Effect of Hydrogen on Post Quench Ductility of HANA-6 Cladding Tube

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1. Introduction

With the implementation of 10 CFR 50.46c, zirconium (Zr)-based cladding tube requires testing to evaluate its performance under loss-of-coolant-accident (LOCA) conditions. Nuclear Regulatory Commission (NRC) has developed new regulatory guidance to set analytical limits appropriate for the proposed rule. According to the guidance, hydrogen absorbed within the Zr-based cladding tube during steady-state operation increases the oxygen solubility and diffusion rate in the Zr matrix, affecting the embrittlement behavior of the cladding tube under LOCA condition [1-4]. In Regulatory Guide (RG) 1.224, a post-quench ductility (PQD) analytical limit was established that correspond to the measured ductile-to-brittle transition for the Zrbased cladding tube, and a test was proposed that can measure the ductile-to-brittle transition (DBT) with unirradiated cladding tube in RG 1.223 [2-3]. Through the test method, it was confirmed that various commercial cladding tube such as Zircaloy-4, ZIRLO, M5, and Optimized ZIRLO satisfied the analytical limit [4-6].

In this study, as-received HANA-6 cladding tube was tested to evaluate PQD. Additionally, to confirm the effect of intrinsic hydrogen on embrittlement, a flash oxidation tests consisting of a 50 seconds oxidation at 1000 $^{\circ}$ C terminated by a direct quenching were performed using prehydrided HANA-6 cladding tubes [6].

2. Test Methods and Results

In this section test method to evaluate PQD and machines are described. According to RG 1.223, specimen preparation for the test consist of hydrogen injection and high temperature oxidation, a ring-compression testing was used as a method to evaluate ductility of the specimen.

2.1 Hydrogen Injection

The hydriding system consists of a tube furnace that can be heated up to 500 °C and gas injection system that can precisely inject hydrogen and argon gas into the furnace. 50 mm-long cladding tube specimens were heated and exposed to hydrogen gas so that hydrogen was absorbed into the specimens depending on the exposure time. To measure the hydrogen concentration of the hydrided specimen, both ends of the specimen were cut and measured by using vacuum hot extraction method. In this way, the specimens containing up to 1377 ppm of hydrogen were produced.

2.2 High temperature oxidation

The high temperature oxidation tester consists of a tube furnace that can be heated up to 1300 °C and a steam generator that turns pure water into steam. This tester was verified by conducting thermal benchmark test and weight-gain benchmark test suggested by RG 1.223 [2].

To evaluate PQD of as-received HANA-6 cladding tube, high-temperature oxidation test was conducted at 1200 °C for about 210 s (16 % CP-ECR) to 350 s (20 % CP-ECR) under steam atmosphere.

2.3 Flash Oxidation

As mentioned before, the flash oxidation test is to evaluate intrinsic hydrogen effect on embrittlement of cladding tubes. Due to the relatively low oxidation time and temperature, the oxidation level does not exceed 0.5 % ECR, which does not lead to significant hardening of the prior- β phase [6].

The flash oxidation test was performed on the prehydrided specimens using high-temperature oxidation tester.

2.4 Ring-Compression Test (RCT)

The universal testing machine manufactured by SHINADZU was used for ring-compression test. An oven was used to uniformly heat the specimen, and upper and lower jigs made of INCOENL were installed. Thermal benchmark test and pre-test were conducted to satisfy the equipment verification requirements suggested in RG. 1.223 [2].

The specimens oxidized at 1200 °C, and flash oxidized specimens were cut into three 8 mm ring-shaped specimen. The specimens were heated to 135 °C and then compressed at a rate of 2 mm/min. Finally, the offset strain was measured through a load-displacement graph generated during the test, and the ductility of the specimen was evaluated.

2.5 Post-quench ductility of as-received HANA-6 cladding tube

Fig. 1 shows the PQD evaluation result for HANA-6 cladding tube. The blue line in the graph represent the ductile-to-brittle transition (DBT) limit, meaning that the point above the line means ductile and below it means brittle. According to the results, DBT was observed in as-received HANA-6 cladding tube at oxidation level of $18 \sim 19$ % ECR, similar to other commercial cladding tubes [4-6].



Fig. 1. Result of ductile-to-brittle transition of as-received HANA-6 cladding tube.

2.6 Intrinsic hydrogen effect

RCT results of flash oxidized specimen were shown in Fig. 2. The ring specimen was compressed up to 7 mm, and the prehydrided specimen with less than 1000 ppm of hydrogen experienced a slight load drop but were compressed to the end without any facture. On the other hand, in the case of the specimen with more than 1000 ppm of hydrogen, the time to facture became shorter as more hydrogen concentration. After the RCT, the specimen was observed with a light optical microscope (LOM) as shown in Fig. 3, the specimen with minor load drop (404 ppm) was shown a micro crack inside of it. Similar to the LOM results, as shown in Fig. 4 (a), a load drop was observed in specimens with more than 400 ppm in hydrogen. As well, low facture energy was calculated for specimen with more than 1000 ppm of hydrogen as shown in Fig. 4(b). Fig. 4 (c) shows that the offset strain tends to decrease as the hydrogen concentration in the cladding tube increases, but the analytical limit from RG 1.224 is sufficiently satisfied.

From the results, it was found that HANA-6 cladding material would be embrittled when it has more than 1000 ppm in hydrogen. Therefore, it is considered that the effect of intrinsic hydrogen on embrittlement is not significant.



Fig. 2. RCT results of flash oxidized prehydrided specimen



Fig. 3. LOM data of specimen with (a) micro and (b) major cracks



Fig. 4. Comparison graph of RCT results according to hydrogen concentration in HANA-6 cladding tube.

3. Conclusions

PQD evaluation of HANA-6 cladding tube was performed via RCT. The results show a similar DBT with other commercial cladding alloys such as ZIRLO, M5, Zircaloy-4 [4-6]. Furthermore, flash oxidation test with prehydrided HANA-6 cladding tube was conducted to evaluate intrinsic hydrogen effect on embrittlement. As a result, it was confirmed that ductility of HANA-6 cladding tube with less than 1000 ppm hydrogen contents was maintained, and even the cladding tubes with more than 1000 ppm of hydrogen also maintain enough strain until fracture occurs.

Recently, PQD evaluation is being conducted with prehydrided and irradiated HANA-6 cladding tube, and the test results will be presented at KNS spring meeting.

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