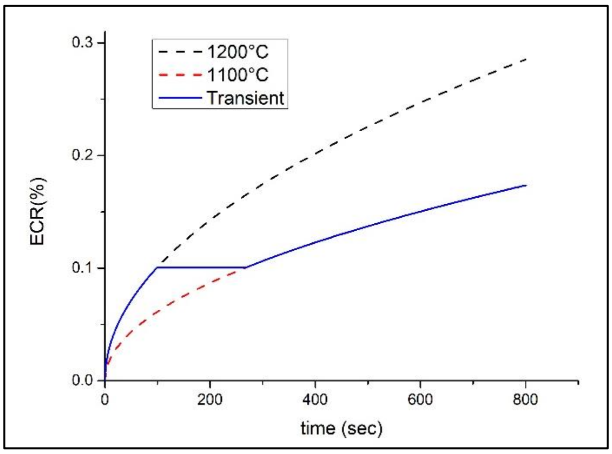

Fig.4. Schematic illustration of isothermal ECR used for non-isothermal transient calculation

This is the basis of the limitation of applying the isothermal correlation for LOCA safety analyses; the use of isothermal correlation is believed to introduce prediction errors for ECR. As anticipated, non-negligible levels of disagreement were found between CP correlation prediction and experimentally obtained ECRs for all tested cases, as can be shown in Fig. 3(b). Considering the remarkable prediction accuracy of the CP correlation for the steady-state oxidation (Fig. xxx), the demonstrated predictions errors for non-isothermal cases (Fig. 3 (b)) and table 1 is non-negligibly big.

It can be noted in Fig. 3(b) that the CP correlation gives a conservative prediction in general. In order to remove unnecessary conservatism in safety analyses, an advanced cladding oxidation model that captures non-isothermal transient behavior is needed.

Table 1 summarizes ECR comparisons for non-isothermal transient tests shown in Fig. 3.

Table 1. Summary of non-isothermal transient ECR

CASE	sample No.	Time(min) 1204°C	Time(min) 1000°C	Experiment ECR(%)	CP based ECR prediction	Error(%)
1	1	1	15	9.63%	13.05%	-26.23
	2			9.39%		-28.06
	3			9.55%		-26.82
2	4	2	20	11.94%	16.31%	-26.75
	5			12.59%		-22.82
	6			11.89%		-27.08
3	7	3	14	14.35%	17.89%	-19.83
	8			14.14%		-20.98
	9			14.39%		-19.58
4	10	4	5	16.15%	18.04%	-10.52
	11			15.69%		-13.07
	12			16.31%		-9.59
5	13	2	23.5	25.62%	18.05%	+41.96
	14			27.51%		+52.40
	15			23.36%		+29.44

3.3.Thermal shock fracture of oxide scale due to steam reflux quenching

For the case 5, Fig.4 shows substantial formations of cracks in oxide scales, implying loss of its protectiveness. This supports significantly high ECR levels for Case 5 (Table 1). This result implies that the structural integrity of cladding oxide scales can be challenged by steam reflux quenching. Steam reflux is an anticipated phenomenon upon LBLOCA in LWRs. Further investigation is needed to understand conditions under which the steam reflux driven oxide fracture takes place.

Fig.5. Picture of fracture

3.4. Comparison of Prior-Beta Layer thickness of specimens through Optical Microscopy

The thickness of prior-beta layer is known to have good correlation of cladding's ductility[2]. Table 5 shows the prior-beta layer thickness obtained from Optical Microscopy(OM). Isothermal specimens corresponding to non-isothermal specimens' ECR were obtained at 1204℃. Fig.6 shows the prior-beta layer thickness of non-isothermal specimens(No.9,12, and 14) and ECR corresponding isothermal specimens which are obtained from Optical Microscopy(OM) shown at Fig.7.

The different thicknesses, which are relevant with ductility ,between non-isothermal specimens and ECR corresponding isothermal specimens implies the different ductility between non-isothermal and isothermal specimens. However, for the small number of the specimens, it can be a hasty conclusion. So, further experiments are needed.

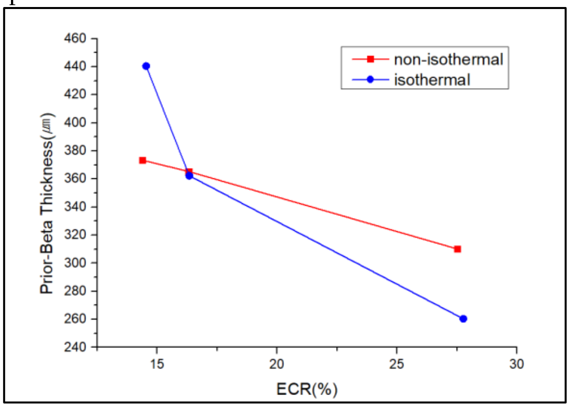

Fig.6.Prior-beta layer thicknesses of non-isothermal specimens(No.9,12, and14) and ECR corresponding isothermal specimens.

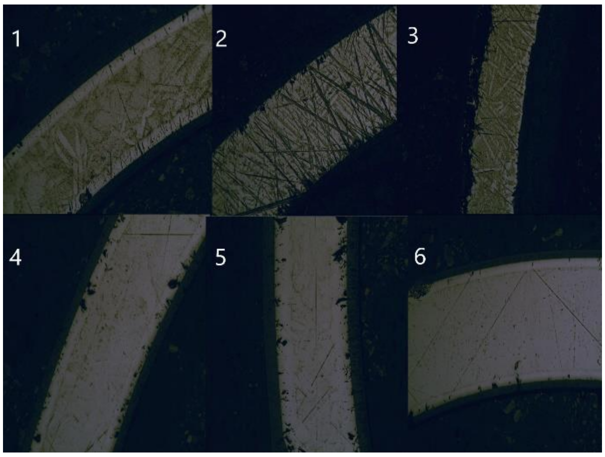

Fig.7. Optical Microscopy of non-isothermal specimens(No.9,12, and 14) and ECR corresponding isothermal specimens.

3.5. Effect of non-isothermal tests on Ring Compression Test (RCT)

To compare the mechanical behavior of non-isothermal specimens(No.3,6) and ECR corresponding isothermal specimens(3`,6`), Ring Compression Test(RCT) was done. The results are organized at Table.2.

Fig.6 shows the difference between the strain-stress curves of specimens(6,6`). For other parameters, it can be seen that when ECR increase, Ultimate Tensile Strength(UTS) and elongation(strain for first failure) decrease, and Young's modulus increase for the non-isothermal cases(3,6) and isothermal cases(3`,6`) respectively. However there are no clear correlation to explain the difference between isothermal and non-isothermal cases. More specimens from experiment are needed.

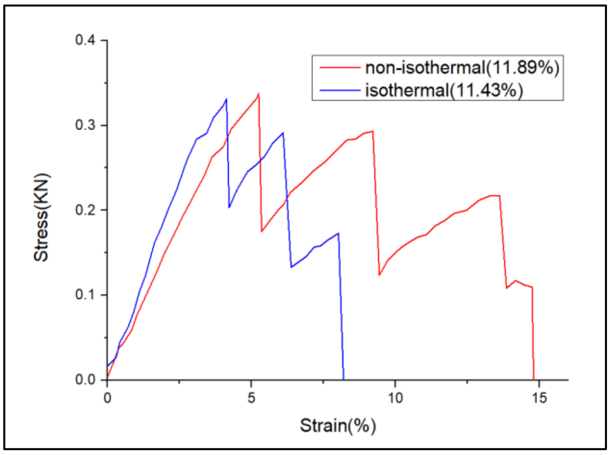

Fig.8. Strain-Stress curve of non-isothermal specimen of ECR 11.89%(specimen 6) and isothermal specimen of ECR 11.43%

Table. 2. Mechanical behavior of specimens

Case	Sample No.	ECR (%)	UTS (KN)	Young's modulus (N)	Elongation (%)
1	3	9.55	0.455	46.214	9.84
	3`	10.69	0.345	87.269	4.4
2	6	11.89	0.338	64.365	5.24
	6`	11.43	0.331	80.243	4.12

3.5. Comparison through on Digital Image Correlation(DIC)

The image comparison and strain comparison during RCT through DIC was also done. The color of the image represent the strain of the ring, so the crack are shown to be red. The first crack position of non-isothermal specimen(No.6) and isothermal specimen(No.6`) was observed to be lower part and upper part of the ring respectively.

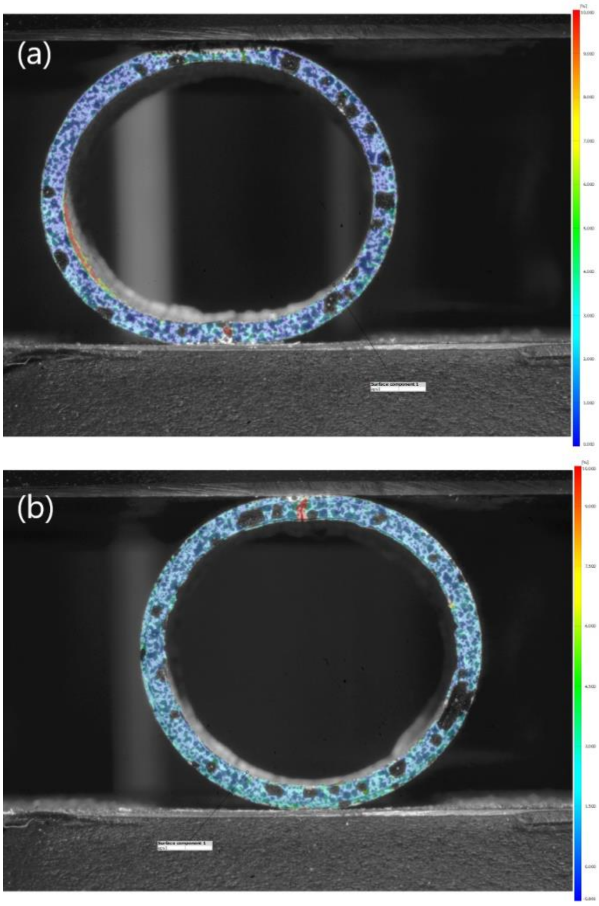

Fig.8. The first crack position(red color) difference of non-isothermal specimen(a) and corresponding isothermal specimen(b).

3. Conclusion

Firstly, from the results of the previous 3.2, it can be inferred that using the current CP correlation for calculation of ECR in the LOCA accident is quite inappropriate with the real ECR. Therefore another method to calculate ECR which can demonstrate the real oxidation process is needed.

Secondly, it can be hard to calculate ECR for the Thermal shock fracture of oxide scale due to steam reflux quenching. This phenomenon should be explained and controlled to predict ECR accurately.

Lastly, there was difference of mechanical behavior between non-isothermal and isothermal specimens of simililar ECR. It can be a hasty conclusion for the small number of specimens, but if there is certainly difference between them it means predicting only ECR is not sufficient to predict the mechanical behavior of the cladding after LOCA accident. Further experiments are needed.

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