Behavior and Rupture of Hydrided ZIRCALOY-4 Tubes and Sheets

F. PRAT, M. GRANGE, J. BESSON and E. ANDRIEU

The mechanical behavior and rupture mechanisms of ZIRCALOY-4 guide tubes and sheet containing 150 to 1200 wt ppm hydrogen have been investigated at room temperature. Sheets were notched to study the influence of geometrical defects on rupture. It is shown that hydrides strengthened the material, as maximum stresses sustained by the material are increased with increasing hydrogen contents. On the other hand, ductility is reduced. The material also exhibits a strong anisotropy due to its pronounced texture. Metallographic examinations have shown that damage by hydride cracking is a continuous process that starts after the onset of necking. Notches reduce ductility. A modified Gurson–Tvergaard model was used to represent the material behavior and rupture. Numerical simulation using the finite element method demonstrates the strong influence of plastic anisotropy on the behavior of structures and rupture modes.

I. INTRODUCTION

ZIRCONIUM alloys are used as structural parts in the nuclear fuel assembly. Their oxidation by water in the reactor produces hydrogen, which diffuses in the bulk material. Hydrogen, which has a low solubility in zirconium, precipitates as zirconium hydrides. The influence of these precipitates on the mechanical properties of zirconium alloys has been the subject of many investigations since the development of nuclear power plants. Previous studies on zirconium alloys showed that cracking of hydrides during straining causes the acceleration of the ductile failure process. The plastic strain yield for hydride cracking depends on the temperature, the stress state, and the hydride orientation. Puls and Choubey and Puls used acoustic emission to detect hydride failure in smooth tensile specimens at different temperatures; they showed that hydride embrittlement is reduced with increasing temperature and that embrittlement is suppressed above 300 °C. The effect of notches has also been studied. Results show a strong decrease of ductility and a decrease of the yield strain needed to start breaking the hydrides. Another way to study the effect of the stress triaxiality ratio on the rupture behavior of hydrided zirconium is the punch-stretch testing technique used by Yunchang and Koss. This method allows the development of a biaxial stress state. They show a reduced ductility; however, the yield strain for hydride cracking remains constant. The comparison of the crack densities for uniaxial and biaxial testing showed a large increase of the void density for specimens tested under biaxial tension. Moreover, the void growth rate was much higher in that case.

In the French pressurized water reactor, where most of the structural parts of the fuel assembly consist of zirconium alloys, the hydride volume fraction remains tolerable with respect to the loads encountered during service (including handling and possible major accidents). However, an increase in the lifetime of the fuel assembly in order to increase the uranium burnup is planned. All the previous studies gave experimental data, but no attempt was made to propose a numerical model able to describe both plastic behavior and rupture. Such a predictive model is necessary to assess the structural integrity of the nuclear fuel assembly. In particular, the material model should be able to describe materials with hydrogen contents up to 1000 ppm.

This work consists of two parts. The first one is concerned with mechanical testing of artificially hydrided recrystallized ZIRCALOY*-4 tubes and sheets containing up to 1200 ppm hydrogen. Tension tests on smooth tubes and notched sheets were carried out to characterize the plastic behavior of the material, its ductility, and its notch sensitivity. These data are used in the second part to propose a mechanical model that could be used to compute complex structures. This model is based on the Gurson model often used to represent plastically voiding metals. This model has been modified to account for both the anisotropic plastic behavior of textured zirconium and void nucleation caused by hydride cracking. It is expected that this type of model could represent the effects observed in previous studies such as the effect of the stress triaxiality ratio on the ductility or the presence of a yield strain for hydride cracking. The present experimental data are used to adjust the parameters of the model, some of which can be determined using quantitative metallography. Finally, the model was implemented in a finite element simulation software, allowing the comparison of experiments and calculations.

II. EXPERIMENTAL

A. Material

The material of this study consists of ZIRCALOY-4 tubes (thickness 400 μm) and sheets (thickness 1.8 mm).

*ZIRCALOY-4 is a trademark of Westinghouse Electric Company, Pittsburgh, PA.
Table I. Composition of ZIRCALOY-4

<table>
<thead>
<tr>
<th>Sn</th>
<th>Fe</th>
<th>Cr</th>
<th>O</th>
<th>Zr</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5 wt pct</td>
<td>0.2 wt pct</td>
<td>0.1 wt pct</td>
<td>1200 ppm</td>
<td>bal</td>
</tr>
</tbody>
</table>

Fig. 1—Notched and smooth sample geometry; hydride distribution in the notched region.

Both materials were cold rolled. The composition of the material is given in Table I. Tubes and sheets were given a recrystallization treatment leading to an equiaxed grain structure (average grain size: 10 μm). Resulting from the elaboration process, both structures exhibit a strong texture with a maximum basal pole density at ±20 deg from the radial (tubes) or short transverse (sheets) direction. The sheet texture is similar to the one of the tube. The long/transverse/short (L, T, S) directions of the sheet correspond to the long/hoop/radial (z, θ, r) directions of the tube (Figure 1).

Specimens were artificially hydrided at 400 °C in an argon/hydrogen gas mixture. They were then slowly cooled down (2 °C/min) to allow the precipitation of the stable δ-hydride. Depending on the exposure time, the resulting hydrogen content ranges from 150 to 1200 wt ppm. This technique leads to a uniform precipitation of δ-hydrides in the material. The hydride distribution in a circumferential cross section of a tube for different hydrogen contents is shown on Figure 2.

The δ-hydrides precipitate as very fine platelets. Precipitation occurs either at grain boundaries or inside the grains. In this study, segregation at grain boundaries was not observed. Precipitation leads to a volume expansion of about 17 pct. This causes plastic deformation in the vicinity of the newly microscopic formed hydrides and enhances hydrogen diffusion toward the existing precipitates. As the local hydrogen concentration is increased, precipitation takes place. This autocatalytic process leads to the formation of mesoscopic hydrides, which consist in microhydride stacks. Transmission electron microscope examinations have shown that hydrides are actually very fine (thickness ≈ 0.1 μm) and tend to build “mesohydrides” (evidenced on figure 2), which consist of these fine precipitates surrounded by microscale matrix ligaments. In the following work, “hydride” will refer to mesoscale precipitates considered as a single entity.

The orientation of the platelets has previously been discussed. It was shown that precipitation occurs in {101} planes, which are close to the basal plane of Zr. The present observations using optical microscopy (Figure 2) show that the hydride platelet normal is close to the S/r direction so that the hydrides lie in planes close to the (T, L)/(θ, z) planes. This observation is consistent with the previously mentioned results, as the S direction is close to the basal pole (texture).

Hydrogen contents given in this study have been measured after mechanical testing close to the rupture zone by fusion at 1800 °C in an inert gas and are an average of three measurements. After chemical polishing, hydrides were revealed by anodization. The measured hydride volume fraction was also determined using image analysis. The measured mesohydride volume fraction $V_H$ can be described by an empirical law as a function of the hydrogen content $C_H$ (ppm) by

$$V_H = \begin{cases} 
0 & \text{for } C_H < 100 \\
0.0003 (C_H - 100) & \text{for } C_H > 100 
\end{cases}$$

B. Mechanical Testing

Two types of specimens were used: smooth tubes and notched sheets.

1. Smooth specimens

All tests were carried out on tube specimens having a total length of 180 mm, a diameter of 12 mm, and a thickness of 0.4 mm. These specimens were used to study both the mechanical behavior and the hydrogen embrittlement. Tests were performed on a 10-ton MTS hydraulic machine at room temperature, as this temperature is more critical than the service temperature (320 °C). Load and axial de-