Effect of Heat Treatment on Delayed Hydride Cracking in Zr-2.5 Wt Pct Nb

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The effect of heat treatments on delayed hydride cracking (DHC) in Zr-2.5 wt pct Nb has been studied. Crack velocities were measured in hydried specimens, which were cooled from solution-treatment temperatures at different rates by water-quenching, oil-quenching, liquid-nitrogen quenching, and furnace cooling. The resulting hydride size, morphology, and distributions were examined by optical metallography. It was found that fast cooling rates, which produce very finely dispersed hydrides, result in higher crack growth rates and a stronger dependence of the crack velocity on the applied-stress intensity factor. Also, the incubation period before cracking commences was found to be relatively short for specimens with fine hydrides, whereas specimens with coarse hydrides required considerably longer incubation periods. These results can be explained by rapid growth of preexisting hydrides within the crack-tip plastic zone. In addition, different solution temperatures were used to investigate the effect of the continuity of the grain-boundary phase (β-phase) on the crack velocity. Transmission electron microscopy was used to examine the structure of this grain-boundary phase. It was found that for heat treatments, which destroyed the β-phase continuity, the crack velocity was significantly reduced, as would be expected from the theory of enhanced diffusion through grain boundaries.

I. INTRODUCTION

The embrittling effect of internal hydrogen in zirconium and other hydride-forming metals has been extensively studied in the past and the mechanisms of the embrittlement are fairly well understood.1-4 The embrittlement in these exothermic absorbers is associated with the precipitation of brittle hydrides below the terminal solid solubility (TSS) temperatures. The presence of the hydrides results in a reduction in ductility2 and fracture toughness3,4 of the material.

In addition to gross embrittlement, hydrides are also responsible for a delayed failure mode via slow, subcritical, crack growth. The first example of this in a zirconium alloy, namely Zr-2.5 wt pct Nb, was reported by Simpson and Ells,5 in an experimental nuclear fuel sheath. Cracking occurred in the weld region after storage at room temperature over a two-year period. A second nuclear-related delayed cracking problem has emerged in some Zr-2.5 wt pct Nb pressure tubes in CANDU™ reactors.6 Through-wall cracks developed in high-stress regions associated with the end fittings. Occurrence of the pressure tube failures led to extensive research efforts aimed at obtaining a better understanding of mechanisms responsible for such delayed failures. As a result, a good understanding of the process has been obtained. The mechanism has been studied both theoretically7-10 and experimentally.10-15 Delayed hydride cracking (DHC) is believed to be produced by a diffusion-controlled precipitation and growth of hydride at a crack tip (under the action of the stress gradient), followed by rapid crack propagation. The crack-tip advances through the embrittled region, then arrests in the ductile matrix, until a new hydrided zone is formed at the crack tip. The process repeats itself, in a step-wise manner, resulting in slow crack growth.

The theoretical model7-10 which was developed to describe the process, assumes a relatively coarse distribution of hydrides, where the plastic zone at the crack tip is initially free of hydride precipitates. Hydrides present outside this highly stressed zone dissolve to provide a source of hydrogen for the hydride precipitate at the crack tip. The model was later improved10,16,17 to include the effect of internal stress (misfit stress associated with the precipitated hydrides) on the flux of hydrogen to the crack tip. The misfit stresses are generated in the vicinity of the hydride as a result of the hydride-matrix misfit (hydrides undergo a volume increase of about 17 pct upon precipitation). The misfit strains can be accommodated by elastic distortion or plastic deformation (generation of misfit dislocations) of the matrix. The associated strain energy has been shown to have a significant effect on the TSS.16 The elastic part (reversible) of the accommodation energy tends to promote hydride dissolution. This results in an increase of the equilibrium, or stress free, TSS (shift in the TSS) during cool-down. The accommodation plastic energy (irreversible), in contrast, tends to facilitate the precipitation of the hydride by substantially reducing the elastic accommodation energy. The hysteresis between cool-down TSS and heat-up TSS, usually observed in exothermic metal hydrogen systems, has been explained by these strain energy accommodation effects.17

In practice, we consider that the coarse distribution of hydrides assumed in the model is not always a totally adequate representation of hydrides in pressure tubes under CANDU™ reactor operating conditions. This is because repeated dissolution and reprecipitation of the hydride will occur during service, due to start-up and shut-down of the reactors, which might give rise to a change in size and distribution of the hydride. Indeed, there is experimental evidence18,19 suggesting that significant hydride refinement and redistribution occurs in zirconium alloys following such thermal cycling.

The present work was thus undertaken with the objective of studying systematically the combined effect of hydride
morphology (size and distribution) and the elastic misfit of the hydride precipitates on the DHC crack velocity in CANDU™ pressure tubes. In addition, we investigated the effect of the morphology of the grain boundary β-phase in Zr-2.5 wt pct Nb on the kinetics of the crack growth. The microstructure of Zr-2.5 pct Nb consists of elongated α-zirconium grains surrounded by a continuous grain boundary network of niobium-rich β-phase. This grain boundary phase was previously found to be a short-circuit path for decomposition and break-up upon aging at temperatures higher than 300 °C. The resulting loss of continuity of the β-phase was previously found to significantly decrease the crack velocity.

II. EXPERIMENTAL METHOD

A. Material and Specimen Preparation

The material used in this investigation is a standard pressure tube alloy, 28 pct cold worked Zr-2.5 wt pct Nb, containing ~1100 ppm oxygen, 10 ppm hydrogen, and 40 ppm nitrogen. The standard manufacturing process of pressure tubes involves extrusion from a billet at 850 °C, followed by cold-drawing (25 to 30 pct) and stress relief (autoclaving) at 400 °C for 24 hours. Compact tension specimens, 17 mm wide and 3.8 mm thick, were prepared from flattened pressure tube with the notch cut in the axial direction of the tube. The texture of the pressure tube material is such that most of the basal plane normals are oriented in the circumferential (transverse) direction (about 60 pct in the circumferential, 36 pct in the radial, and 4 pct in the longitudinal direction). The specimens were hydrided gaseously at 350 °C for 4 hours to a concentration of about 50 µg/g (0.46 at. pct) hydrogen. These specimens were then homogenized at either 390 °C, 350 °C, or 310 °C for about 10 days. These different homogenization temperatures were used to study the effect of the continuity of the β-phase (present as a continuous grain-boundary network surrounding the elongated α-grains) on the crack velocity. The β-phase has been shown to undergo decomposition and agglomeration at about 500 °C after a 0.5-hour heat treatment. With a TSS temperature of about 280 °C for this hydrogen concentration (50 µg/g), it is expected that all the hydrogen will be in solution at these homogenization temperatures. Some of the solution-treated specimens were then quenched, either in ice-water, oil, or liquid nitrogen, to produce fine hydride dispersions. Others were furnace cooled from the solution temperature, to produce a much coarser hydride distribution. All specimens were precrack under dynamic fatigue, using an initial maximum stress intensity factor of about 12 MPa√m. The R-ratio ranges from 0.3 at the beginning of the fatigue precracking to 0.4 at the end. The maximum load on the specimen was subsequently reduced in steps so that, for the last 50 pct of fatigue precrack extension, the maximum stress intensity factor was kept below 5 MPa√m.

B. Testing Procedure

The specimens were loaded in a furnace, using a 10:1 lever loading system, at an initial stress intensity factor of about 6 MPa√m, once the desired test temperature was reached. Isothermal DHC tests were performed at various temperatures, ranging from 150 °C to 216 °C. The load on the specimen was increased by small increments (generally about 1 MPa√m) to establish the velocity-stress intensity factor curves. Crack velocity was measured using the potential drop technique. Acoustic emission equipment (Dunegan 3000) was used to estimate the incubation periods (time required for cracking to initiate) and to study the fracture mechanisms at different stages of the crack growth, using amplitude distribution analysis.

C. Metallography and Fractography

Specimens for optical examination were prepared from the test specimens before the DHC tests, to study the hydride morphology resulting from the different cooling rates. Some specimens were also examined after the DHC test to determine whether a change in the hydride morphology occurs during testing. Transmission electron microscopy examination was undertaken to determine the microstructural changes of the grain boundary β-phase resulting from heat treatment at the different homogenizing temperatures.

The fracture surface was examined on the broken specimens, after the DHC tests, using light microscopy. Oblique illumination, with the light directed in the crack propagation direction, was used to observe the intermittent nature of the crack growth process. In this way, the individual steps of crack propagation can be resolved on the fracture surface as striations, the spacing of which represent incremental crack growth. The average striation spacing was estimated for both quenched and furnace-cooled specimens.

III. RESULTS

A. Hydride Morphology

The size and distribution of the hydrides in quenched and furnace-cooled material are shown in Figures 1 and 2, re-