Post-LOCA Ductility of Cr-coated cladding and its Implications on Accident Coping Time

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1. Introduction

Coated cladding has been developed to extend accident coping time by suppressing steam oxidation during accidents. There is a time period between the post-LOCA ductility limit and either melting or significant loss of coolable geometry upon reflood quenching which marks an initiation of severe accidents[1]. An accident coping time may refer to the time period before the initiation of severe accident. Violation of the Design Basis Accident (DBA) criteria (i.e., Emergency Core Cooling System – ECCS Criteria) can be considered an initiation of severe accident phase, although its extent on the accident progression may still be limited. Hence, it is important to quantify the increase in accident coping time of coated-Zircaloy cladding in terms of time required to reach the ECCS limit.

It is noteworthy Accident Tolerant Fuel (ATF) candidates may as well comply with the current Emergency Core Cooling System (ECCS) criteria based on post-LOCA ductility in order to main the consistency in regulatory framework. In that, the extension of accident coping time can be evaluated in terms of time extended to reach the post-LOCA ductility limit. Such definition may be taken as the minimum, yet most conservative, extension of accident coping time.

Single-sided coating on the outer cladding surface leads to the internal oxidation upon cladding rupture followed by ballooning at ~ 600-700 °C[2]. Hence, it is anticipated that the coated Zircaloy cladding undergoes appreciable embrittlement due to internal oxidation. In this study, high-temperature steam oxidation and water quenching experiments were conducted to quantitatively assess the effect of coating on post-LOCA ductility, and discusses its implications on extension of accident coping time.

2. Experiments

Zircaloy-4, Cr coated Zircaloy-4 claddings were cut into 0.8 mm and washed with water, ethanol, and acetone, and then the weight and coating thickness before oxidation were measured. Cr coating was applied via cold-spray at 400°C, to avoid mechanical property change. Coated specimes were provided by fuel research group at Massachusetts Institute of Technology (MIT). Oxidation experiments were conducted at 1200°C through the oxidation device shown in Fig. 1, and Zircaloy-4 and Cr coated Zircaloy-4 claddings were simultaneously oxidized while measuring the temperature of the specimen by attaching a thermocouple to the specimen. After oxidation at 1200°C for a certain period of time, the temperature was lowered to 800°C, then quenched into boiling water. Weight gain after oxidation was measured to calculate Equivalent Cladding Reacted(ECR), and Ring Compression Test(RCT) was conducted at 135 °C to evaluate the ductility of the oxidized claddings. The test procedures and protocols for steam oxidation, water quenching, and RCT were conducted in compliance with the U.S NRC's guidelines (DG-1262) to ensure direct regulatory implications [3].

The resulting microstructures of the post-LOCA specimen were compared by Scanning Electron Microscope (SEM). Oxygen concentrations of oxidized cladding were measured with Field Emission Electron Probe Microanalyzer (EPMA).



<Fig 1. Schematic diagram of oxidation facility>

3. Results and Discussion

3.1 Ductility based Accident Coping Time Test matrix and results are summarized in Table 1. Specimens judged to be brittle based on the U.S NRC's offset strain criteria were indicated with red letters.

Oxidation	Cladding	ECR	Offset
Time	Туре		Strain
310s	Cr-coated	8.24%	17.88%
	Zircaloy-4		
	Zircaloy-4	16.55%	3.46%
620s	Cr-coated	x	x
	Zircaloy-4		
	Zircaloy-4	22.72%	1.63%
900s	Cr-coated	13.83%	6.31%
	Zircaloy-4		
	Zircaloy-4	27.38%	0.47%
1200s	Cr-coated	15.53%	1.92%
	Zircaloy-4		
	Zircaloy-4	31.09%	0.80%
1800s	Cr-coated	19.49%	0.63%
	Zircaloy-4		

<Table 1. Results of Oxidation experiment:

	Zircaloy-4	38.76%	0.10%
Fig. 2 shows	temperature	profiles of tested	specimen

prior to water quenching. Specimens were held at constant temperature (~1204°C) and lowered to 800 °C with cooling rate of 2° C/s ~ 3.5° C/s.



<Fig 2. Experiment temperature profile>

Fig. 3 shows the ECR and offset strain of Zircaloy-4 and Cr coated Zircaloy-4 claddings according to oxidation time. Zircaloy-4 agrees well with the doublesided oxidation Cathcart-Pawel (CP) correlation. The coated cladding gives a good agreement with the onesided oxidation CP correlation. As the oxidation time increased, both claddings' offset strain decreased, and when oxidized for the same time, the ductility of coated cladding was better than uncoated cladding for its extent of oxidation is nearly half of the uncoated specimens. It takes 620s for uncoated cladding to reach the post-LOCA ductility limit whereas it takes 1200s for coated cladding. Therefore, the additional accident coping time that can be obtained by coating the cladding outer surface is about 10 minutes which is twice longer than uncoated cladding.



<Fig 3. ECR and post-LOCA offset strain of uncoated and Cr-coated Zircaloy-4: circled points represent brittle specimens based on the U.S NRC's offset strain criteria>

3.2 Oxygen Distribution of Zircaloy-4 and Cr coated Zircaloy-4

Fig.4 shows oxygen distribution measured by EPMA over SEM images of zircaloy-4 and Cr coated zircaloy-4 claddings oxidized at 1200°C for 620 seconds. Zircaloy-4 is double-sided oxidized and result in oxygen-affected region on both sides, whereas Cr-coated Zircaloy clearly exhibits oxygen-affected region on the inner side, demonstrating the performance of the outer coating layer. For each experiment, the ECR of the two claddings are shown in Table 1. It can be seen that the coated cladding is oxidized almost half of the uncoated cladding in terms of ECR. It can be noted that the Zr matrix adjacent to the coating layer is characterized with the prior- β phase which is responsible for post-LOCA ductility.



<Fig 4. SEM and EPMA analysis of oxygen distribution for (a) uncoated Zircaloy and (b) Cr-coated Zircaloy. Oxidized at 1200°C for 620s>

3.3 Offset strain and ECR relation

Fig. 5 shows the ECR and offset strain of the oxidized Zircaloy-4 and Cr coated Zircaloy-4 claddings, and the post-LOCA ductility criterion of U.S.NRC's guideline (DG-1262) [3]. For substantially high ECRs,

the offset strains of uncoated and coated specimens agree closely. However, for relatively low ECRs (<15%), an apparent difference in offset strain is observed, demonstrating lower offset strain, hence ductility, for coated specimens compared to uncoated specimens. This is because when ECR of the one-sided oxidized cladding is the same with that of double-sided oxidized cladding, the oxidation time is longer hence oxygen diffused a longer distance into the beta layer, which is responsible for the ductility of the cladding tube [4]. This explains reduced offset strain for Crcoated Zircaloy-4.



<*Fig 5. Ductility of oxidized* Zircaloy-4 and Cr coated Zircaloy-4>

4. Conclusions

Single sided coating is shown to extend the accident coping time for ~10 minutes based on the post-LOCA ductility criteria. This may serve as the least amount of coping time extension excluding time period between the violation of ECCS limit and appreciable progression of severe accidents. Nevertheless, since the violation of ECCS criteria (DBA limit) may represent a possibility of immediate progression into severe accidents, the explored coping time extension may be taken as a conservative, yet potentially reasonable, evaluation. Double-sided coating may be necessary to further extend the accident coping time in terms of post-LOCA cladding ductility.

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REFERENCES

[1] K. Terrani, Accident tolerant fuel cladding development: promise, status, and challenges, J. Nucl. Mater. (2018), 10.1016/j.jnucmat.2017.12.043

[2] Geelhood, K.G., Luscher, W.G., 2019. Degradation and Failure Phenomena of Accident Tolerant Fuel Concepts, PNNL-28437,

https://www.nrc.gov/docs/ML1903/ML19036A716.pdf.

[3] Draft Regulatory Guide DG-1262, Testing for Postquench Ductility. 2011, U.S. Nuclear Regulatory Commission, Washington, DC.

[4] M. Billone, Y. Yan, T. Burtseva, R. Daum, NUREG/CR-6967/ANL-07/04: Cladding Embrittlement During Postulated Loss-of-coolant Accidents, US NRC, July, 2008