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Stress-induced reorientation of hydrides and mechanical properties of Zircaloy-4 cladding tubes

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Abstract

Stress-induced reorientation of hydrides and its effect on the stress–strain response of Zircaloy-4 cladding tubes were investigated. The reorientation of hydrides along the radial direction was most pronounced if the tube was cooled from 300 to 200 °C under circumferential loading. Reorientation occurred much less frequently at either higher (cooled from 400 to 300 °C) or lower (cooled from 200 to 100 °C) temperature range. The population of radial hydrides in R43H7 (which was cooled from 400 to 300 °C and maintained at 300 °C for 7 h) increased drastically during annealing at 300 °C, suggesting time dependent stress-aided dissolution of circumferential hydrides and reprecipitation of radial hydrides. The drastic decrease of the strength and the complete loss of the ductility were observed in R32AC and R43H7.

1. Introduction

Zircaloy-4 is known to have satisfactory mechanical strength and corrosion resistance and has been used as nuclear fuel cladding materials [1–5]. The life of zirconium alloy claddings, however, can be limited by the degradation of mechanical properties caused by cracking of brittle hydrides [6–10]. When the alloy is used in a nuclear reactor, its hydride is formed from external hydrogen sources such as waterside corrosion, dissolved hydrogen in coolant water and water radiolysis, and from internal sources such as hydrogen content in fuel pellets and moisture absorbed by the uranium dioxide fuel pellet [11].

The extent of hydride embrittlement depends not only on the quantity of hydride present but also on its morphology and in particular the orientation of hydrides with respect to the applied stress [3,4]. For a tubular component internally pressurized in service, it is desirable to have the hydride platelets oriented with their major axis in the circumferential direction. Hydride orientation is to a large extent determined by the manufacturing process, which has a great influence on the texture and the shape and size of grains [12–14]. The cold tube reduction process (pilgering), which is used in the fabrication of cladding tubes, fortunately produces material with a strong tendency to form circumferentially oriented hydrides. However, hydrides are usually dissolved at high temperatures, and hydrogen in solid solution diffuses in the presence of concentration, temperature and

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stress gradients, and precipitates out as radial hydrides under the presence of the circumferential stress. In this study, the reorientation of hydrides and its effect on the mechanical properties of Zircaloy-4 tubes were studied.

2. Experimental procedure

Zircaloy-4 tubes used in this study were supplied by the Korea Nuclear Fuel Company. They were manufactured in the same way as the typical stress-relieved fuel claddings, and the basal poles were oriented at about 30° from the tube radial direction [10,15–18]. The tubes were cold-pilgered in three steps with the reduction of diameter in the final step equal to 83% and then annealed at 460 °C for 7 h. The grain size in the transverse section was 5.6 μm and the length in the transverse section was 7.9 μm. The outer and inner diameters of the tubes were 9.4 and 8.4 mm, respectively. The chemical composition of the tube was Zr–1.34Sn–0.22Fe–0.11Cr–0.13O in weight percentage.

The tubes were charged with hydrogen using the high temperature cathodic hydrogen charging method [10]. The electrolyte solution used for hydrogen charging was 500 mg $NaHSO_4$ (mg) + 50 mg Na_2SO_4 + 10 mg K_2SO_4 + 100 ml H_2O. A direct current was supplied galvano-statically to Zircaloy-4 specimens with the current density of 0.5 A/cm^2. In order to stabilize hydrogen in the specimens as hydride form, vacuum annealing was carried out at 400 °C for 3 h after chemically cleaning of the specimen surface with acetic acid. After annealing, the tubes were cooled in air. The cooling rate for air-cooling was approximately 126 °C/s The amount of hydrogen pick-up after charging, assuming no loss of hydrogen, is 1260 wt.ppm [8]. The hydrogen concentration was measured using the inert gas fusion thermal conductivity detection method by LECO hydrogen analyzer RH404 (St. Joseph, Michigan, USA) and the real hydrogen concentration was 220 wt.ppm. The difference between the amount of hydrogen pick-up and the measured hydrogen concentration can be attributed to the difficulty of stabilization of hydrogen above the solubility limit and the release of over-charged hydrogen during annealing.

Ring specimens with a width of 4 mm were cut transversely from the cladding tube and the circumferential stress was applied using a special grip for reorientation treatments and tensile tests. Fig. 1 shows the schematic configuration of the ring specimen and two half-cylinders that open and strain the ring specimen. As shown in this figure, 1 mm wide space was given between two half-cylinders to allow free plastic flow during tensile testing. The diameter of the half-cylinders in Fig. 1 was 8.35 mm. Loading of the sample was applied using a United Testing Machine, SFP 10 (Huntington Beach, CA, USA) with a three-zone split furnace. Various reorientation treatments were employed in this study. In R21AC, the specimen was heated to 200 °C and cooled to 100 °C at a cooling rate of 4 °C/min under loading and air-cooled to room temperature after unloading when the temperature was lowered to 100 °C. The loading was applied up to (67.5 ± 2.5)% of the yield stress at each temperature when the designated maximum temperature was reached. With cooling, the applied stress was continuously increased to keep the stress level at (67.5 ± 2.5)% of the yield stress throughout the cooling process in the furnace (from 200 to 100 °C in case of R21AC). Likewise, in R32AC, the specimen was heated to 300 °C and cooled to 200 °C in the furnace at a cooling rate of 4 °C/min under loading and air-cooled to room temperature after unloading and, in R43AC, the specimen was heated to 400 °C, cooled to 300 °C in the furnace under loading and air-cooled to room temperature after unloading. In R43H7, the specimen was heated to 400 °C, cooled to 300 °C at a cooling rate of 4 °C/min under loading, maintained at 300 °C for 7 h under loading and air-cooled to room temperature after unloading to examine the time dependent reorientation of hydrides.

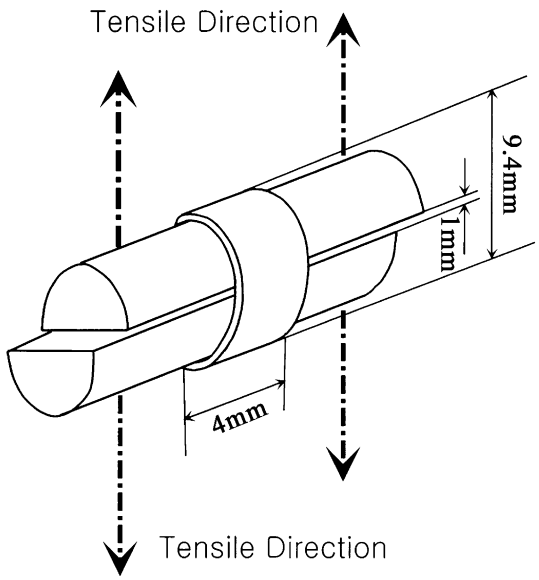

Fig. 1. Schematic configuration of tensile testing sample and grip.

Recently, Singh et al. [1] reported that the threshold stress for reorientation of hydrides decreased and the ratio of the threshold stress (σ_{th}) to the yield stress (σ_{ys}) (σ_{th}/σ_{ys}) decreased with increasing temperature. For example, σ_{th}/σ_{ys} was reported to be approximately 0.55 and 0.4 at 227 °C and 327 °C, respectively [1]. The applied stress level in the present study was maintained well above the threshold stress reported for reorientation