A Microscopic Analysis of a Cleavage Crack Propagation of a Nuclear Fuel Cladding in an Iodine Environment

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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 diameter of a cladding is decreased, while the outer diameter of a fuel pellet is increased. Eventually the pellet and the cladding contact each other after some cycles have elapsed. Also, during such a burn-up, the concentration of the iodine inside a fuel rod gradually increases to cause a stress-corrosion cracking (SCC). The possibility of both a mechanical and a chemical interaction which cause an iodine-induced stress-corrosion cracking (ISCC) is also increased.

Recently, it has been proposed that grain-boundary pitting coalescence (GBPC) and pitting-assisted slip cleavage (PASC) models are appropriate for the modeling of an ISCC crack nucleation and growth in Zircaloy-4 [1]. However, the reason why a re-crystallized structure has a lower crack propagation rate than a stress relieved one, has not been clearly explained. This study is aimed at supporting these models by using a quantitative and microscopic analysis of ISCC test results as well as detailed observations of the fractured surfaces of a pre-cracked cladding tube, especially, the effects of the microstructure was focused on.

2. Experimental

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 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 [2] 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 (KISCC). 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 [1, 3].

3. Results and Discussion

Fig.1 shows the schematic crack shape, crack propagation mode and SEM fractographs of a fully recrystallized (RX) Zircaloy-4 specimen which was pre-cracked and pressurized at 145 MPa for 12.3 hr in an iodine environment. At an early stage, the crack was propagated by an intergranular (IG) ISCC cracking, at a later stage, however, the crack propagation mode was switched to a cleavage-like transgranular (TG) cracking. However at an intermediate stage (a/t = 0.48 ~ 0.85), the crack was propagated by a mixed IG and TG mode. In this region, a TG crack that passed through one grain could not penetrate further into a neighboring grain so it changed its direction along the grain boundary to reveal an IG crack along several grains. A crack by the TG mechanism will appear after some IG cracks have proceeded it.

The crack is propagated by a TG mode with a fluting and a cleavage at an early stage for the SR Zircaloy-4. A cleavage and a fluting were arranged in an alternate position, and nanometer scale pits were formed on the whole fracture surface. It means that a cleavage took place by the action of nanometer scale pits in spite of a low K_I. In addition, the TG cracking behavior depended on the microstructure.

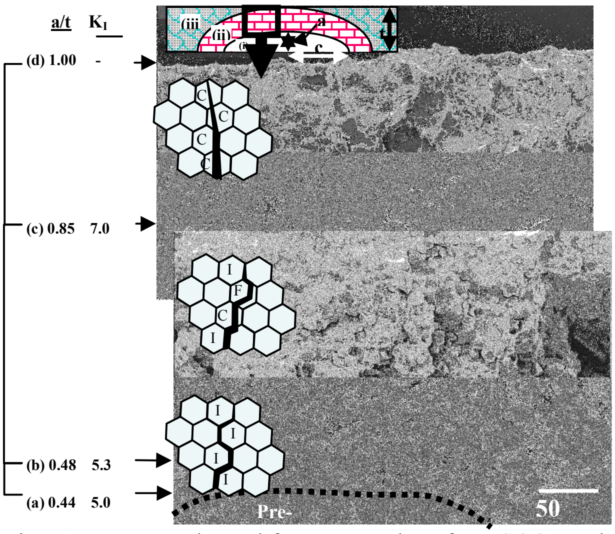

Fig. 1. Fractographs and fracture modes of an ISCC crack for the re-crystallized Zircalloy-4 cladding pressurized in an iodine environment at 350°C; (i) Fatigued pre-crack, (ii) ISCC (iii) ductile fracture after test; I=IG, C= TG by cleavage, F=TG by fluting .

Newman[4] has presented a stress-intensity factor equation for semielliptical surface cracks in finite elastic plates subjected to tension or bending loads. We tried to use this equation for an RX and an SR grain, which are 6 X 6 X 6 μm^3 for the RX and 30 X 2.4 X 3 μm^3 for the SR, respectively.

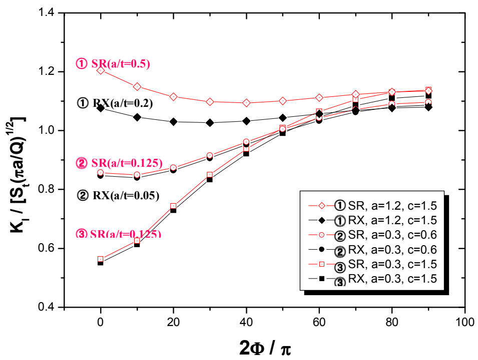

Fig. 2. Typical results of the stress intensity factors for the surface crack formed by the GBPC mechanism on the SR and RX grain surfaces.

Fig.2 shows the stress intensity boundary-correction factor ($K_I / S_t \sqrt{\pi a / Q}$) applied at the surface cracks. In spite of the same size of the surface crack, the stress intensity of the SR grain is always higher than that of the RX grain. The larger the surface crack is, the higher the stress intensity factor applied to the SR grain along the ϕ = 0 direction is, which means the possibility of a cleavage cracking increases in the case of the SR grains. Since the laminar type SR grains have a larger value of a/t than that of the RX ones, their surface crack in the SR structure suffers from a higher stress intensity than that in the RX one. In spite of the small pits or a surface crack of a nanometer scale, the a/t value in the SR grain is so large that it can initiate a cleavage or a fluting. If a surface crack becomes parallel to a cleavage habit plane, a cleavage cracking takes place in a grain. Otherwise, a fluting crack takes place in a grain.

4. Conclusions

From a microscopic analysis regarding ISCC test results as well as detailed observations of the fractured surface of a pre-cracked Zircaloy-4 cladding with either SR or RX microstructures to investigate the ISCC process, the following can be summarized.

During the ISCC cracking, it was revealed that the grain shape and cleavage habit plane played important roles in the rate determining step (RDS) by generating a crack in the grain, which resulted in an IG-TG$_c$ (TG cracking by cleavage) or a TG$_f$ (TG cracking by fluting)-TG$_c$ cracking mode. An IG-TG$_c$ cracking took place for the RX microstructure through a GB pitting, however, a TG$_f$-TG$_c$ cracking did occur for the SR one which increased its propagation rate.

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Reference

[1] in press as: S.Y. Park, et al., J. Nucl. Mater. (2007), doi:10.1016/j.jnucmat.2007.03.258.
[2] C. Lemaignan, Int. J. Pres. Ves. & Piping, 15 (1984) 241
[3] S.Y. Park, J.H. Kim, B.K.Choi, Y.H. Jeong, Met. Mater. –Int, 13 (2007) 155
[4] J.C. Newman, Jr and I.S. Raju, Engineering Fracture Mechanics, 15 (1981) 185