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Near threshold delayed hydride crack growth in zirconium alloys

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Delayed hydride cracking (DHC) in zirconium alloys arises as a consequence of the diffusion of hydrogen atoms to a crack tip, precipitation of hydride platelets and then the fracture of a hydrided region that has formed ahead of the crack tip. This process repeats itself and, consequently, a crack grows in a series of steps. There is a threshold value, K_{IH}, of the crack tip stress intensity below which DHC crack growth is unable to proceed. The present paper provides a physical picture of the near threshold situation, accounting systematically for the manner in which hydrided material fractures, and consequently obtains an expression for K_{IH} in terms of the hydrided material's flow and fracture characteristics.

1. Introduction

When zirconium alloys contain hydrogen in excess of the TSS level (the terminal solubility limit above which hydride platelets are able to precipitate), they are susceptible to fracture by a delayed hydride cracking (DHC) mechanism. DHC is a discontinuous process. Under the influence of a stress gradient, hydrogen atoms migrate to the vicinity of a crack tip and precipitate as hydride platelets. A lenticular shaped hydrided region then grows ahead of the crack tip along the crack plane, until a critical condition is reached when the hydrided region fractures. The crack then propagates forward through the hydrided region and arrests when the crack tip is confronted by the ductile metal matrix at the front end of the region. This process keeps repeating itself, and results in a stepwise stable crack extension (when averaged out), but consisting of successive increments of unstable crack extension through hydrided material.

Most of the experimental data with regard to DHC crack growth in zirconium alloys are for cold-worked Zr–Nb, as used in CANDU (Canada deuterium uranium) nuclear reactor pressure tubes, where the specimen–crack plane configuration usually studied in experiments (these simulating actual pressure tube behaviour) is one in which the DHC crack grows in the axial direction in an axial–radial plane. Zr–Nb pressure tube material has a strong texture with basal normals in the circumferential direction, i.e. normal to the DHC crack growth plane, and since Hardie and Shanahan [1] have shown that hydride precipitation is favoured when grains have their basal crystallographic planes normal to the local tensile stress at a crack tip, such a textured material is particularly susceptible to DHC. An important characteristic [2] of DHC crack growth in Zr–Nb material is that the average crack velocity, v, shows a two stage dependency on stress intensity factor, K; in stage I, v rises very rapidly as the stress intensity increases above a threshold value, K_{IH}, until the intervention of stage II when there is little or no K dependency. The practical importance of K_{IH} stems from the fact that if a pressure tube contains a crack for which the crack tip stress intensity is less than K_{IH}, then such a crack is unable to grow by a DHC mechanism.

Against this background, it is clearly important to have an understanding of the physical parameters that affect the value of K_{IH}. With this objective in mind, and as part of a wideranging theoretical research programme on DHC initiation at stress concentrations, the author has presented [3] the results of a very preliminary attempt at modelling DHC initiation at a sharp crack. Particular features of this work were the way in which the compressive stress induced by hydride precipitation was modelled and quantified and, most importantly, the use of a critical tensile stress criterion for failure of a hydrided region. The limitations of this earlier attempt, and also a similar attempt by Shi and Puls [4], which has also used a critical tensile stress criterion, are highlighted in Section 2.1. It is against this background that the present paper goes beyond the earlier attempt [3], and presents a suggested physical picture of the way in which DHC crack growth proceeds at near threshold conditions, while obtaining an expression for K_{IH} in terms of the hydrided material's flow and fracture characteristics.

2. Background

2.1. Appraisal of earlier modelling attempts

In his earlier modelling attempt [3], the author took due account of the stress distribution in the vicinity of a sharp crack due to the applied loadings, and the geometrical parameters associated with the hydrided region since these affect the stress that is induced by the hydride precipitation (in its unconstrained state

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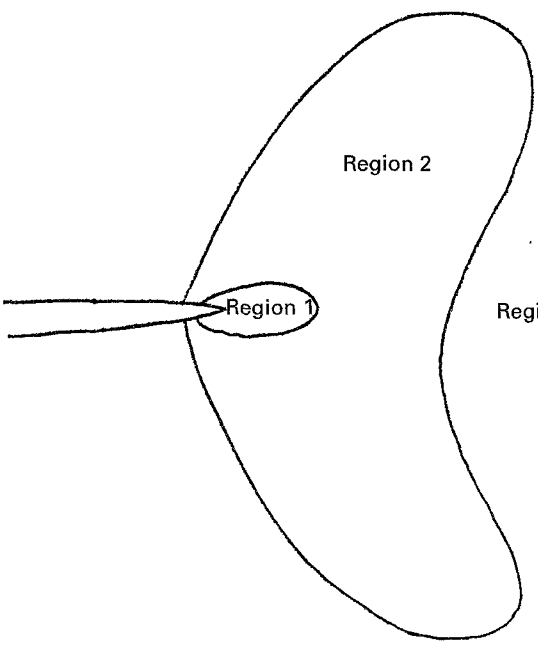

Figure 1 The various zones ahead of a crack tip.

the formation of a hydride platelet is associated with an expansion normal to the platelet). As regards the deformation pattern near a crack tip, in the absence of hydriding, essentially three different zones can be identified [5] as shown in Fig. 1.

1. Region one, a very small zone in the immediate vicinity of the tip within which there are high local strains; the size, r_I of this zone is approximately equal to twice the crack tip opening displacement, $\delta \sim K^2/E_o\sigma_o$, where σ_o is the tensile yield stress of the Zr–Nb material and $E_o = E/(1-\nu^2)$ with $E =$ Young's modulus and $\nu =$ Poisson's ratio.

2. Region two, a plastic zone surrounding the high strain local zone, the boundary of this zone being at a distance, $r_p \sim 0.32\, K^2/\sigma_o^2$ from the crack tip, when measured along the crack plane.

3. Region three, an elastic zone which surrounds the plastic zone, (region two). Rice and Johnson [5] (see also [6]) have analysed the stress pattern for small scale yielding conditions. The position of the hydrostatic stress peak is directly ahead of the crack tip and approximately coincides with the boundary between zones one and two, and following Eadie and coworkers [7], the author assumed [3] that this peak hydrostatic stress position is the location where hydride precipitation first occurs.

With respect to fracture of the hydrided material, the author focused on the magnitude of the tensile stress normal to the crack plane in the region where the hydrided region forms and grows, i.e. ahead of the crack and along the crack plane. This tensile stress has a maximum value of $\sim 3\sigma_o$ at the boundary between regions one and two and gradually decreases on moving towards the boundary between regions two and three, where the stress has a value $\sim 2\sigma_o$ [5]. In fact, apart from the very small high strain region one, the simple elastic stress value $K/(2\pi x)^{1/2}$ (x is the distance ahead of the crack tip) provides a reasonable approximation [5] for the tensile stress ahead of the crack tip. In the author's earlier work [3], a constant stress $2.5\sigma_o$ was assumed for zone two, so as to simplify the analysis.

In modelling the fracture of hydrided material, the effects of the crack tip (tensile) stress field and the (compressive) stress associated with hydride precipitation were decoupled, and the crack tip tensile stresses referred to in the preceding paragraph were used [3]. The compressive stress associated with hydride precipitation was determined by viewing the hydrided region as residing within a cracked elastic solid. A hydrided region of constant thickness, t, was assumed to extend outwards into the material from the peak hydrostatic stress position, i.e. the boundary between regions one and two. Assuming that threshold conditions correspond to the case where length-wise growth of the hydrided region was unrestricted, the author's analysis, which allowed for image effects associated with the hydrided region–crack interaction, showed that the threshold, K, value is given by the expression

$$K_{IH} = E_o\left[\varepsilon_* f_*\, t/20\pi 2^{1/2}\left(1 - \frac{\sigma_{HF}}{2.5\sigma_o}\right)\right]^{1/2} \quad (1)$$

where ε_* is the assumed hydride precipitation strain perpendicular to the hydrided region, f_* is a measure of the extent of hydriding within the hydrided region, and σ_{HF} is the tensile fracture stress of the hydrided region (it was assumed that hydrided region fracture commences at the interface between regions two and three when the total tensile stress at that position attains the value σ_{HF}). With typical input values for the various parameters, Equation 1 was used to infer that K_{IH} is in the range 5–10 MPa m$^{1/2}$, a prediction that accords with experimental results [8].

However, upon examination of Equation 1, one observes that if hydride precipitation were not to be associated with a strain, ε_*, then K_{IH} should be equal to zero; this clearly cannot be the case since fracture of hydrided material must be associated with a non-zero K value. Something is clearly missing in the K_{IH} expression, and the author now believes that this difficulty stems from the assumption that DHC initiation is linked with the concept, which the author now believes to be over simplistic, that the hydrided region fractures when the tensile stress in the region attains a critical value, σ_{HF}. Failure of an element of the hydrided region involves the fracture of hydride platelets and also the failure of zirconium alloy matrix material, and consequently some degree of plastic strain must also be involved in the failure of a hydrided region. As will be shown later in the paper, as soon as this point is accepted, and the fracture of hydrided material is accounted for in a more realistic manner, the whole character of the problem changes and the inconsistency disappears.

Shi and Puls [4] have also modelled K_{IH}, using a similar approach to that adopted by the author. However, they assumed that the (constant thickness) hydrided region (in fact a single platelet) of infinite length extended from the crack tip and not from the hydrostatic stress peak as in the author's analysis [3]. Shi and Puls argue that image effects associated with the hydrided region–crack interaction can be ignored, in contrast to the author's approach [3] which gives prominence to such interactions (see detailed discussion of the importance of image effects [9]), and assume that fracture occurs at the peak applied tensile stress position at the boundary between regions one and two. Despite these differences, Shi and Puls obtained a K_{IH} expression that is similar in form to Equation 1, though with slightly different constants. The Shi–Puls approach, as summarized here, is therefore subject to essentially the same limitation as the author's [3] due to the same assumptions of a hydrided region fracture criterion based solely on the attainment of a critical tensile stress. The remainder of the paper proceeds from the recognition that one should incorporate the concept that some degree of plastic strain is involved in the failure of a hydrided region.

2.2. A simple physical picture of near threshold crack growth in Zr–Nb material

This section provides a very simple physical picture of the near threshold stress situation proceeding from the basis that DHC is a discontinuous process. As indicated in the introduction, under the influence of a stress gradient, hydrogen atoms migrate to the vicinity of a crack tip and then precipitate as hydride platelets. A lenticular shaped hydrided region therefore grows ahead of the crack tip along the crack plane until a critical condition is reached when the hydrided region fractures. The crack therefore propagates forward through the hydrided region and arrests when it is confronted by the ductile metal matrix at the front end of the region. This process keeps repeating itself, and results in a stepwise stable crack extension (when averaged out), but consisting of successive increments of unstable crack extension through a hydrided region.

In quantifying this process, it should be recognized that there are two ways of interpreting the threshold stress intensity K_{IH}:

1. the lowest value of K at which DHC first initiates at a crack tip, and
2. the lowest value of K at which a growing DHC crack arrests.

The author's earlier research [3] and that of Shi and Puls [4] focused on the former (first initiation) situations. However, the K_{IH} value associated with first initiation (in increasing K tests) exceeds the K_{IH} value associated with an arresting DHC crack (decreasing K tests), for which the experimental database is more extensive. Accordingly, the present paper focuses on the arrest situation. The objective is to obtain an expression for the lowest K value at which crack extension will proceed from an arrested crack and this will be equated with K_{IH}.

In considering this situation, one proceeds from the recognition (see previous section) that propagation of a crack through a hydrided region involves the fracture of hydride platelets together with the failure of ligaments of ductile zirconium alloy matrix material between the cracked hydride platelets. With regard to such propagation, for Zr–Nb alloy whose crystallographic texture is such that the material is particularly susceptible to DHC, the author's picture [10] is that each increment of unstable crack extension is likely to proceed via what Cottrell has categorized [11] as a cumulative mode, so called because with a formal dislocation description of the process, the contribution made by a dislocation moving (climbing) through the material ahead of the crack, accumulates continuously with the distance travelled by the dislocation. Thus, the propagation of a crack tip through a hydrided region during each unstable increment of crack extension can be viewed in terms of the tip being associated with a fracture process zone and that there is a stress, p_*, (viewed in an average sense) at its leading edge. p_* is either the tensile stress required to crack hydrided material within a lenticular shaped hydrided region or it is the maximum stress that the region can sustain as the bridges between the cracked hydride platelets deform and rupture, whichever stress is the larger. On the basis of acoustic emission experiments, Simpson [2] (see also [12]) has taken the view that hydride platelets within a hydrided region are able to fracture fairly easily, presumably much more readily than the single platelets as studied by Puls [13], because of the enhanced strain incompatibility effects within a hydrided region, and that rupture of the intervening ligaments is the controlling mechanism. If this view is accepted, then p_* is the stress associated with the deformation and rupture of the bridges between the microcracks caused by fracture of hydride platelets. The crack arrests because there is no longer hydrided material to fracture, and the arrest is associated with a K value, K_{ARR}, which will be quantified later. If the applied K value exceeds K_{ARR}, then there will be plastic deformation in the vicinity of the arrested crack tip, which manifests itself as a striation on the fracture surface, a characteristic of DHC fracture surfaces [2, 8, 12, 14]. Hydrogen atoms then diffuse to the high stress region ahead of the arrested crack and eventually a critical condition will be reached in the vicinity of the arrested crack tip when there is another increment of unstable crack propagation through a hydrided region. With this picture K_{IH}, as determined in a K reduction experiment, is the lowest value of the applied K at which this sequence of events is able to operate. The author believes that K_{IH} is not overly in excess of K_{ARR}, a view that is supported by the experimental observation [2] that striations are observed on most (but not all) DHC fracture surfaces. There will clearly be a scatter in K_{ARR}, for reasons that will soon become apparent, and this means that the extent to which striations appear, which depends on the difference between the applied K and K_{ARR}, will show variability. The author believes that K_{ARR} provides a reasonable estimate of K_{IH}, though recognizing that there will be variability in these values.

In quantifying K_{ARR}, one recognizes that immediately behind a crack tip at the instantaneous moment of arrest, there is a fracture process zone consisting of fractured hydride platelets and unfractured ligaments of ductile zirconium alloy matrix material. Thus in very simplistic terms, and against the background of the comments in the preceding paragraph, one immediately sees that the threshold value of K, i.e. K_{IH}, required for reinitiation of DHC at the arrested crack tip is given by the very simple expression

$$K_{IH} = (E_o p_* v_*)^{1/2} \tag{2}$$

where v_* is the displacement between the crack faces that is required to rupture the ductile zirconium alloy ligaments between the cracked hydride platelets. As indicated earlier, p_* is the stress associated with the deformation and rupture of the bridges between the microcracks, and implicit in the formulation of Equation 2 is the assumption that the stress (when averaged out) is constant (with a value p_*) within the fracture process zone, an assumption that allows for a very simple description of the DHC mechanism. $p_* \sim (1-f) \times q\sigma_o$, where σ_o is the tensile yield stress of the matrix material, f is the areal fraction occupied by cracked hydride material and q is a plastic constraint factor, i.e. the elevation of yield strength due to the mechanical constraint associated with the ligament regions, with the maximum value of q expected to be ~ 3 [11]. Since the cumulative semiductile mode of crack propagation through hydrided material is only able to operate if p_* is less than the tensile yield stress, σ_o, of the material [11], it follows that f must exceed $\sim 2/3$ if $q \sim 3$. As indicated earlier in the paper, Zr–Nb material has a crystallographic texture with a predominance of basal phase normals perpendicular to the DHC crack growth plane, and consequently this condition is expected to be satisfied. Expression 2 for K_{IH} can therefore be written in the form

$$K_{IH} = [3(1-f)E_o\sigma_o v_*]^{1/2} \tag{3}$$

As indicated earlier, v_* is the displacement between the crack faces that is required to rupture the ductile zirconium alloy ligaments between the cracked hydride platelets. Even though v_* is a displacement viewed in an average sense, this will display scatter, as also will the texture parameter, f, and it is believed that scatter in these parameters are responsible for the variability observed in K_{IH} measurements.

Before concluding this section it is worth mentioning that the present section's perspective of near threshold crack growth in Zr–Nb material, and its emphasis on the fracture toughness of hydrided material via a fracture process zone concept is very much in accord with views expressed both by Simpson [2], and also by Eadie and Smith [12], the latter having emphasized the usefulness of the concept of a "semicohesive zone", stemming from ideas developed by Moody and Gerberich [15, 16] for hydride cracking in titanium alloys. However, the picture of near threshold crack growth as presented in this section, contrasts with that presented by the present author in his initial attempt [3], and also with the approach developed by Shi and Puls [4]. These earlier attempts gave, what the author now believes to be, undue prominence to the effect of hydride precipitation strains; the limitations of these earlier attempts have been highlighted in Section 2.1, and have provided the motivation for what the author now believes is the more realistic picture as developed in this section.

3. Discussion

This paper has been concerned with DHC crack growth in Zr–Nb pressure tube material, with consideration being focused on the near threshold situation. A systematic account has been taken of the way in which hydrided material fractures during DHC crack growth and the author therefore believes that the work described in the paper is a marked improvement upon the earlier attempt [3] at modelling the near threshold situation. The objective has been to formulate a criterion for reinitiation of DHC at an arrested crack tip, and the lowest value of K for reinitiation has been equated with the threshold stress intensity, K_{IH}. Thus, a very simple expression (Equation 3) has been obtained for K_{IH}, and this expression recognizes that failure of hydrided material involves both the fracture of hydride platelets and the rupture of ductile matrix ligaments between the cracked platelets, with the latter process believed to be the controlling step in the failure of a hydrided region.

Proceeding from the basis that K_{IH} is given by Equation 3, it is immediately seen that texture (through the parameter f) has a pronounced effect on the K_{IH} value. For Zr–Nb material that has a pronounced texture, a not unreasonable value for f is $f = 0.75$. On the other hand, Zircaloy-2 has a less pronounced texture (smaller f) and it is expected that, other things being equal, K_{IH} should be higher; this prediction is in accord with experimental results for Zircaloy-2, for which K_{IH} is up to a factor of two higher than it is for Zr–Nb material [8]. It is not easy to assess the effect of material strength or irradiation on K_{IH}, since, while increasing the yield stress, σ_o, these are also likely to reduce the rupture ductility, v_*, of the matrix ligaments, an effect which acts in the opposite direction. With a K_{IH} value ~ 5 MPa m$^{1/2}$, $f = 0.75$, $\sigma_o = 700$ MPa, $E_o = 80 \times 10^3$ MPa, Equation 3 shows that $v_* \sim 0.7$ µm, which is a small fraction of the grain size and a not inappropriate value for ductile failure of ligaments between cracked hydrides.

Before concluding this discussion, it is worth noting that the considerations in this paper have been essentially descriptive. Ongoing research is aimed at quantifying these considerations in the following areas

1. the progressive growth of a crack through a hydride region prior to its arrest,
2. the situation at the arrested crack tip as a specimen rings up to the applied K level, and
3. the reinitiation of fracture at the arrested tip and the influence of hydride distribution, taking into account residual damage at the arrested tip.

Accounts of this work will be the subject matter of subsequent publications.

4. Conclusions

The paper has provided a physical picture of near threshold DHC crack growth in Zr–Nb pressure tube material, accounting systematically for the manner in which the hydrided material fractures, and an expression has been obtained for the threshold stress intensity, K_{IH}, in terms of the hydrided material's flow and fracture characteristics.

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