Effects of Hydride and microstructure on the PCI behaviors of the nuclear fuel claddings

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1. Introduction

Nowadays, many power plants adopt high burn-up operation which includes high power, enlarged fuel cycle, and so on. In this high burn-up condition, PCI and hydrogen embrittlement were serious problems in relation to cladding safety. Firstly, as the burn-up increase, the diameter of the cladding decreases but the outer diameter of the fuel pellets increases so that the PCI behavior will be very severely. In addition, the concentration of iodine inside the fuel rod increased so that the possibility of both a mechanical and a chemical interaction which causes a ISCC become more severe [1-4].

The cladding tubes of the LWR pick up part of the hydrogen evolved during reactor operation by coolant-metal corrosion reaction. Hydrogen concentration in the cladding tube increased sharply with the burn-up. Hydrogen present in excess of terminal solid solubility (TSS) precipitates out as hydride phase, which affects the PCI behaviors of high burn-up nuclear fuel claddings. Therefore, in this study, we focused on the time-to-rupture behavior of pre-hydrided nuclear fuel cladding in high temperature and high-pressure iodine environment. The rupture time of un-hydrided and pre-hydrided cladding was evaluated and was compared.

2. Experimental Procedure

The specimens for this study were cut from a commercial grade low-tin Zircaloy-4 (Zr–1.3Sn–0.2Fe–0.1Cr). Their outer and inner diameters were 9.50 mm and 8.36 mm, respectively. Their length was 130 mm. They were all in as-received states in a stress-relieved condition. To investigate the effect of the microstructure, a specimen was heat-treated at 620°C for 3 h to have a fully re-crystallized structure. Using hydrating methods for minimum change of cladding surface and microstructure, various concentration of hydrided specimen were prepared and tested.

Test specimen was put inside an autoclave, and then a medium, which was pure argon mixed with iodine, was pressurized inside the cladding after reaching a constant test temperature of 350°C. The iodine used in this study, which had a purification of 99.99%, was supplied by Aldrich. To minimize oxidation at the outer surface, we evacuated the autoclave and filled it with argon gas before each test. The test was stopped whenever a leakage occurred around the test specimen or, if the specimen did not leak, after 100 h of testing. The iodine concentration was kept constant at 1.5 mg/cm^2.

3. Results and Discussion

Fig. 1 shows the hydride morphology of the stress-relieved (SR) and re-crystallized (RX) Zircaloy-4 specimen. For the specimen of SR structure, the direction of hydride platelet is same as to the circumferential; however, the hydride platelet of a RX structure is randomly distributed. This figure implies that the shape of grain boundary plays an important role in the direction of hydride platelet.
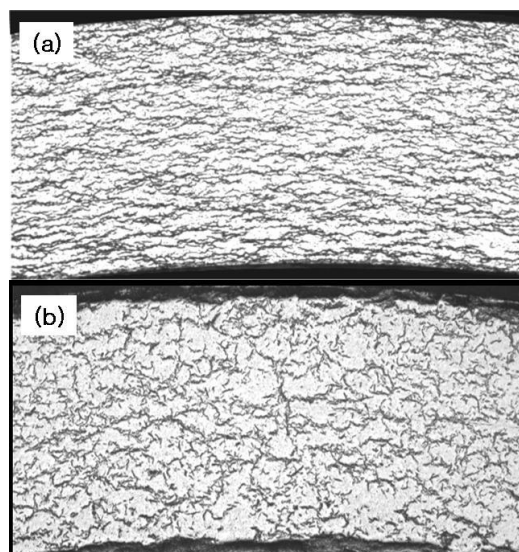

Fig. 1. Hydride morphology of the (a) SR and (b) RX Zircaloy-4 specimen

Fig. 2 shows the time-to-rupture behavior of un-hydrided and pre-hydrided Zircaloy-4 cladding, and Fig. 3 shows the comparison of the rupture time at the hoop stress of 300 MPa. For SR Zircaloy-4, the rupture time of hydrided specimen decreased rather than that of un-hydrided specimen. It means that hydride increase the ISCC crack velocity because of its embrittlement. We reported that ISCC crack propagation rate and K_{ISCC} of the un-hydrided RX Zircaloy-4 is superior to those of un-hydrided SR one [5-7]. But the rupture time of the pre-hydrided RX Zircaloy-4 is seriously decreased

rather than that of pre-hydrided SR one. The direction of hydride of SR structure is circumferential, but that of RX one is random as shown in Fig. 1. Texture of two samples is similar to each other; therefore, the radially distributed hydride did not form along hydride habit plane, but formed along the grain boundary. The radially distributed hydride is parallel to the ISCC crack direction, which decrease the rupture time severely. From this result, we can conclude that the microstructure plays a critical role in the PCI behavior of pre-hydrided Zircaloy-4.
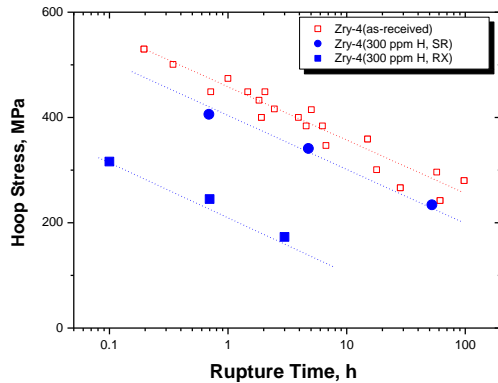

Fig. 2. The time-to-rupture behavior of the as-received(SR) and pre-hydrided (SR and RX) Zircaloy-4 cladding.
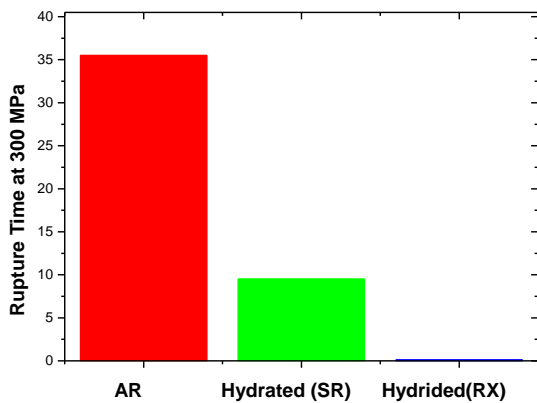

Fig. 3. Comparison of the rupture time at the hoop stress 300 MPa.

Recently, we have proposed that grain-boundary pitting coalescence (GBPC) and pitting-assisted slip cleavage (PASC) models are appropriate for evaluating crack nucleation and growth in a commercial grade of nuclear fuel cladding without hydriding [5-7]. The main factors affecting the crack initiation step were pitting nucleation, growth, and agglomeration in that the pits are preferentially formed around a grain-boundary (GB) as a result of a local corrosion reaction by iodine. When

these pits were combined in a crystallographic slip plane, a cleavage crack was generated in that grain.

More detail study on the relationship between hydride and grain-boundary pitting will be evaluated in further study.

4. Conclusions

Using hydrating methods for minimum change of cladding surface and microstructure, pre-hydrided Zircaloy-4 specimen were prepared and time-to-rupture behavior was estimated in a high temperature iodine environment. The rupture time of pre-hydrided SR Zircaloy-4 was decreased by 35% as compared with that of un-hydrided one. Moreover, the rupture time of pre-hydrided RX specimen was severely decreased rather than that of SR one. It is considered that RX microstructure could be inferior to SR one for a high burn-up nuclear cladding.

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