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Hydride reorientation in Zircaloy-4 cladding

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Abstract

The formation of radial hydrides in stress-relief annealed Zircaloy-4 cladding was studied. Specimens were firstly hydrided to different target hydrogen levels from 100 to 600 wt ppm and then thermally cycled in an autoclave under a constant hoop stress to form radial hydrides by a hydride reorientation process. The effect of thermal cycling on the hydride reorientation was more significant than that of isothermal treatment. Based on the experimental data, a thermodynamic model was proposed to elucidate the stress reorientation behavior of hydrides in Zircaloy cladding. According to the model, the bounds of stress and temperature to stress reorientation of hydride precipitates were developed. The threshold stress for hydrides to reorientation was a function of solution temperature and specimen hydrogen concentration.

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1. Introduction

The mechanical properties of Zircaloy fuel cladding can be adversely affected by the presence of hydrides, especially when they are oriented towards the radial direction of the tubing (i.e. radial hydride) [1–4]. The formation of radial hydrides in Zircaloy materials is closely related to the fabrication history, texture and stress [5–8]. In order to retain sufficient ductility to keep its integrity during reactor service, Zircaloy fuel cladding tube is manufactured under well controlled condition to ensure that only circumferential hydride platelets are to develop during reactor service. However, radial hydrides can be formed, by a reorientation process, when a specimen is cooled down under stress from temperatures at which hydrides are dissolved. As is often the case, Zircaloy fuel cladding tends to be subjected to larger internal pressure and to have higher hydrogen content as its burnup increases. The higher hoop stress will make these hydrides more susceptible to stress reorientation when fuel cladding undergoes a temperature variation during reactor operation or under dry storage conditions [9–12]. Therefore, the factors determining the preferential orientation of hydride precipitates and the effects of radial hydrides on the integrity of cladding materials have been examined extensively.

The occurrence of hydride reorientation in Zircaloy fuel cladding usually involves the dissolution of circumferential hydrides and the formation of radial hydrides. Of the two processes, precipitation dominates the hydride reorientation process mechanistically. Hydride precipitation is a complicated function of the solubility of hydrogen in cladding materials, cladding hydrogen concentration, stress state, temperature, cooling rate and thermal cycling. So when characterizing the stress reorientation behavior of hydride precipitates in cladding materials, it is necessary to take into account these factors separately and synergistically. In this work, specimens of hydrogen levels between 130 and 600 wt ppm were thermal-cycled under a constant hoop stress to form radial hydrides. The dependence of the

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stress reorientation on the hydrogen concentration, thermal cycle number and solution temperature was investigated experimentally. A thermodynamic model for evaluating the susceptibility to radial hydride formation in the stress-relief annealed (SRA) Zircaloy-4 cladding was proposed and verified with the experimental results. With the model, the bounds of stress and temperature to stress reorientation of hydride precipitates were developed.

2. Experimental

2.1. Material and hydriding process

Stress-relief annealed (SRA) Zircaloy-4 cladding with an outside diameter of 9.5 mm and wall thickness of 0.58 mm was used. Its chemical composition is given in Table 1. Cladding tube, cut into 13 cm lengths, was first uniformly hydrogen-charged by a thermal cycling process. The specimen was encapsulated with a pre-determined amount of pure hydrogen in a Pyrex capsule of sufficient volume such that a low hydrogen partial pressure could be obtained to avert the formation of hydride layers. The encapsulated cladding specimen was then thermally cycled between ~200 °C and 300 °C for a certain number of cycles, depending on the target hydrogen concentration level. The heating and cooling rates were at 3 °C/min and 2 °C/min, respectively [13]. The target hydrogen levels ranged from 100 to 600 wt ppm. Hydrogen concentrations of Zircaloy-4 cladding specimens were determined by an inert-gas fusion method using a LECO RH-404 hydrogen determinator. Typically, hydrides were oriented in the circumferential direction and homogeneously distributed across the cross-section of the cladding specimen.

2.2. Hydride reorientation experiment

In order to obtain radial hydrides, the as-hydrided specimen was further subjected to thermal cycling in an autoclave under a constant hoop stress by regulating the differential pressure between its internal and external pressures with a constant differential pressure control system [13]. The tube was heated to 400 °C under a constant differential pressure of 20.7 MPa that was equivalent to a hoop stress of 160 MPa being applied on the tubing wall. After solution-annealed at 400 °C for 2 h, the specimen was slowly cooled down at a cooling rate of 1 °C/min to 170 °C to make up one thermal cycle. The experimental parameters of solution temperature and cooling rate were chosen to simulate dry storage conditions as close as possible. In this work, cladding tubes were treated under the same thermal and pressure conditions but different cycling numbers, i.e. 1, 2, 4, 8 and 12 cycles, to obtain specimens with various fractions of radial hydride precipitates. The reorientation experiment run was conducted in duplication for each of combination conditions of hydrogen concentration and cycle number. Besides, to study the effect of solution temperature on the hydride reorientation behavior, some specimens were solution annealed at the temperature 300 °C or 450 °C for 2 h and then cooled down to 170 °C at a rate of 1 °C/min. Afterwards, the specimens were further furnace-cooled to room temperature, not thermally cycled.

Table 1
Chemical compositions of Zircaloy-4 cladding tube (wt%)

Sn	Fe	Cr	O	N	C	H	Zr
1.26	0.22	0.12	0.13	0.0029	0.01	0.0007	Balance

2.3. Analysis of hydride orientation

Because the majority of hydride traces observed was either along the specimen hoop direction or perpendicular to it, hydride stringers were classified into two groups: circumferential and radial hydrides. The former was defined as the clusters with their precipitate planes oriented

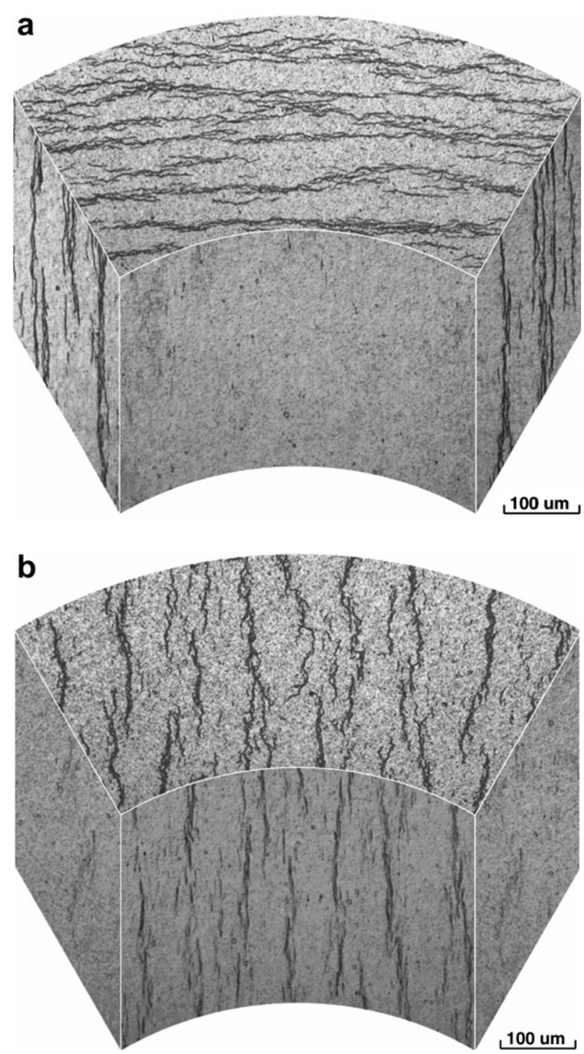

Fig. 1. Micrograph showing orientation of hydrides in Zircaloy-4 cladding of ~230 ppm: (a) as-hydrided and (b) after 8 cycles of thermal treatment.