# Effect of Pre-oxide and Pre-hydride on the Zircaloy-4 Cladding in a Simulated LOCA Transient

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#### 1. Introduction

Maintaining fuel integrity in a postulated design-based accident, as well as during a normal operation is of importance. Loss of coolant accident (abbreviated as LOCA) is one of the most important accidents at the design of a light water reactor (LWR). When LOCA occurs, pressurized coolant is discharged so that fuel cladding balloons and even ruptures due to the pressure differences across the cladding. Temperature of the fuel system rises so that the cladding undergoes oxidation in the mixture of the water and steam. After a time interval, the emergency core cooling system is activated, which inevitably accompanies thermal shrinkage of the cladding. When the embrittled cladding cannot stand the stress involved, it fragments, which results in the loss of the barrier preventing fission product release. Nowadays, many power plants adopt high burn-up operation which includes high power, enlarged fuel cycle, and so on. At high burn-up such as 60,000MWd/MTU, oxide and absorbed hydrogen amount of Zircaloy-4 reached the value as 80 µm and 600ppm caused by extended waterside corrosion [1]. It is widely known that thick oxide and large hydrogen uptake caused by normal operation can cause deleterious effect on LOCA properties of the cladding [2]. The objectives of this study are to investigate the effects of pre-oxide and pre-hydride which simulate high burn-up, normal operation on the LOCA property of zirconium cladding. Studies were focused on the separate effects of pre-oxide and pre-hydride, high temperature deformation and thermal quench behavior were carried out.

# 2. Experimentals

#### 2.1. Specimen and sample preparation

The cladding used in this study is the commercial low Sn Zircaloy-4(Zr-1.3Sn-0.2Fe-0.1Cr) which has the length of 200mm. To simulate oxide during high burnup operation, claddings were oxidized in the 500 muffle furnace at 4 and 8 weeks to obtain cladding oxide as 20 and 50  $\mu$ m. To investigate the hydride effect absorbed during normal operations, claddings were hydrided by the gaseous method to the value of 300 and 600 ppm. Hydrogen analysis after hydriding revealed that little

variation of hydrogen content occurred along axial direction. Fresh cladding in as-received condition was used as a reference condition.

### 2.2. LOCA experiment

The detailed explanation of LOCA simulating facility was described elsewhere [3]. Two types of tests were performed, namely the transient ballooning test and the thermal quench test. Transient ballooning was conducted, where the pressurized cladding was heated at a rate of 10, 100 per second until rupture. The other is the quench test where the specimen was oxidized in a flowing steam at a desired temperature and time followed by water quenching. To determine the oxidation rate more quantitatively, the term ECR (Equivalent Cladding Reacted) was introduced to define the ratio of the converted metal thickness to initial cladding thickness.

# 3. Results and Discussions

#### 3.1. High temperature ballooning

Fig. 1 shows the total elongation of the fresh Zircaloy-4 under the transient heating condition. It showed two peaks at 800 and 1100 , which can be explained as intragranular slip and intergranular slip caused by the phase transformation of the zirconium [4]. Effect of the pre-oxide and pre-hydride on the high temperature ballooning property of the Zircaloy-4 were collated in the Fig. 1. Increase of pre-oxide thickness dropped circumferential elongation. Zirconium cladding balloons according to the mechanism of circumferential grain elongation. Microstructure of the pre-oxidized, ballooned sample revealed that pre-oxide restrains subsurface deformation. Local deformation detours along the subsurface 'strain-free' region and total deformation decreases. In terms of the pre-hydride effect, second peak transformation temperature shifts lower as the hydrogen contents increased. Hydrogen is the beta-phase stabilizer, which expands  $\beta$  phase temperature also shifts  $\alpha$  to  $\beta$ phase transformation temperature to lower value. Increase of  $\beta$  phase fraction result in the grain boundary sliding caused by 'Hard'  $\beta$  phase as hydrogen content increases, therefore increase of elongation occurs at a lower temperature in hydrided claddings.



Figure 1. Total elongation of zirconium claddings with the burst temperature. Open symbol represents small thermal ramp rate (= 10 °C/sec), closed symbol represents fast thermal ramp (= 100 °C/sec)

# 3.2. Thermal Quench behavior

Fig. 2 shows a failure map of the Zircaloy-4 cladding thermally quenched after subsequent high temperature oxidation. An open symbol represents the specimen that survived during the water quenching after a high temperature oxidation. A closed symbol represents the one that failed. From the previous study [3], threshold ECR value of fresh Zircaloy-4 cladding ranges into the value of 20%. Pre-oxide cannot act as a barrier against high temperature oxidation. Thickness of newly generated oxide in fresh cladding during LOCA is about 65 µm when exposed 1100 , 3000sec. At cladding pre-oxidized at 20 and 50 µm, oxide thickness generated during LOCA were 43 and 69  $\mu$ m. In the pre-hydrided claddings, threshold ECR decreases to the value of 17%. Absorbed hydrogen increases oxygen solubility in the  $\beta$  phase [5]. As the hydride contents increases, increase of oxygen content inside residual metal may decrease its ductility and is susceptible to fail under rapid thermal quenching such as LOCA.

# 4. Conclusions

Simulated LOCA test was performed at pre-oxided, pre-hydrided Zircaloy-4 claddings in order to evaluate the property of simulated high burn-up situation under the design based accident. High temperature ballooning and thermal quench test was carried out and the followings were driven out.

1) Pre-oxide has an influence on the deformation behavior rather than thermal quenching by restraining subsurface deformation. 2) Pre-hydride expanded the  $\beta$  phase temperature as well as oxygen solubility in the  $\beta$  phase, resulted in the increase of elongation above 1000 and the decrease of ductility during thermal quenching.



Figure 2. Failure map of zirconium claddings with respect to the oxidation temperature and time. Solid line represents ECR values calculated by Baker-Just equation. Open symbol denotes cladding survived during water quenching. Closed symbol failed.

#### Acknowledgement

This study was supported by Korea Institute of science & Technology Evaluation and Planning (KISTEP) and Ministry of Science & Technology (MOST), Korean government, through its National Nuclear Technology Program.

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