INFLUENCE OF HYDRIDE MICROSTRUCTURE ON THROUGH-THICKNESS CRACK GROWTH IN ZIRCALOY-4 SHEET

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Abstract

The fracture toughness of cold-worked and stress-relieved Zircaloy-4 sheet subject to *through-thickness* crack growth within a "sunburst" hydride microstructure was determined at 25°C. The results were obtained utilizing a novel testing procedure in which a narrow linear strip of hydride blister was fractured at small loads under bending to create a well-defined sharp pre-crack that arrested at the blister-substrate interface. The hydriding procedure also forms "sunburst" hydrides emanating from the blister that were aligned both in the plane of the crack *and* in the crack growth direction. Subsequent tensile loading caused crack growth initiation into the field of "sunburst" hydrides. Specimen failure occurred under near-linear elastic behavior, and the fracture toughness for crack growth initiation into sunburst hydrides was in the range $K_Q \approx 10\text{-}15$ MPa√m. These results, when combined with those of a previous study [1], indicate that the through-thickness crack growth initiation toughness at 25°C is very sensitive to the hydride microstructure.

1 Introduction

During reactor exposure, the ductility of thin-wall zirconium components such as fuel cladding can be degraded by hydrogen ingress associated with waterside corrosion on the component surface. The resulting uptake of hydrogen has long been identified as a main contributor to limiting the fracture resistance of cladding even at high burnup levels [2, 3]. For thin-wall components such as cladding, a particularly relevant form of hydrogen-induced loss of ductility occurs when a surface crack initiates within a hydride rim or blister and subsequently propagates in a *through-thickness crack-growth mode* [4-7]. Predicting failure under such conditions requires knowledge of the crack depth and its geometry, the appropriate fracture mechanics, and a fracture toughness value for through-thickness crack growth. Depending on temperature, the latter may be quite sensitive not only to the total hydrogen content but also to the hydride distribution (i.e., solid hydride blister, sunburst, radial or circumferential hydride particles) [1, 8-14].

Based on bend testing, the fracture toughness (K_Q) behavior has been recently determined for thin Zircaloy sheet for through-thickness crack growth within either (a) predominantly in-plane hydrides (akin to circumferential hydrides in cladding tubes), or (b) a field of mixed in-plane/out-of-plane hydrides.[1] The room temperature behavior indicates that K_Q is sensitive to the orientation of the hydride platelets such that K_Q decreases by roughly a factor of three when a significant component of hydride platelets are oriented parallel to both the crack plane *and* the growth direction, as in the mixed in-plane/out-of-plane hydride case.

In the previous study mentioned above, the two hydride microstructures examined for through-thickness K_Q-behavior consisted of discrete hydride particles separated by a ductile Zircaloy matrix. However, hydride blisters that form on the surface of components such as cladding tubes are often accompanied by *sunburst* hydrides that emanate from the blister in a radial orientation. Because of their orientation, the sunburst hydrides offer a convenient crack path for the propagation of a crack that initiates within the brittle hydride blister. Thus, it seems likely that the crack growth initiation toughness within a sunburst hydride field may be significantly lower than that for discrete hydride microstructures examined previously [1], especially if the crack is co-planar with hydride platelets. The issue of through thickness fracture toughness in the presence of radial hydrides is also relevant to possible cladding failure by hydride reorientation during long term storage of spent fuel which could affect its ductility during transportation.

It is important to recognize that crack growth through the cladding wall thickness and along sunburst hydrides occurs under both stress-state *and* hydride microstructure conditions that are distinctly different from previous studies. For example, when the fracture toughness of hydrided zirconium alloys determined for the case of mode I crack growth of plate material [8-13], plane-strain crack growth creates a locally high degree of stress triaxiality, but crack growth occurs under conditions where the hydride platelets are usually not co-planar with the crack.

Alternatively, while fracture toughness has been determined under low degrees of stress triaxiality for thin-wall hydrided Zircaloy cladding tubes, crack growth was along the tube axis under a plane stress condition at the crack tip in mixed mode I/III crack propagation *and* under conditions where the crack plane also does not coincide with the plane of the hydride platelets [14]. The crack growth conditions in this study differ; through-thickness crack growth occurs under a plane-strain condition along the crack front, *and* importantly, it occurs under the condition wherein the crack plane is co-planar with the sunburst hydrides within the microstructure.

The purpose of this study is thus to investigate the room temperature fracture toughness behavior for *through-thickness crack growth into a sunburst hydride microstructure* using thin sheet specimens of cold work and stress relieved Zircaloy-4 as a model material. The experimental approach utilizes a novel procedure in which a narrow linear strip of brittle hydride blister across the specimen width creates a well-defined pre-crack upon initial loading in bending. The subsequent crack growth resistance is then characterized by tensile loading of the specimen and an elastic fracture mechanics analysis. These new results provide further insight into the relationship between crack growth resistance and hydride microstructure and should contribute to a better understanding of the failure of thin-wall Zircaloy-4 components at low temperatures under through thickness crack growth conditions.

2 Experimental Procedure

2.1 Materials and specimen preparation

The material used in this study is 0.68 mm thick Zircaloy-4 sheet, obtained from ATI Wah-Chang initially in the cold-worked (CW) state, and subsequently heat treated at 520°C in vacuum to obtain a cold-worked and stress-relieved (CWSR) state.[1] Table 1 shows that the Kearns parameters [15] are similar to those previously reported for Zircaloy-4 cladding tube [16, 17] in the three orthogonal directions. As also shown in Table 1, the yield strengths and plastic anisotropy parameter in the transverse direction of the sheet [1] are similar to those of cladding tube material.[6] However, the strain-hardening exponent (n) is about 0.009, which is significantly smaller than that of the previously studied cladding but similar to that of irradiated cladding [17].

As previously done in [1], a novel specimen preparation procedure was employed in which a narrow 200-700μm wide linear strip of hydride blister was created across the width of Zircaloy-4 sheet specimen (initially ≈ 35 mm wide) using a sequential hydriding procedure.[1] In addition to forming the linear blister, the hydrogen charging developed a sunburst hydride microstructure beneath the blister; it should be recognized that after pre-cracking through the hydride blister, these sunburst hydrides limit the crack growth initiation toughness.

Table 1: Comparison of Kearns factors for the present CWSR Zircaloy-4 sheet material with those of Zircaloy-4 cladding tube. Tensile properties of either CWSR Zircaloy-4 sheet oriented transverse to the rolling direction or Zircaloy-4 cladding tube oriented transverse to its axis.

Material	Kearns Factors			Yield Strength 0.2% (MPa)	Anisotropy factor R'	Strain-hardening exponent n
	Normal/ Radial	Transverse/ Circumferential	Rolling/ Axial			
CWSR sheet [1]	0.57	0.34	0.16	597	2.6	0.009
CWSR tube [5, 6]	0.58	0.32	0.10	610	2.3	0.068

Where $n = \dfrac{d(\ln\sigma)}{d(\ln\varepsilon)}$ and $R' = \dfrac{\varepsilon_{width}}{\varepsilon_{thickness}}$ as determined in the transverse direction.

2.2 Fracture toughness testing

Fracture toughness testing was performed using the tensile specimen configuration shown in Fig. 1. For the purpose of determining the initial crack length as accurately as possible, all fracture toughness test specimens (initially ≈35 mm in width and ≈0.68 mm in thickness) were pre-cracked at small loads (well less than yielding) in four-point bending using acoustic emission signals emitted from the specimen during bending to detect cracking within the blister. The crack depth was then determined from two length-wise strips of the sample (~5.5 mm wide) taken from each edge of the specimen. These metallographic specimens also served to characterize the hydride microstructure of each individual test specimen.

Pre-cracking of the hydride blister generated a surface crack extending across the entire width of the tensile specimens (≅ 24 mm wide), resulting in a single-edge-cracked specimen being loaded in uniform tension (analogous to a Single-Edge-Notched Tension, or 'SENT' specimen). In two of the four specimens tested, the pre-cracking procedure resulted in the formation of only one crack of well-defined length within the blister, as shown in Figure 2. In the other two specimens, while a pair of pre-cracks formed, only the longer crack propagated. After specimen fracture, the shorter crack did not display any crack opening, blunting, or growth; thus, the fracture toughness value was based on the length of the longer crack, as would be expected from fracture mechanics considerations [18].

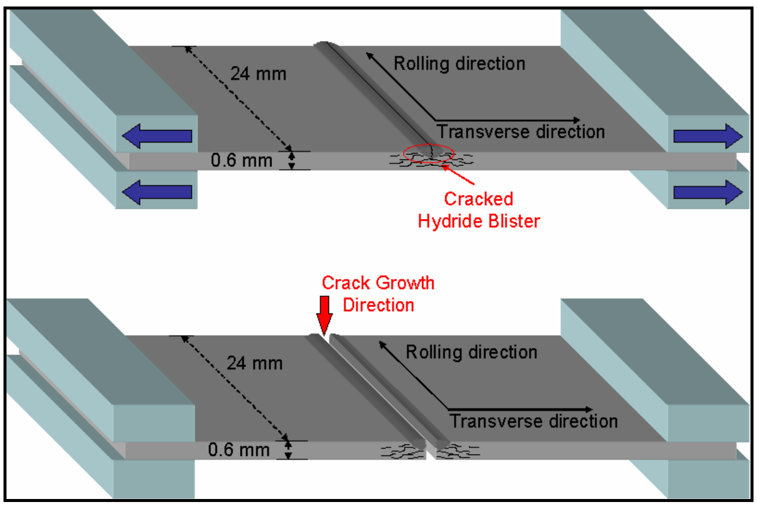

Figure 1: Tensile test configuration used for fracture toughness testing

Pre-cracked specimens with a gauge section of ≈ 53 mm long were tested in uniaxial tension at a crosshead displacement rate of 1.0 mm/min. One test failed at a net section stress equal to ~60% of the yield stress, giving rise to linear-elastic load-displacement response until failure. In the other tests, failure occurred when the net-section stress was roughly equal to the yield stress and in which case the load-displacement response deviated slightly from the linear elastic behavior prior to failure, as shown in Figure 3. In these cases, the load P_Q at the onset of crack growth was determined using the 95% secant rule as per ASTM Standard E1820 [19]. In all cases where failure occurred near yielding, the load at failure P_{max} was within 3% of the value of P_Q such that the ratio P_{max}/P_Q did not exceed the 1.10 limit for the use of linear elastic fracture mechanics. Importantly, no acoustic emission was detected prior to fracture during tensile loading, indicating the absence of any large scale crack growth of the pre-crack prior to fracture. The fracture toughness K_Q was thus calculated based on the length of the pre-crack, the load P_Q at the onset of crack growth, and the application of linear-elastic fracture mechanics for single-edge-crack geometry in uniform tension (SENT) for which [19]

$$K_Q = \left(0.265\left(1 - a/W\right)^4 + \frac{0.857 + 0.265\, a/W}{\left(1 - a/W\right)^{3/2}} \right) \frac{P_Q}{B_N W} \sqrt{\pi a} \qquad (1)$$

where a, B_N and W are the crack length, width and thickness of the specimen, respectively.

3 Results and Discussion

In the present study, the experimental hydrogen charging procedure creates not only the solid hydride blister extending across the specimen width but also a microstructure consisting of hydride particles beneath the hydride blister. All the specimens tested in this study had similar microstructural features in the region beneath the blister, where the microstructure varied with depth below the specimen surface in the manner shown in Fig. 2. Although three distinct forms of hydrides are present at different depths within the Zircaloy substrate beneath the blister, "sunburst hydrides", which emanate radially from the hydride blister, dominate the microstructure directly below the blister. Normally, the depth of the sunburst hydride zone increases with blister depth in which case the "sunburst" microstructure tends to be better developed.

The solid hydride blister is known to be brittle at temperatures at least up to 400ºC [18]. Thus as shown in Fig. 2, pre-cracking the specimen at small loads in bending causes the blister to fracture and forms a sharp crack that extends across the specimen width at near constant depth and that arrests near the interface between the blister and the sunburst hydrides that emanate from the blister. Once the pre-crack is present, the subsequent crack growth initiation behavior and K_Q-value depends on the character of the substrate hydride microstructure directly ahead of the crack tip, since it is well known that the hydride particles directly affect the fracture process in zirconium-base alloys at room temperature.

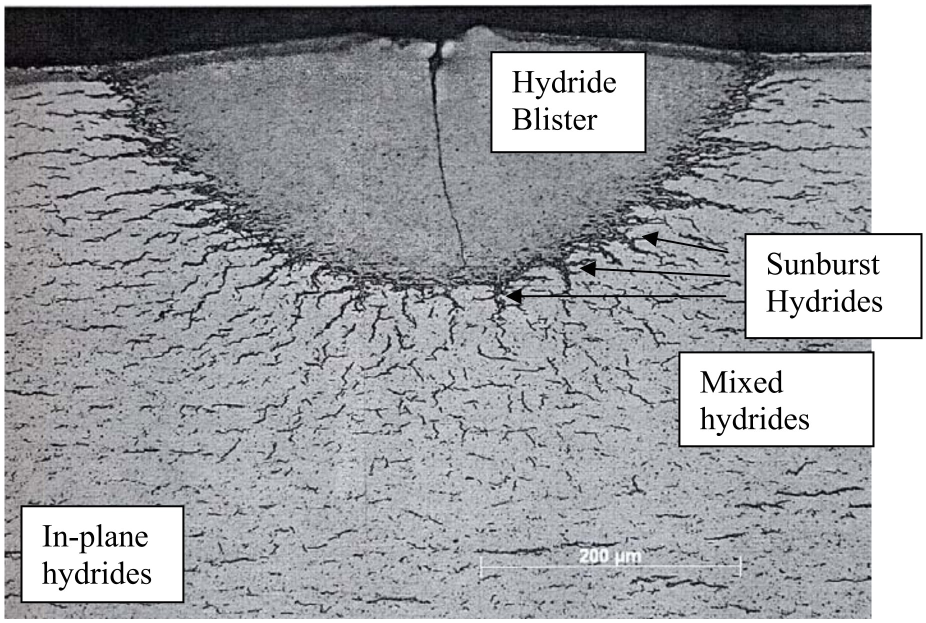

Figure 2: Light micrograph showing a pre-crack within the hydride blister. Note the sunburst hydrides emanating from the blister through which crack growth initiation occurs.

Upon tensile loading, crack growth initiation occurs into the sunburst hydride zone and unstable fracture follows immediately, as evidenced by the nearly linear elastic load-displacement curve shown in Figure 3. The four fracture toughness tests in this study show K_Q-values in the range of 10 to 15 MPa√m, which are summarized in Table 2. The smallest through-thickness fracture toughness value (K_Q = 10 MPa√m) is associated with the deepest (and most prominent) sunburst hydrides (≈85μm) and resulted in specimen failure in a linear elastic manner at roughly 60% of the net section yield stress. In contrast, specimens such as that in Fig. 2 with smaller (and less prominent) sunburst hydrides (in the range 20μm to 45μm), exhibited somewhat higher fracture toughness levels of $K_Q \approx$ 15 MPa√m. In all cases, crack growth occurred in a mode I manner through the thickness of the specimen.

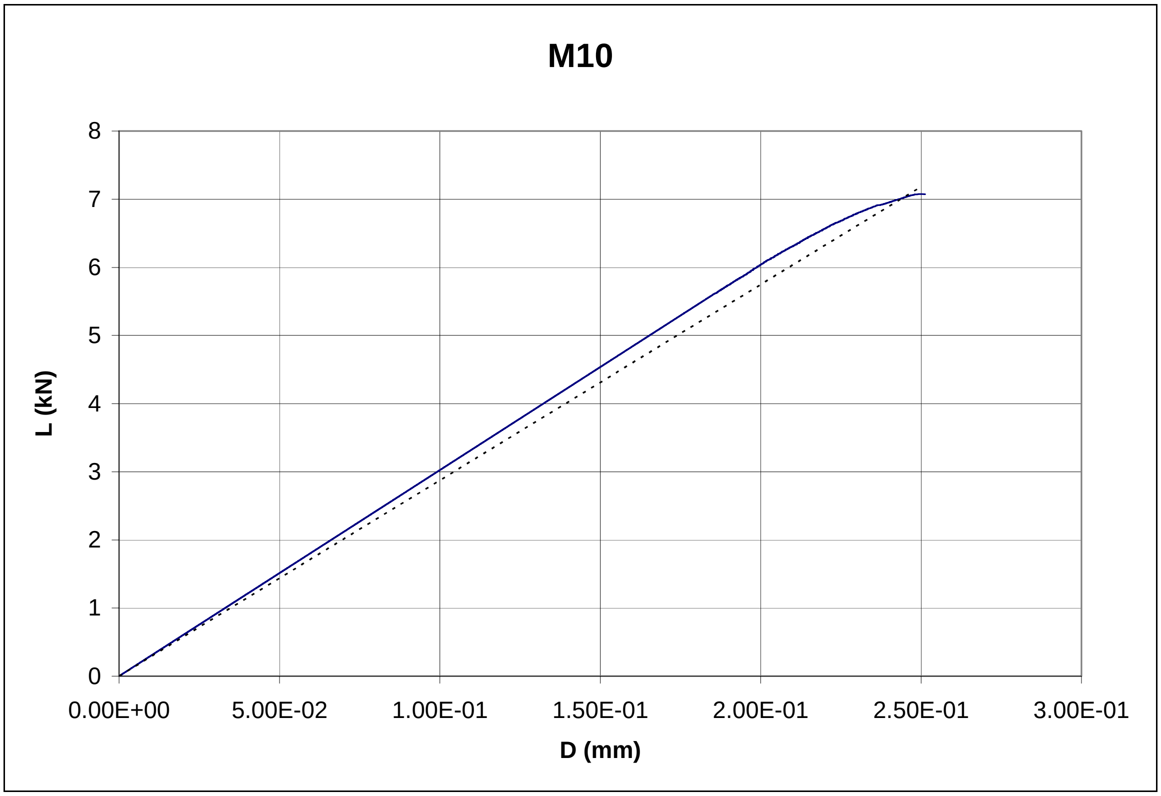

Figure 3: The load-displacement curve (solid line) of hydrided Zircaloy-4 sheet under tensile loading as compared to the 95% secant line (dashed line) as per ASTM E1820 [19]. This specimen fractured at K_Q = 15.5 MPa√m.

In addition to the sunburst hydrides, two other distinct forms of hydrides are present within the Zircaloy substrate beneath the blister. Adjacent to and below the sunburst hydrides in Fig. 2 is an extensive "mixed hydride zone" within which discrete hydride platelets with differing orientations are present. A significant fraction of these hydride platelets have their normal oriented parallel to the sample surface (akin to radial hydrides within a cladding tube), so that the platelet surfaces are parallel to the crack plane. At a further distance beneath the mixed hydride zone in Fig. 2 is an "in-plane hydride zone". This zone occupies the remainder of the Zircaloy sheet and is characterized by discrete hydride platelets that are parallel to the sample surface and thus perpendicular to the crack growth direction *and* the crack plane. These hydrides are akin to the "circumferential hydrides", typically observed in Zircaloy cladding tubes.

Table 2: The through-thickness fracture toughness (K_Q) as related to the microstructure in the region of crack growth initiation for the present study and that previous [1]. The load ratio P_{max}/P_Q at fracture is also reported for the present study.

Test configuration	Crack growth into:	P_{max}/P_Q	**K_Q (MPa√m)**
Tensile	~85µm deep out-of-plane sunburst hydrides branching from the hydride blister in a continuous fashion	1.00	**10.0**
Tensile	~45µm deep out-of-plane sunburst hydrides branching from the hydride blister in a continuous fashion	1.03	**14.8**
Tensile	~25µm deep out-of-plane sunburst hydrides branching from the hydride blister in a continuous fashion	1.01	**15.5**
Tensile	~20µm deep out-of-plane sunburst hydrides branching from the hydride blister in a continuous fashion	1.00	**14.5**
Four-point bending [1]	Random/mixed hydrides with platelet orientations both in-plane and out-of-plane of sheet.	N/A	**26 (average of 3 tests)**
Four-point bending [1]	Predominantly in-plane hydrides with significant presence of large particles that cause crack-tip branching.	N/A	**73 (average of 2 tests)**

The K_Q-values for through-thickness crack growth have been recently determined for both the mixed hydride and the in-plane hydride microstructures described above [1], and these values are also reported in Table 2. As a comparison with the tensile fracture behavior of the current study, Fig. 4 (taken from Raynaud et al. [1]) shows the corresponding crack growth process within the hydrided zone beneath the hydride blister in a four-point bend specimen. In this test, crack growth through the sunburst hydrides occurred in the *absence* of any discernible load drop prior to unstable fracture, as evidenced in the load-displacement curve shown in Fig. 4a. The first load drop at (corresponding to Fig. 4b) was associated with crack growth initiation into the field of mixed hydrides for which $K_Q \approx 26$ MPa√m. The crack then arrested in a region where in-plane hydrides began to dominate the microstructure, and subsequent loading (Fig. 4c) resulted in large-scale yielding (specimen is plastically bent) and was characterized by crack arrest and significant crack-tip deflection.

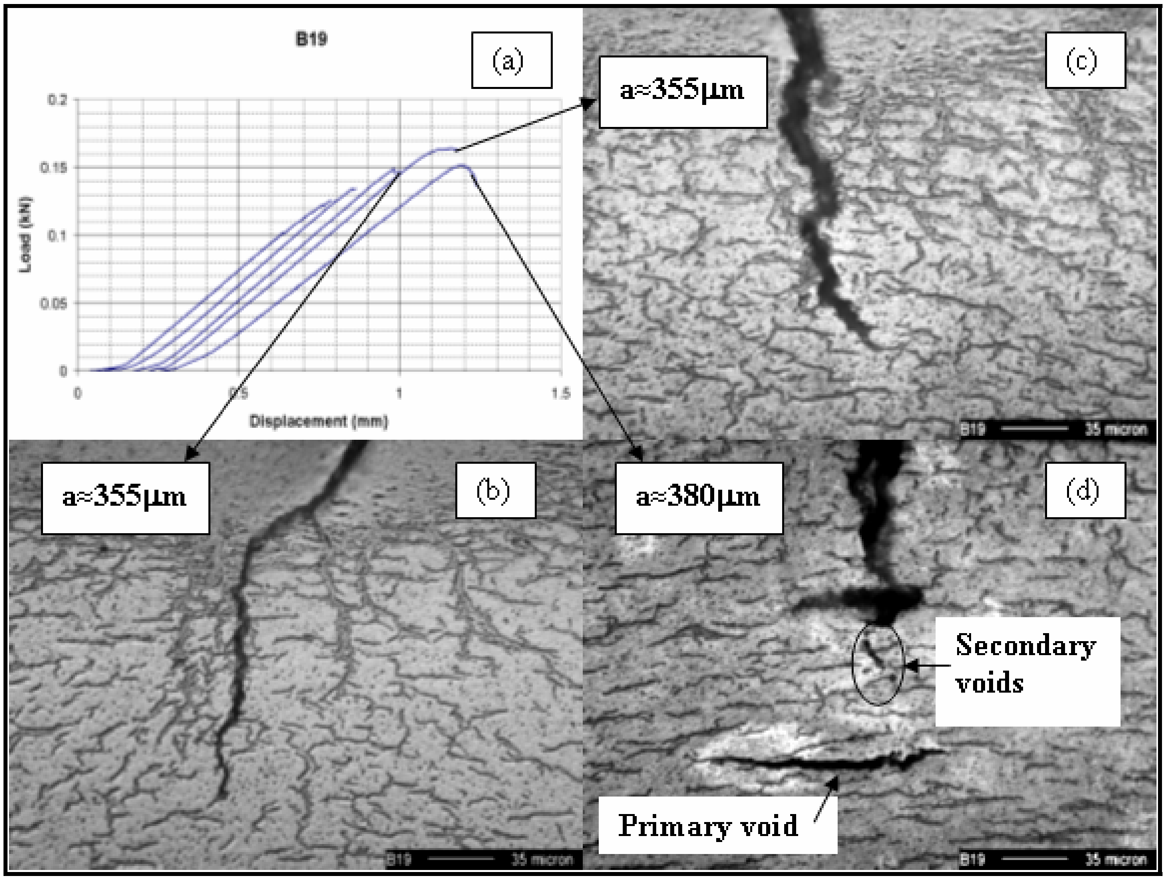

Figure 4: The evolution of the crack profile with increased bending during the interrupted four-point bend testing of a specimen with a ~215 μm blister. From Raynaud et al [1]

When compared with crack growth into mixed in-plane and out-of-plane hydrides for which $K_Q \approx 26$ MPa√m [1], the present data obtained under tensile loading shows that through-thickness crack growth initiation into the *sunburst* hydride region is characterized by a much lower fracture toughness value with an average $K_Q \approx 13.7$ MPa√m. This behavior can be rationalized on the basis that the relatively thick sunburst hydride platelets provide a nearly continuous crack path as they are oriented in the crack plane and are continuous with the hydride blister. Preliminary examinations of crack profiles and underlying microstructure support this hypothesis. This behavior as well as the dependence of toughness on the depth of the sunburst hydrides (see Table 2) suggests that the fracture resistance of Zircaloy-4 cladding decreases with increasing *continuity* of radial hydrides either in the form of sunburst hydrides (more continuous) or mixed hydrides in the form of discrete particles (and less continuous).

As also depicted in Fig. 4, crack growth into the in-plane hydride field is difficult as it requires significant plastic deformation of the specimen (Fig. 4d) to cause a small degree of crack extension (≈ 25 μm). Fig. 4d also shows that at this point in the bending process the crack tip is quite blunted (indeed in Fig. 4d, crack-tip branching has occurred) with large primary voids forming along the plane normal to the crack plane following elongated hydrides ahead of the crack tip. As a consequence of the behavior described above, K_Q is much higher ($K_Q \approx 73$ MPa√m) when the hydrides are oriented predominantly in the plane of the sheet and (importantly) normal to both the crack plane and the crack growth direction.[1] In this case, despite the presence of elongated hydride particles that initiate large primary voids, their presence also causes crack-tip deflection and crack branching which contribute to increasing the crack growth resistance.[1]

Finally, the room temperature results presented here are in agreement with previous studies by Bertolino et al. [12, 13] that suggest that the hydride microstructure into which crack growth occurs has a very significant impact of the crack growth resistance. In fact, this study suggests that radial sunburst hydrides are most detrimental to through-thickness crack growth resistance with a corresponding $K_Q \approx$ 10-15 MPa√m; mixed in-plane and out-of-plane hydrides result in a slightly higher but still low fracture toughness with $K_Q \approx 26$ MPa√m [1]. In contrast, through-thickness crack growth is difficult into an in-plane hydride field, at least at concentrations in the range of the present study for which $K_Q \sim 73$ MPa√m.[1]

4 Conclusions

1. A tensile testing procedure has been developed to determine the fracture toughness of hydrided CWSR Zircaloy-4 sheet under *through-thickness* crack growth conditions. The procedure relies on forming a pre-crack within a narrow strip of brittle hydride blister that

extends across the specimen width. The subsequent crack growth initiation conditions can be determined using linear elastic fracture mechanics.

2. The fracture toughness behavior at room temperature is sensitive to the hydride microstructure within which crack growth initiation occurs. For through-thickness crack growth into a microstructure dominated by the radial sunburst hydrides branching from the blister, $K_Q \approx$ 10-15 MPa√m. This low toughness value is consistent with the observation that the large sunburst hydrides provide a near continuous fracture path that is nearly co-planar with the initial crack
3. When combined with the results from a previous study of fracture toughness using a four-point bend test[1], the room temperature fracture toughness K_Q for through-thickness crack growth initiation into a thin sheet of hydrided Zircaloy-4 is dependent on the microstructure and varies as follows:
    - $K_Q \approx$ 10-15 MPa√m for crack growth initiation into sunburst hydrides
    - $K_Q \approx$ 26 MPa√m for crack growth initiation into mixed hydrides
    - $K_Q \approx$ 73 MPa√m for crack growth initiation into in-plane hydrides

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