EXPERIMENTAL INVESTIGATION OF STRAIN, DAMAGE AND FAILURE OF HYDRIDED ZIRCONIUM ALLOYS WITH VARIOUS HYDRIDE ORIENTATIONS

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ABSTRACT

This experimental investigation is devoted to the hydride embrittlement of fuel cladding tubes and especially to the influence of the orientation of hydrides with respect to the applied stress on strain, damage and failure mechanisms. Ring tensile tests are performed on cladding tube material (unirradiated cold worked stress-relieved Zircaloy-4). The average hydrogen content of the material is about 200ppm, and orientations of hydrides are either "tangential" (hydride platelets parallel to the tensile direction) or "radial" (perpendicular to the tensile direction). Tangential hydrides are usually observed in cladding tubes, in particular because of the texture of the material. However, hydrides can be reoriented after cooling under stress and become radial and then trigger brittle behaviour. In this investigation, we first perform "macroscopic" tensile tests, on smooth rings, which give us the mechanical response of the material as a function of hydride orientation. Then, we perform SEM in-situ tensile tests, on rings with the same geometry, in order to observe damage and failure mechanisms. In both cases, digital image correlation techniques are used to estimate local and global strain levels. For macroscopic tests, airbrush speckle painting is used to mark the samples while 2µm-pitch micro-grids are used for in-situ tests. The "macroscopic" tests underline the strong influence of the hydrides orientation: the specimen with radial hydrides suddenly fails at 1700N, within the elastic domain, whiles the specimen with tangential hydrides reaches 4300N and develops macroscopic shear band. The SEM in-situ tests allow us to improve the understanding of failure mechanisms: with radial hydrides, a crack propagates along a path of aligned hydrides and leads to failure. With tangential hydrides, macroscopic shear bands lead to large plastic strain and final ductile failure; damage can only be observed in the very late stage of deformation.

1 INTRODUCTION

In PWR (pressurized water reactors), the fuel assemblies are mainly made of zirconium alloys. During irradiation, these components are oxidized by the water of the primary circuit (Zhang [1]). Part of the hydrogen produced by this reaction is picked up by the cladding tubes, made of Zircaloy. When the solubility limit is reached, hydrogen precipitates into zirconium hydrides. These are known to strongly embrittle the material, in particular in cold conditions. But the changes in the mechanical response induced by hydrides (loss of ductility, reduction of fracture toughness) depend on several parameters: hydrogen content, hydride phase, morphology and spatial distribution of the hydrides and in particular their orientation with respect to the applied stress, since hydrides, because of their platelet morphology, are strongly anisotropic particles.

This investigation is devoted to the influence of the orientation of hydrides with respect to the applied stress on strain, damage and failure mechanisms. Indeed, even if hydrides are mainly tangential (i.e. perpendicular to the radial direction of the tubes) because of the texture induced by the manufacturing process and not noxious for usual loading conditions, they can be reoriented under some conditions of temperature and stress, and in the case of radial hydrides (normal to the circumferential direction of the tubes), they can lead to a dramatic loss of ductility and embrittlement of the material (Bai [2], Racine et al. [3]).

2 EXPERIMENTAL PROCEDURES

Specimens are extracted from unirradiated cladding tubes made from cold-worked stress-relieved Zircaloy-4 (460°C during 1h30). Tubes have been hydrogen charged by a gaseous method, which was performed at 400°C in an argon-hydrogen atmosphere. After hydrogen charging (about 230ppm for the tests reported here), hydrides are reoriented by heating to 400°C and then cooling specimens under stress. This hydrides reorientation process does not imply plastic deformation of tubes and allows us to get different hydrides orientation depending on the charge applied. Then we work on material charged with 230ppm hydrogen, with tangential or mostly radial hydrides (Figure 1) or with both tangential and radial hydrides in the same specimen (with respect to the applied stress). This last case will be subsequently called "intermediate" (Figure 1). After hydridation and reorientation treatments, the ends of tubes are cut off and heated to 1250°C during 20mn: the hydrogen content of this sample is given by the quantity of gaseous hydrogen emitted. The hydrogen is supposed to be homogeneously distributed in the tubes. Meanwhile hydrogen content of the specimen studied here will be precisely measured at the end of this investigation (not before, because of the destructive nature of these tests).

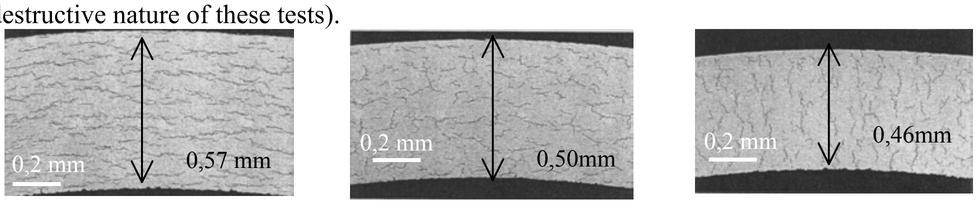
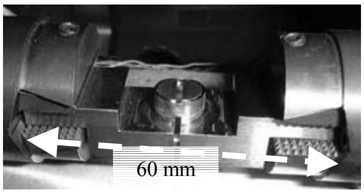


Figure 1: Metallographic cross sections of hydrided tubes (about 230ppm) with tangential (left), "intermediate" (middle) and radial hydrides (right).

Above the solubility limit of hydrogen in zirconium, hydrogen precipitates and forms zirconium hydrides. Three phases of zirconium hydrides can form in zirconium-hydrogen system, γ, δ, ε. Their formation depends on different parameters, which are, for instance, the cooling rate, the amount of hydrogen… The hydride phase observed here is the delta phase, as revealed by X-ray diffraction, which is also the one formed in reactor. Composition of these hydrides is ZrH_x ($x = 1{,}53$ to $1{,}66$). The metal atoms form a face centered cubic lattice, in which the hydrogen atoms occupy tetrahedral interstitial sites.

Hydrides (delta phase) are usually described as platelets. But, different scale can be used to observe them. On cross-sections normal to the axis of the tubes observed under the optical microscope, we can see "macroscopic hydrides" which form thin and long bands in the section of tubes. These bands are approx. 80µm apart each other. With a scanning electron microscope (SEM), we can see about 10 to 50µm long and 0,3 to 0,5µm wide hydrides and also very little platelets (5 to 10µm long and 0,3µm wide) which form the segments of these macroscopic hydrides. According to the literature micro-platelets are also visible with a transmission electron microscope (TEM). Here, the word "hydrides" is used to refer to the "macroscopic hydrides".

For both unhydrided and hydrided (with different hydrides orientations) specimens, two tensile tests are performed, one macro tensile tests and one in-situ SEM tensile tests, on smooth rings, whose characteristic dimensions are: thickness = 0,57mm, axial length = 5mm, external diameter = 9,5 mm. Smooth rings are chosen to make possible the observation of the lateral faces of the rings (normal to the one of the tube) during the SEM in-situ tests. All tests are carried out at 20°C. Specimens are tested up to fracture, at a displacement rate of 1µm/s. Load and displacement are recorded.



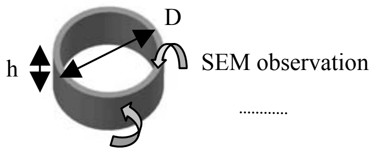


Figure 2: Ring testing system (left) and scheme of the ring with the observed surfaces (right)

The macro tensile test is performed first to get the mechanical response of the material for this kind of ring tensile test, and secondly in order to “prepare” the SEM in-situ tests : that is to say in order to forecast what will be the maximal charge and where and when rupture and/or localization bands (if there are any) appear. Finally, the SEM in-situ tensile test allows us to observe the damage mechanisms of the specimen during the deformation ; in these tests attention is focused on area where fracture and localization have been observed during the macroscopic tests. We can also gain access to the deformation modes at the microstructural scale, by means of local strain field measurements techniques. Indeed, in both tests, the digital image correlation method (Doumalin[3]) is used to gain access either to the global (macroscopic tests) or to the local strain (in-situ tests) field.

During the macroscopic tensile tests, the non-plane area is observed with an optical camera (with a 1300 x 1030 pixels CCD). A specklegram is deposed with an airbrush on the observed surface with a pattern as fine as possible (roughly 50 µm-diameter spots). One image each second is acquired all along the test. Local details on the speckled surface make identification of homologous points and then digital image correlation possible. Strain maps (gage length for local strain values of about 150µm) are plotted and allow us to characterize the macroscopic deformation modes: the principal point of interest is to know if macroscopic strain bands develop or not, and to get a better quantitative knowledge of the actual strain in the sample, since overall displacement provide only an approximate value because of the complex geometry and loading mode of the test.

During the SEM in-situ tensile tests, the section perpendicular to the tube axis is observed. This surface is mechanically polished (with silicon carbide papers and oxide particles suspension): this way, hydrides can be observed in the SEM with back-scattered electrons and no artefact like those induced by chemical etching (disappearing of hydrides and grooves in the surface instead of platelets) is noticed (Racine et al.[4]). After polishing, about 13 microgrids are deposed by a microelectrolithographic technique (Allais et al.[5]) on both sides of the section to be observed. Each microgrid covers a 500µm x 500µm area. The steps of the grids is 2 µm and allow us to get qualitative and quantitative indications on the local deformation mechanisms at the scale of the hydrides. High resolution images (4096 x 4096 pixels) of several domains of interest are acquired for different strain levels. Since it takes 15 minutes to acquire one image, the load is stopped (and slightly reduced, in order to limit relaxation during acquisition). By using the image correlation technique, local strain maps (with a gage length of 4µm) are plotted: the strain distribution can be characterized visually and isolevel curves indicate where the strain localizes. In addition, averages of the strain over area of about 200µm x 200µm provide the overall loading conditions under which the local deformation or damage mechanisms are activated and these averages can be compared to the local strain values measured optically during the macroscopic tests.

Besides, the microstructure remains visible: correlation between the local strain levels, damage mechanisms, and the microstructure, in particular the spatial distribution of hydrides, can be investigated. Situations without hydrides, with tangential, radial or intermediate orientations of hydrides are compared.

3 EXPERIMENTAL RESULTS

Load-displacement curves (Figure 3) clearly show the differences in the response of hydrided Zircaloy-4 according to different hydrides orientations (these curves can be compared because the specimens have all exactly the same geometry but strain-stress curves cannot be deduced because the local mechanical state is not uniform in the sample, as discussed later). First, the unhydrided specimen reaches the maximal force (4400N) and final fracture occurs later than for the other specimens. The global response is ductile, macroscopic strain bands develop and necking occurs. Final fracture occurs along macroscopic strain bands. For hydrided specimen (228ppm) with tangential hydrides, the maximal force reached is slightly but probably not significantly smaller (4300N) and the response is almost the same, with in particular final failure for almost the same displacement. In the specimen with radial hydrides and charged with 245 ppm hydrogen, the response is very different from the first two cases : while the first part of the load-displacement curve is almost the same, the fracture happens suddenly (1700N), without prior development of plasticity and the crack propagates along a straight line, normal to the applied load. For a specimen with the same hydrogen content (228ppm) and with "intermediate" hydrides, the maximal force is almost the same (4400N) as without hydrides, plasticity mechanisms are of the same kind (macroscopic strain bands and necking are observed at the same positions), but the fracture mode is totally different : final fracture occurred along a straight line, normal to the applied load, it is brittle like in the case with radial hydrides.

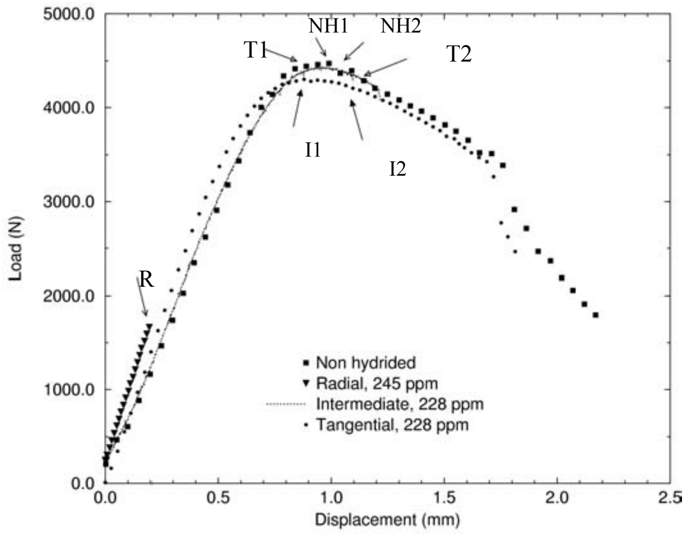


Figure 3: Load-displacement curves obtained from macroscopic tensile tests for non-hydrided and hydrided material (roughly 230ppm) with 3 different hydrides orientations.

Strain maps given by the image correlation technique can be linked with the load-displacement curves, in order to gain access to the local deformation modes for different load levels. These maps cover an about 5 x 7mm area and represent the local strain values of the non-plane area of the ring. As previously discussed, the strain gage length for these maps is about 150µm.

On figure 4, we notice for the first three cases (non-hydrided, tangential or intermediate) the similarities of local deformation modes: apparition of localization bands at the maximum load (bands begin to form for NH1, T1 and I1, and are easily recognizable for NH2, T2 and I2) and initiation of ductile failure at the intersection of these bands in the first two cases. In the case of radial hydrides, the strain map just before final failure (R1) confirms that the rupture is brittle: no significant deformation can be noticed, the deformation heterogeneities must be due to the test itself: indeed at the beginning the ring is mostly submitted to flexion and not to pure tension. Besides, the images from which correlation is done are plane projections of a non-plane area and motion: artefacts are induced and especially visible for such low strain levels.

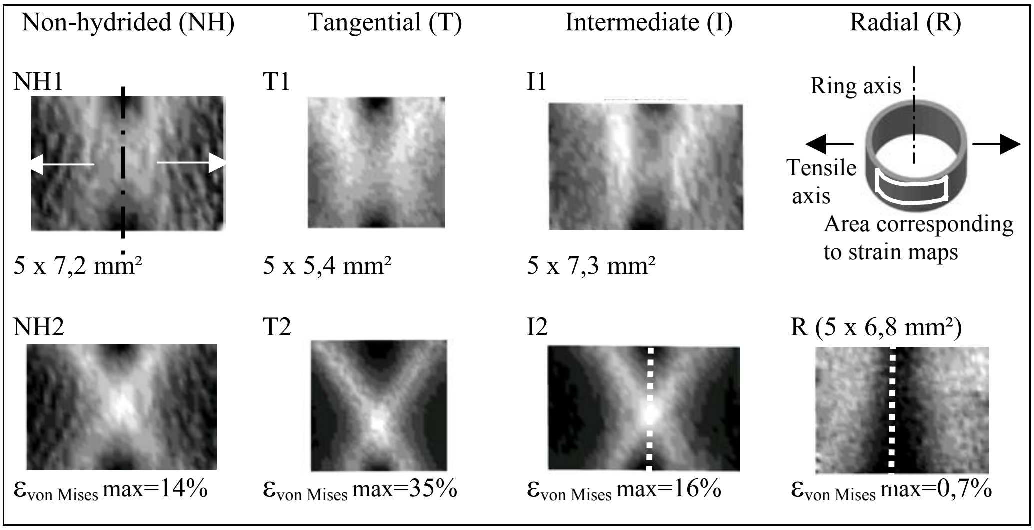


Figure 4: Strain maps of the different materials studied before and after the localization strain bands apparition (if any). The maximal deviatoric strain value is given by the highest gray level (white color). In the last two cases the dotted line represents the crack path.

According to previous studies on material with higher hydrogen contents (Grange [5]), damage in hydrides can be observed only if the global level of plastic strain is relatively significant (at least 15%). That is why we use indications given by macroscopic tensile tests to forecast either where, at the surface of the ring which is observed during tensile tests, strain localization bands will appear and therefore where the plastic strain level will be the highest, or where the fracture will occur (in the case there will be no strain bands).

In specimen with tangential hydrides strain level (averaged value on a domain) reaches 40% in domains (100 x 260µm) inside the macroscopic localization bands whereas in others domains (140 x 200µm) outside the bands it is only 1%. Strain maps (relative to a 4µm gage length) represented by isovalues curves are superimposed on images of the microstructure in order to observe whether strain heterogeneities are linked or not with the presence of hydrides. Until now, in the case of tangential hydrides, it seems that strain localizes in microbands at the microstructural scale, and that hydrides are preferably outside these bands : hydrides seem to affect the local plastic flow. Nevertheless, a more detailed statistical analysis is required and currently under progress in order to be able to draw more quantitative conclusions about the role of hydrides on deformation mechanisms; in particular, these microscopic strain bands and their statistical properties (characteristic length, witdth, spacing,…) will be compared in different cases: hydrided or not hydrided material, with different hydrides orientations and different hydride contents. About microdamage mechanisms, in the most deformed domains, during the final steps just before final failure, damage is observed in some hydrides: cavities appear in their thickness. No decohesion has been observed until now.

Unlike this case, in specimen with radial hydrides, the fracture occurs suddenly, in the median plane of the specimen. Just before failure, the strain level in this domain (350 x 300µm) is less than 2%. The behaviour of such a material is brittle. For several similar tests, we managed to reconstitute the path followed by the crack which led to failure: indeed, the crack propagates along radial hydrides which are more or less aligned along a radius of the ring, in a domain where the stress is normal to the direction of this radius.

4 CONCLUSIONS

These experimental results underline the influence of the orientation of hydrides platelets on failure mechanisms of hydrided Zircaloy-4. Indeed, for almost the same hydrogen content (230ppm), material is brittle (radial hydrides), ductile (tangential hydrides) or undergoes plastic flow and a beginning of necking but then fails in a brittle mode ("intermediate" case). Such different micromechanisms need to be explained with more insight in order to be able to propose a micromechanically-based failure criterion. For instance according to the responses given by the macroscopic tests, unhydrided and hydrided material with tangential hydrides seems to have almost the same behaviour. Nevertheless, we need to investigate more precisely micro-mechanisms to know if a correlation exists between the strain modes and the presence of hydrides. Investigations need to be carried on: macro and in-situ tensile tests will be performed on material with about 500ppm hydrogen, and comparisons of strain modes at the microstructural scale between hydrided and unhydrided materials are planned. We hope it will allow us to draw more quantitative conclusions on the role of hydrides on deformation, damage and failure mechanisms of hydrided material.

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