HYDROGEN DECREASED DUCTILITY OF A U-6%NB ALLOY

David F. Teter, Robert E. Hackenberg, Michael E. Mauro, Rodney McCabe & Patricia O. Dickerson
Materials Science & Technology: MST-6 Metallurgy
Los Alamos National, Laboratory, Los Alamos, NM 87544 USA

ABSTRACT

Hydrogen decreases the ductility of uranium alloys without resulting in a brittle failure. Due to the low solubility of hydrogen and the propensity to form hydrides in uranium and its alloys, it might be expected that the mechanism of embrittlement would be the stress-induced hydride formation and cleavage mechanism. However at 2-5 wppm levels, hydrogen significantly decreases the ductility of the alloy without resulting in a cleavage-based mechanism. We have chosen the uranium-niobium (6wt.%) alloy system to investigate the hydrogen embrittlement phenomena in uranium-based alloys. The alloy was gas-phase charged at 800 ºC at pressures ranging from 10^{-4} to 10^{-1} MPa using ultra-high purity hydrogen gas (99.999%) followed by water quenching. Hydrogen contents ranged from 0 wppm (vacuum outgassed) to 20 wppm. Tensile tests were performed at strain rates of 10^{-3} and 10^{-5} sec^{-1}. The elongation and reduction in area decrease rapidly from 0 wppm to 5 wppm with little further reduction at 20 wppm. Hydrogen has no effect on the yield strength, ultimate tensile strength and work hardening coefficient. Scanning electron microscopy of the fracture surfaces shows that all of the samples fail by ductile microvoid coalescence. Concomitant with the reduction in ductility due to increased hydrogen content, the microvoid size also decreases with increasing hydrogen content. This alloy has numerous inclusions and the microvoids all nucleate at the inclusions. At low hydrogen contents, the microvoids appear to nucleate only on the largest inclusions (~10μm). As the hydrogen content increases, the microvoids tend to also nucleate on smaller inclusions (< 1 μm). Hydrogen appears to be decreasing the inclusion/metal interface strength of the smaller inclusions and localizing deformation near the debonded inclusion. Since many more microvoid nuclei are active due to hydrogen, the microvoids link together and cause fracture at a lower overall macroscopic strain. (LA-UR-04-3343)

1 INTRODUCTION

Since uranium and uranium alloys react readily with hydrogen to form hydrides [1, 2], it would be expected that a mechanism of hydrogen embrittlement for these materials would be a hydride based mechanism. Powell et al. [3]studied the effect of hydrogen on the mechanical properties of a U-5.7%Nb alloy and showed that hydrogen decreased the measured ductility. However the actual fracture mechanism for the highest hydrogen content examined was still ductile microvoid coalescence. No evidence was found for uranium hydrides causing the reduced ductility. Liang et al. have recently modeled hydrogen interactions with inclusions causing debonding of the metal-inclusion interface. This study is aimed at revisiting hydrogen reduced ductility in U-6wt.% Nb with particular emphasis on quantitative fractography and metallography to try to understand the mechanism of hydrogen reduced ductility in this alloy in light of the recent modeling efforts by Liang et al [4, 5].

2 EXPERIMENTAL

The alloy used in this study, uranium – 6 wt.% niobium was processed using a triple melting technique called VIM-VAR-VAR (Vacuum Induction Melting – VIM, Vacuum Arc Remelting – VAR) followed by rolling to achieve a plate geometry. Tensile specimens were then machined from 3 mm plate with a gage section of 6 mm wide by 41 mm long. After machining, the samples were heat treated at 1073 K under vacuum (10^{-4} Pa) and hydrogen charged by exposing to a partial pressure of hydrogen gas (0.1 to 101 kPa) for 30 min. followed by water quenching. Initial specimen dimensions, width and thickness, were measured using an optical microscope or micrometer. Tensile tests were performed on an MTS servo-hydraulic testing machine at room temperature using a constant strain rate of 10^{-3} mm/mm/sec. Loads were measured on a calibrated 245 kN load cell and strains were measured using a calibrated 25.4 mm clip gage extensometer. After testing, the final dimensions of the sample were again measured using an optical microscope or micrometer. Fracture surfaces were examined using a scanning electron microscope (SEM). The microvoid size of the fracture surfaces was measured using a method similar to the ASTM E112-96 circular intercept procedure for measuring the mean lineal intercept value, which is equivalent to the mean microvoid diameter at fracture. Optical microscopy and scanning electron microscopy was also used to characterize the inclusion size and distribution present in this alloy. Hydrogen contents were measured using an optimized inert gas fusion method [6].

3 RESULTS AND DISCUSSION

Hydrogen does not appreciably affect the 0.2% offset yield strength, ultimate strength or uniform elongation in U-6 wt.% Nb for the hydrogen contents examined in this study as shown in Table 1. In fact no significant difference in the stress-strain response can be seen up to the point of instability when the stress-strain curves for all of these alloys are overlaid. The ductility as measured by the plastic elongation appears to decrease with increasing hydrogen content but is difficult to see due to the appreciable experimental scatter. However, this decrease in ductility is most readily apparent by examining the reduction in area which measurably decreases with increasing hydrogen content. This suggests that hydrogen is not affecting the uniform mechanical response of this alloy, but is affecting the necking and fracture mechanism of this alloy.

Table 1: Mechanical property summary of hydrogen charged U-6%Nb alloys.

Hydrogen Content (wppm)	0.2% Offset Yield Strength (MPa)	Ultimate Tensile Strength (MPa)	Uniform Elongation (%)	Plastic Elongation (%)	Reduction in Area (%)
0.25±0.03	112 118	980 1007	26.5 28.1	29.4 30.5	32.3 30.1
2.2±0.3	117 119	995 993	27.0 25.8	30.4 27.7	32.9 30.3
4.8±0.3	122 126	994 1011	26.1 29.0	27.9 30.7	28.3 28.5
20 (calc.)	112 115	1002 984	26.8 25.4	27.3 26.4	23.6 24.8

SEM fractography shows a significant decrease in the microvoid size as a function of hydrogen content as shown in Figure 1 and 2. The dimples become smaller and shallower with increasing hydrogen content. At 20 wppm, the dimples appear to form in rows in certain areas as is seen in the lower left hand corner of Figure 1c. The spacing of these rows is consistent with the twin spacings that are often observed in these alloys. This suggests that hydrogen is affecting the deformation mechanism of certain twins depending on their relative orientation to the tensile axis. An example of the inclusion distribution found in this alloy is shown in the micrograph of Figure 3. Energy dispersive x-ray spectroscopy has shown that the types of inclusions typically found are niobium carbides, uranium carbonitrides, and uranium oxides. Inclusions of various sizes are often found at the bottom of the microvoids. For example a 10 μm inclusion is visible at the bottom of the large microvoid in the center of Figure 1a and smaller 1 μm inclusions are visible in other microvoids of Figure 1a. A statistical analysis of the inclusion distribution shows an asymmetric distribution with a peak frequency at 2 μm radius inclusions with a broad tail out to larger inclusions with radii up to 20 μm observed.

A postulated mechanism for the decreased ductility in hydrogen-charged uranium-niobium alloys is that the hydrogen is decreasing the cohesive strength of the inclusion/matrix interface with concomitant hydrogen enhanced localized plasticity generating microvoids leading to fracture. At low hydrogen concentrations, only the largest inclusions generate a sufficient stress field to cause hydrogen to diffuse to the interface, debond the interface and form microvoids. As the bulk hydrogen content increases, hydrogen may accumulate at smaller inclusions and hence decrease the inclusion-matrix interface strength. The result would be more microvoid nucleation sites, and hence the final size of microvoids on the fracture surface, since the number of small inclusions is greater than the large inclusions. Since this mechanism would be dependent upon the inclusion size and distribution, one might expect a minimum microvoid size plateau which is insensitive to further increases in hydrogen but limited by the distribution of the smallest inclusions. Similarly this mechanism would be dependent upon the diffusivity of hydrogen in the metal, and the fracture mode should be sensitive to strain rate. Therefore future studies will be aimed at examining a wider range of hydrogen contents as well as strain rates.

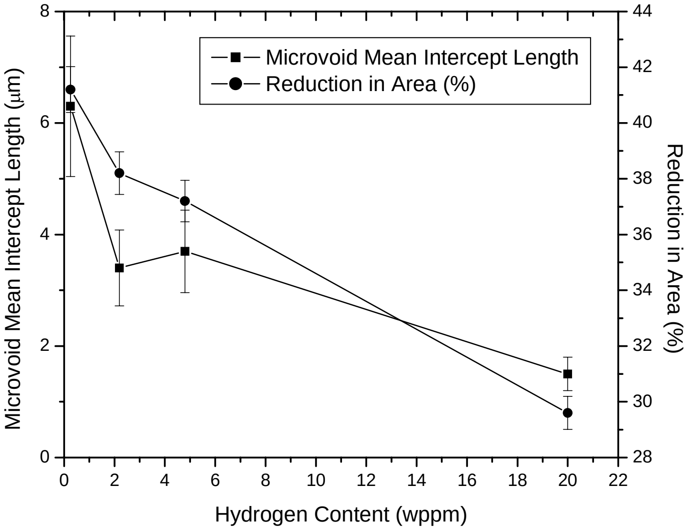

Figure 1: Variation of microvoid mean intercept length and reduction in area with bulk hydrogen content.

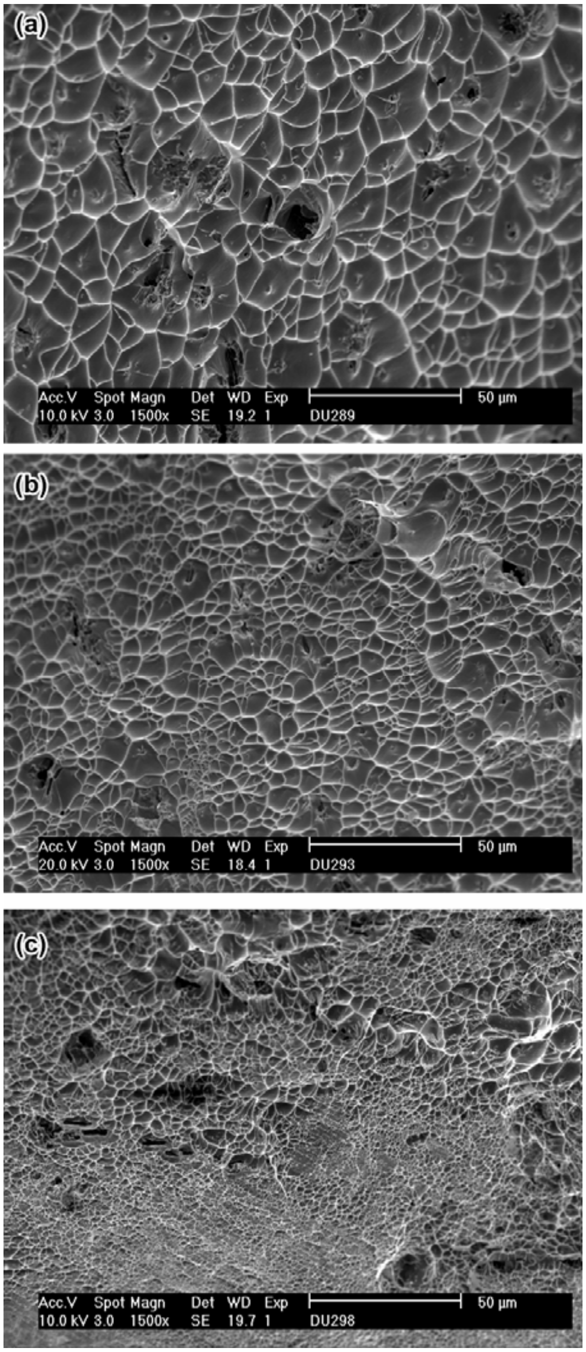

Figure 2: SEM Fractographs of hydrogen charged U-6%Nb alloys illustrating the decrease in microvoid size with dissolved hydrogen content; (a) 0.25 wppm, (b) 4.8 wppm and 20 wppm.

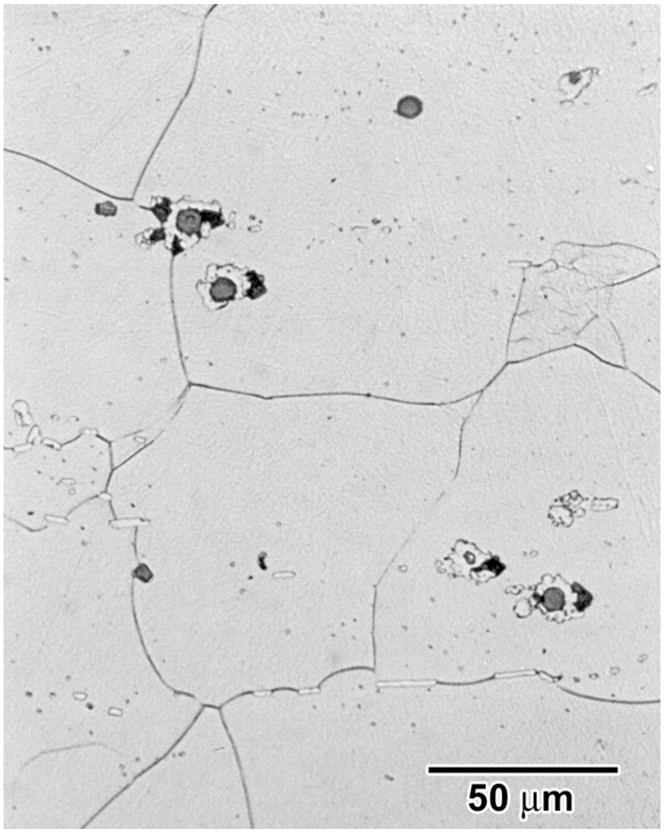

Figure 3: Optical micrograph showing the size and distributions of inclusions in this alloy.

4 SUMMARY

For the hydrogen contents examined, up to 20 wppm, no change in yield or ultimate strength was observed. Hydrogen has no appreciable effect on the uniform deformation of this alloy. However, the plastic elongation and reduction in area both decrease with increasing bulk hydrogen content. Even though the ductility of the material is decreasing due to hydrogen, the mode of fracture for this material is ductile microvoid coalescence. The microvoid diameter decreases by about a factor of five with increasing hydrogen content. Hydrogen appears to be decreasing the inclusion/metal interface strength of the smaller inclusions and localizing deformation near the debonded inclusion. Since many more microvoid nuclei are active due to hydrogen, the microvoids link together and cause fracture at a lower overall macroscopic strain.

5 ACKNOWLEDGEMENTS

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