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ISCC Properties of HANA claddings

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1. Introduction

Nowadays, most nuclear power reactors are adopting a high burn-up to increase their fuel economy, where the refueling cycle of the fuel bundles is extended. As a fuel burn-up increases, the possibility of an iodine-induced stress-corrosion cracking (ISCC) is increased.

KAERI had developed six kinds of advanced Zr alloy claddings named HANA (High performance Alloy for Nuclear Application) in order to meet the global demand for an extension of the fuel discharge burn-up to more than 70 GWd/MtU. In this high burn-up condition, the diameter of the cladding is decreased but the outer diameter of the fuel pellet is increased so that the PCI behavior will be very severely. Also, the concentration of iodine inside the fuel rod increased so that the possibility of both a mechanical and a chemical interaction which causes a SCC become more severe. So the PCI resistance of HANA cladding needs to increase. This study has been done to estimate the PCI behavior of HANA-4(Zr-1.5Nb-0.4Sn-0.2Fe-0.1Cr) and HANA-5 (Zr-0.4Nb-0.8Sn-0.3Fe-0.1Cu) claddings.

2. Experimental

The specimens for this study were cut from a HANA-4 (Zr-1.5Nb-0.4Sn-0.2Fe-0.1Cr) and HANA-5(Zr-0.4Nb-0.8Sn-0.3Fe-0.1Cu) cladding. Their outer and inner diameters were 9.50 mm and 8.36 mm, respectively. Their length was 130 mm. They were all used in their as-received states namely a stress-relieved condition. To investigate the effect of the microstructure, a specimen was heat-treated at 620^{o}C for 3 hours to have a fully recrystallized structure then it was used for the ISCC test. A specimen with an initial crack inside its surface was used in the test. A pre-crack was created by the fatigue cracking method which Lemaignan [1] employed.

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^{o}C. The iodine used in this study, which had a purification of 99.99%, was supplied by Aldrich. The iodine concentration was kept constant at 1.5 mg/cm^2. After the test, the specimen was examined using a scanning electron microscope (SEM) to determine the actual crack propagation depth during the ISCC test, and then the crack propagation velocity was calculated. ISCC crack propagation rate with respect to the applied K_I was evaluated to determine the threshold stress intensity factor (K_{ISCC}). The K_I value was adjusted so that the stress state around a crack tip was a plane strain condition. The detailed experimental procedure regarding a fatigue pre-cracking and the ISCC procedure can be found in previous papers [2, 3].

3. Results and Discussion

Recently, grain boundary pitting coalescence (GBPC) and pitting-assisted slip cleavage (PASC) models have been proposed to explain the ISCC crack initiation and propagation mechanism of a Zircaloy-4 surface. This is depicted in Fig.1 [2]. When iodine is adsorbed into the zirconium cladding, the zirconium bond at the grain boundary (GB) will be weakened, so its surface energy will be reduced [4]. In addition, any free iodine can react with the zirconium to form solid iodides and a gaseous zirconium tetra-iodide (ZrI_4) [4]. This gaseous ZrI_4 can be decomposed easily into the iodine and Zr at a strained surface by an applied hoop stress, thus pits are formed due to a localized attack on the GB. Therefore, the pits or pitting clusters can become a crack nucleation site. The ISCC crack is nucleated by pits at first and they cluster with each other along the grain boundary, and then the crack grows by an IG mode during an early step of a cracking. This pitting mechanism on the surface of HANA-alloy explains well about the GBPC model.

In the early stage of a cracking, an ISCC crack nucleates at a grain-boundary by a GBPC mechanism that is purely an IG mode. During a crack growth and propagation, TG_c (TG cracking by cleavage) and TG_f (TG cracking by fluting) modes take place with an increase of the stress intensity at a crack tip. TG_c takes place in low stress intensity with a GB pitting and a cleavage habit plane, however, a TG_f takes place in high stress intensity without the aid of a cleavage habit plane. For the SR microstructure, the stress intensity boundary-correction factor on the GB pitting is higher than that of the RX one because of its laminar type grain structure. Therefore TG_f-TG_c is the main factor for the cracking mode during a crack propagation for the SR microstructure; however, IG-TG_c is the main factor for the RX microstructure which reduces the propagation rate. So, the interactions among the stress intensity, the direction of the crystallographic plane, the pitting resistance at the GB and the grain shape play important roles in a crack propagation behavior.

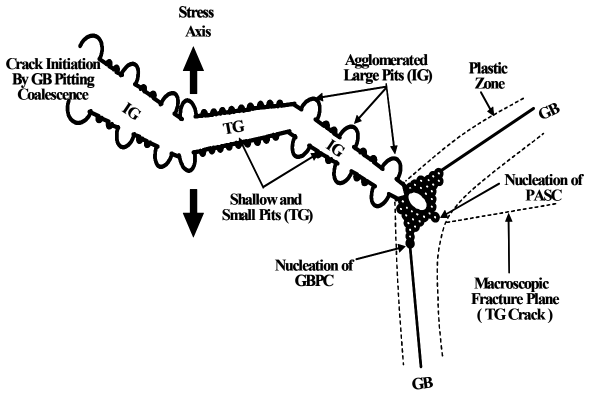

Fig.1 Schematic diagram showing an ISCC crack propagation by grain-boundary pitting coalescence (GBPC) and pitting-assisted slip cleavage (PASC) models.

Fig.2 shows the crack propagation rate of the Zircaloy-4 and HANA-5 claddings with SR and RX structures with respect to the applied K_I. For Zircaloy-4, the K_{ISCC} value and the crack propagation rate in the region II (at 6.0 $MPa.m^{0.5}$) are 3.3 $MPa.m^{0.5}$ and 1.3 x 10^{-6} m/sec, respectively. For the HANA-5 cladding, they are 4.8 $MPa.m^{0.5}$ and 4.0 x 10^{-8} m/sec, respectively. It means that the ISCC resistance of the HANA-5 cladding is much higher than that of Zircaloy-4.

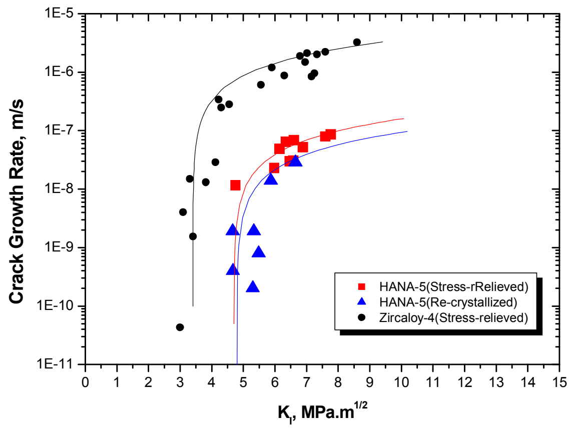

Fig.2 Plots of da/dt for Zircaloy-4 and HANA-5 cladding.

Test results for Zircaloy-4, HANA-4 and HANA-5 claddings are summarized in Table 1. The crack propagation rate at 6.0 $MPa.m^{0.5}$ of Zircaloy-4 is 100 times higher than that of the re-crystallized HANA-4 cladding. The increase of the ISCC resistance for HANA claddings result from adding of Nb element in the Zr-alloy. Many pitting clusters appear on the IG surface of the Zircaloy-4, but small and shallow pits appear on the IG surface of the HANA claddings. Some pitting clusters appear on the border of the TG surface in Zircaloy-4, but no pitting appears in the HANA claddings. Thus, the addition of Nb to Zr-alloy seems to suppress the pitting generation around the grain boundary and increase the ISCC resistance.

Table 1 Comparisons of the threshold stress intensity factors and crack propagation rates between Zircaloy-4 and HANA claddings.

Alloy	Micro-structure	K_{ISCC} ($MPa.m^{0.5}$)	da/dt(at 6$MPa.m^{0.5}$) (m/sec)
Zircaloy-4	SR	3.4	1.3E-06
HANA-4	SR	4.5	8.1E-08
	RX	4.8	1.0E-08
HANA-5	SR	4.8	4.0E-08
	RX	4.8	2.0E-08

4. Conclusions

The grain-boundary pitting coalescence (GBPC) and pitting-assisted slip cleavage (PASC) models explain the ISCC crack initiation and propagation behavior well in an iodine environment for Zircaloy-4 and HANA claddings. The ISCC resistance of HANA-4 and HANA-5 claddings is higher than that of Zircaloy-4 in all the conditions. Especially, the crack propagation rate of the recrystallized HANA-4 cladding is 100 times lower than that of Zircaloy-4. The increase of the pitting resistance at the grain boundary plays an important role in reducing the crack propagation rate of HANA claddings.

Acknowledgement

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