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EFFECT OF HYDRIDES ON THE DUCTILE-BRITTLE TRANSITION IN STRESS-RELIEVED,  
RECRYSTALLIZED AND BETA-TREATED ZIRCALLOY-4

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**Effect of Hydrides on the Ductile-Brittle Transition in Stress-relieved, Recrystallized and  $\beta$ -treated Zircaloy-4**

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It is generally established that the embrittlement of zirconium by hydrides is caused by the presence of the brittle hydride phase, localized at the crack tip or gross embrittlement due to high hydride volume fractions. Important factors are the hydride morphology (volume fraction, size and orientation) and the matrix crystallographic characteristics (texture and grain size). This paper is concerned with the influence of  $\delta$ -hydrides on the mechanical properties of three heat treated cold-rolled Zircaloy-4 sheets (stress-relieved, recrystallized and  $\beta$  treated), tested at room temperature and 350°C. Smooth tensile specimens of two thicknesses: 0.5 and 3.1 mm, containing different hydride volume fractions, up to 18% (about 1400 ppm H), have been tested. Metallographic and fractographic analysis were carried out in order to examine the fracture morphology near and on the fracture surface, and to determine the evolution of the fracture mechanism of hydrides as a function of temperature, hydride orientation and volume fraction.

#### Part I - Influence of Hydrogen Content and Temperature

The specimens with the tensile axis along with the transverse direction of cold-rolled sheets were gaseously hydrided at 400°C, and cooled down to room temperature with a cooling rate  $< 1.4^\circ\text{C}/\text{min}$ . After this treatment, the hydride morphology, controlled by optical analysis, was strongly dependent on the previous thermo-mechanical treatment of Zircaloy-4:

- for the stress-relieved state, the hydride platelets are usually aligned along the rolling plane and have the same normals as the sheets;
- for the recrystallized state, hydrides are coinciding with the grain boundaries so their morphology is controlled by grain size and grain boundary orientation;
- for the  $\beta$  treatment, hydrides precipitate at the  $\alpha$  platelets interfaces, in the big  $\alpha$ -grain born of the high temperature  $\beta$  phase, independently of the sheet thickness.

It should be noted a remarkable hydride morphology difference between 0.5 and 3.1 mm sheets for stress-relieved and recrystallized states. Generally, thin sheets exhibit relatively more randomly distributed and smaller hydrides than thick ones.

At room temperature (20°C), for the three metallurgical states, the effect on strength of increasing hydrogen concentration is not significant and about 10% variation of yield strength and tensile strength was observed for all hydride volume fractions. The ductility (elongation and reduction of area) is drastically reduced when a critical hydride volume fraction is reached, as shown in figure 1. This critical value varies for the three states, about 600 ppm for 0.5 mm stress-relieved sheet, 700 ppm for 0.5 mm recrystallized sheet, less than 250 ppm for 0.5 mm b sheet; the presence of an oxide layer tends to decrease this value. The specimen thickness influence has been also studied.

However, at 350°C, the specimens are ductile up to very important hydrogen concentrations, higher than those for the ductile-brittle transition observed at 20°C. At this temperature the mechanical properties are only slightly affected by hydrides.

During hydride formation, the anisotropic misfit strains between hydrides and the hexagonal-close-packed zirconium matrix result in a preferred orientation of the hydride platelets in the anisotropic stress field caused by non-relieved cold-rolled residual stresses and misfit stresses. Coupled with the metallographic features corresponding to the three heat treatment states, this stress field, which is different for each state, consequently results in various hydride morphologies. For the  $\beta$  state, within very big  $\alpha$ -grain, the hydrides laying at the interfaces contribute to the decohesion between these interfaces then to the final fracture, thus making this state more susceptible to hydrides. There is no great difference for the critical hydride volume fraction between stress-relieved and recrystallized states, which is only slightly higher for the recrystallized state. Of special note is that the transition for stress-relieved state is more spread out.

Another important factor which should be taken into account when one discusses the susceptibility to hydrides is the hydrogenation rate: for the same exposure time to hydrogen at 400°C,  $\beta$  specimens absorb less hydrogen than those of the two other states. Furthermore, to introduce the same hydrogen content, the time needed is 20-30 times greater for thick sheets than for thin ones.

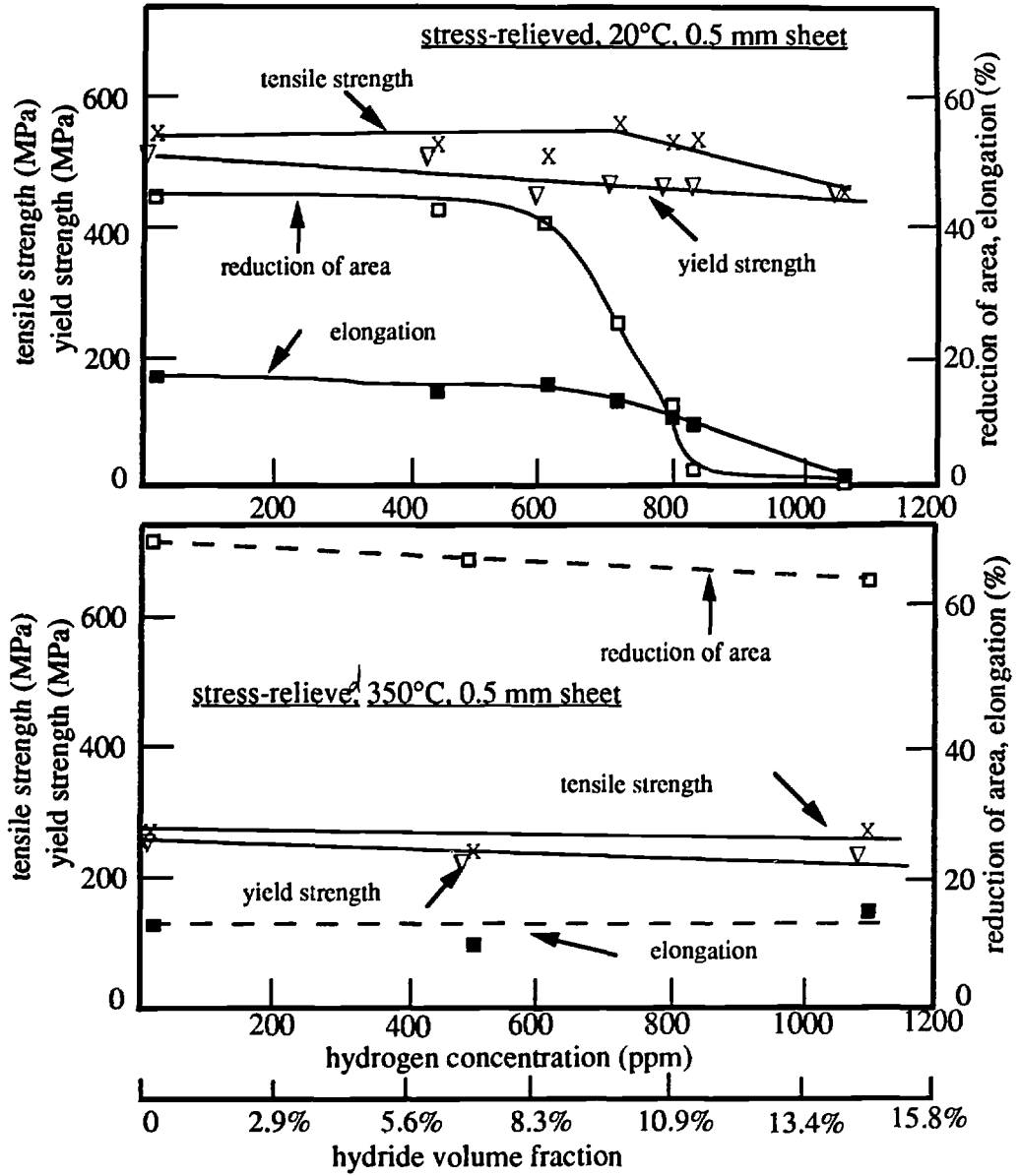


Fig.1: Influence of hydrogen concentration and temperature on the mechanical properties of Zircaloy-4.

## Part II - Stress Orientation of Hydrides and its Influence on the Mechanical Properties of Zircaloy-4

Because of the deleterious effects of oriented hydrides on the fracture behavior of zirconium and its alloys, the factors which give rise to preferential orientation of the precipitate (radial hydride for tubes) have been extensively examined. However, in these previous works, the oriented hydrides have been produced by cooling specimens under stress (called reorientation). It is possible that for tests with hydrogen concentrations higher than the terminal solid solubility at the temperature considered, full reorientation could not be realized in this way. In this study, a quantitative technique was developed for evaluating the susceptibility of cold-rolled Zircaloy-4 sheets (for three heat treatments) to the formation of preferential hydride orientation. The external applied threshold stress was determined by using tapered specimens (similar to those used by Leger and Danner) and image analysis technique. Then, the influence of hydride orientation on the mechanical properties of this material was estimated by tensile test on smooth sheet specimens hydrided under stress.

When hydrogenated under stress and for sufficiently high stresses, the hydrides can be totally oriented perpendicularly to the applied stress. For stress-relieved and recrystallized specimens, this threshold stress increases with the yield stress at 400°C. An influence of specimen thickness and heat treatment was observed and attributed to:

- 1) the orientation of grain boundaries and the textured microstructure, because hydrides precipitate frequently at grain boundaries or interfaces, with habit plane,
- 2) the stress field resulting from the residual stresses in the sheets (demonstrated by initial hydride morphology) and the misfit stresses.

In stress-relieved and recrystallized tapered specimens, the proportion of oriented hydrides diminishes along the stress reduced direction. However, for the  $\beta$  treatment, the morphology is more complicated, and hydrides can change their initial precipitation site to cross the interfaces but no universal orientation was observed.

The oriented hydrides have a noticeable influence on the yield strength and tensile strength of Zircaloy-4 sheets for small or middle hydrogen contents compared with specimens containing the same

hydride volume fraction. Using microscopy analysis, it is shown that radial hydrides break earlier than others. For a critical matrix plastic deformation criteria, this means that radial hydride needs smaller deformation for fracture. This induces the earlier fracture and the ductility reduction in sheet specimens. The complete brittle behavior, however, needs a sufficient quantity of radial hydrides corresponding to small interdistances between hydrides.