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ON THE INFLUENCE OF AN EMBRITTLED RIM ON THE DUCTILITY OF ZIRCALOY CLADDING

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ABSTRACT

In a reactivity-initiated accident (RIA), the Zircaloy cladding must survive the deformation caused by fuel expansion and pellet-cladding interaction from a prescribed fuel energy deposition. In this paper we examine the ductility of Zircaloy cladding under RIA-like conditions in the context of sheet metal deformation and failure. In particular, we explore the susceptibility of the cladding to failure due to a localized necking instability which can cause failure at expectedly small strains. We show that localized necking failure is especially sensitive to the presence of surface flaws, such as might be introduced by fracture within a thin rim of hydrided cladding. A straight-forward application of existing localized necking analyses is used to predict the increment of cladding strain required to trigger a localized necking failure once a surface flaw is created. The analysis indicates that, given the deformation behavior of irradiated cladding and the likely deformation path imposed during an RIA event, the thickness of any pre-existing embrittled rim is critical in determining the ductility of cladding and its survivability during the RIA.

I. INTRODUCTION

In a light water reactor (LWR), a control rod ejection or drop can initiate a reactivity initiated accident (RIA). One of the consequences of a RIA is a large instantaneous energy deposition in the fuel. The resulting increase in fuel and cladding temperature, with the attendant fuel expansion and pellet cladding interaction can lead to fuel cladding failure.[1] Recent results indicate that the energy deposition threshold for cladding failure decreases significantly as the fuel burnup increases.[2] Several experiments performed at different facilities have recently become available that, coupled with previous U.S. experiments, suggest that during a RIA event failures at lower energy deposition values are possible in high burnup Zircaloy cladding. In an experiment performed in the CABRI facility in France[3], Zircaloy-4 cladding irradiated to 65 Gwd/ton failed at a peak averaged fuel enthalpy of 30 cal/g. Failure of the cladding wall occurred due to crack growth with little or no thinning of the cladding wall and on a plane normal to the cladding surface (i.e. radial cracks). In contrast, for the HBO experiments conducted in the NSRR facility in Japan, a different failure mode was observed.[4] Occurring at lower temperatures (~50° C) and much lower hydrogen content (~190 ppm average), failure in these experiments is in some ways of greater safety concern. First, despite cladding failure, plasticity of the cladding (~2%) is reported in the form of hoop extension with comparatively little axial extension of the cladding. Thus, the cladding appears to deform plastically in a near plane-strain tension path transverse to the cladding axis. Second, the failure of the HBO cladding has a much different fracture profile than in the CABRI experiment. Post-irradiation examination shows the cladding has many surface cracks (< 0.1 mm long) which have arrested within the ductile cladding. The resulting fracture profile thus involves short segments of fracture surface normal to the cladding surface combined with shear on planes inclined about ± 45° through the thickness. Thus, while most of the HBO-1 cladding thickness exhibits good crack growth resistance, failure occurs with relatively little cladding extension, dominated by through-thickness slip. It is also significant that there is a significant concentration of hydrides are present within a rim (about 50μm) near the outer surface of the HBO-1 cladding, and that the cladding from a companion test which did not fail, HBO-3, contained a thinner rim of hydrided material, approximately 24 μm.

The purpose of this communication is to examine the sensitivity of the ductility of Zircaloy cladding to the presence of surface flaws such as might form due to failure within an embrittled rim, as would occur if excessive hydriding occurred near the outer surface of the cladding. We focus here on the role of the cladding while recognizing that the evolution of the fuel microstructure may play an important role in the process.[5] It is well known that during reactor exposure, the Zircaloy cladding undergoes corrosion, accompanied by hydrogen pickup.[6,7] Both the corrosion and the hydrogen pickup can lead to the formation of a brittle rim in the cladding after long reactor exposures. Since the thickness of the oxide layer can vary within a given sample, a non-uniform brittle cracked rim can be formed by uniform corrosion. Likewise, hydrogen pickup and redistribution to colder regions of the cladding can lead to the formation of a non-uniform hydrided rim layer. The object of this paper is to present an analysis showing that the formation of a cracked brittle rim can severely limit the cladding ductility under the combination of deformation conditions imposed during an RIA.

II. ON LOCALIZED NECKING AS A FAILURE CRITERION

Our basic premise is that the thin-wall Zircaloy cladding obeys sheet metal plasticity as it deforms during a RIA event. Basically, this assumption means that through-thickness slip dictates a condition of plane-stress deformation which is typical of sheet metal with thicknesses of approximately 2 mm of less. The thickness of the HBO cladding, which is about 0.6 mm, easily meets this deformation criterion. Given a condition of sheet metal plasticity, the important issue that we address here is the <u>susceptibility of sheet metal to failure due to a localized necking instability</u>. This failure mode differs from that usually assumed for cladding failure; i.e., either crack growth leading to brittle failure, or global accumulation of damage throughout the cladding thickness, leading to a general damage accumulation type of failure. The important feature of localized necking is that, as an alternate mode of failure, it can cause cladding fracture <u>before</u> any global failure criterion, such as strain energy density[8], is satisfied. Thus, it is possible that a localized necking instability may be the controlling failure criterion in a RIA-induced cladding failure, such as that in HBO-1.

III. THE ANALYSIS

The failure of sheet metal, especially with regard to sheet metal forming, received considerable attention in the 1970's and 1980's (see references 9-11, for example). The analysis presented below is a straight-forward application of basic formability concepts in which the ductility of sheet under multiaxial deformation is limited by the onset of a localized necking instability. The fundamental characteristic of localized necking is that through-thickness deformation can concentrate within a narrow band of material such that the adjoining material ceases to deform. The maximum principal strain within the material at the onset of localized necking thus determines fracture strain <u>if</u> no other failure mechanism (such as crack growth) occurs at smaller strains. Furthermore, since final failure occurs as a result of intense through-thickness deformation on planes of high shear stress, the orientation of the failure plane is ± 45° to the sheet surface. As will be shown below, localized necking is especially sensitive to the presence of linear imperfections in the thickness (or strength) of the sheet. However, it is important to recognize that localized necking failure of sheet will occur even in the <u>absence</u> of an imperfection.[12]

With the susceptibility of cladding to RIA-induced failure in mind, we present an analysis which indicates that a thin layer (or rim) of embrittled material, which fails at small strain, creates a thickness imperfection which, acting as a surface flaw, triggers localized necking. The result is a serious degradation of the ductility of otherwise undamaged cladding. Our starting point of this analysis is that the cladding contains a thin outer layer or rim of metal which has been embrittled due to a combination of radiation damage, hydriding,

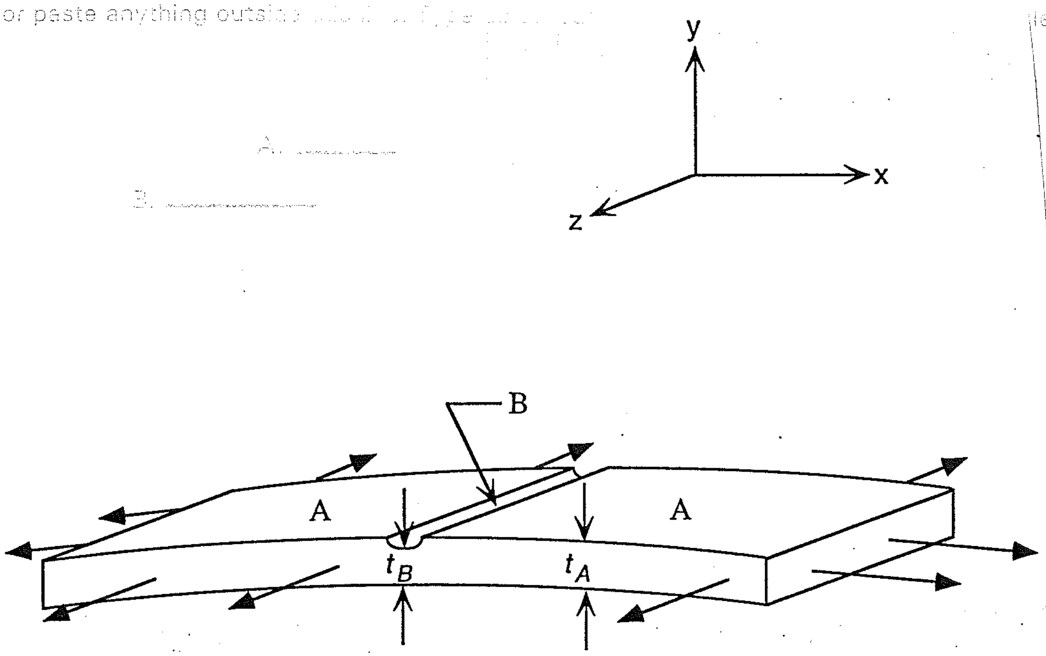

Figure 1. A schematic drawing indicating an imperfection reducing the thickness of the sheet from t_A^o to t_B^o. Plane-strain conditions are indicated during sheet extension.

and oxidation. The fundamental characteristic of this layer is that, while it is load-bearing, it fails at a small plastic strain. This behavior is common in Zircaloy cladding containing a rim of material in which hydrides have formed. The following analysis predicts the remnant ductility of the cladding after failure of the embrittled rim. The geometry, shown in Figure 1, consists of two regions, A and B, of initial thicknesses t_A^o and t_B^o.

If localized necking controls the cladding failure strain, $\bar{\varepsilon}_f$ then

$$\bar{\varepsilon}_f = \bar{\varepsilon}_e + \Delta\bar{\varepsilon}_\gamma \tag{1}$$

where $\Delta\bar{\varepsilon}_\gamma$ is the equivalent plastic strain increment to initiate localized necking once the embrittled layer fails at $\bar{\varepsilon}_e$. To estimate $\Delta\bar{\varepsilon}_\gamma$ we assume, as is the case in the MATPRO data compilation[13], that the material obeys the power law hardening relationship.

$$\bar{\sigma} = K\bar{\varepsilon}^n\bar{\varepsilon}^m \tag{2}$$

where $\bar{\varepsilon}$ is the equivalent plastic strain, $\bar{\varepsilon}$ is the strain rate, n is the strain hardening exponent, m the strain-rate hardening exponent, and K the strength coefficient. In assuming isotropic plasticity, we recognize the inherent anisotropy of Zircaloy but also note that the onset of localized necking should be relatively insensitive to plastic anisotropy for the case of plane strain tension[14] expected to be present in this case.

From equilibrium we can write

$$\sigma_A t_A = \sigma_B t_B \tag{3}$$

Defining the severity of the flaw as

$$f = 1 - \frac{t_A^o}{t_B^o} \tag{4}$$

and using von Mises' flow theory, we can rewrite equation 3 as[11]

$$\left(\varepsilon_1^A\right)^{n/m} \exp\left(-\varepsilon_1^A / m\right) d\varepsilon_1^A = (1 - f)^{1/m} \left(\varepsilon_1^B\right)^{n/m} \exp\left(-\varepsilon_1^B / m\right) d\varepsilon_1^B \tag{5}$$

where ε_1^A and ε_1^B are the principal strains in regions A and B. Equation 5 permits us to predict the development of a localized necking instability as a function of the severity of the imperfection *f*, the strain hardening exponent n, and the strain-rate hardening exponent m. Of these three variables, two are material properties, n and m, while *f* is dependent on the depth of the surface flaw created by the fracture of the brittle rim of the cladding. For sheet metal or cladding whose ductility is limited by localized necking, the "limit" strain, which is the far-field strain at the onset of necking, is the measure of ductility. It should be recognized that the limit strain value does not include the strains within a localized neck, and thus its value will necessarily be less than the more commonly used "elongation to failure" strain.

In assuming the constitutive relation in Equation 2 and the resulting relation given by Equation 5, we ignore any contribution from elasticity. Introducing an elastic component of strain introduces only minor effects on this plasticity-based analysis. This conclusion is supported by a preliminary analysis in which we incorporate the elastic yield strain through the use of the Ludwik equation:[13]

$$\sigma = \sigma_o + K\varepsilon^N \tag{6}$$

taking proper care to use the correct value of the strain-hardening exponent in that relationship (note, the n-value from Equation 2 does not equal the N-value in Equation 6). The analysis shows that the limit strain increases by at most 1% for small imperfections, and much less for large imperfections, when the elastic contribution is considered, so it appears to be justified to neglect elastic strain in the derivation above.

IV. APPLICATION OF THE ANALYSIS TO ZIRCALOY CLADDING

In order to predict the remnant ductility of Zircaloy cladding after the failure within a thin rim of embrittled material, we apply the above analysis under the following conditions:

- the power law constitutive equation given by Equation 2 relating stress and plastic strain, as used in MATPRO compilation.[13]
- plane-strain hoop extension of the cladding.
- isotropic plasticity assuming von Mises flow theory (plastic anisotropy should have no effect on localized necking under plane-strain conditions,).[14]
- a long wavelength assumption for the imperfection such that plane stress is maintained and the stress triaxiality due to the blunted crack is ignored (the thin wall of the cladding and relative ease of through-thickness slip support this assumption).
- RIA-induced cladding heating is ignored, although it will enhance localization once it is initiated.
- any irradiation-induced deformation softening is ignored; if present, this effect would also accelerate the onset of localized necking.

In order to apply the analysis, the strain-hardening exponent, n, and the strain-rate hardening exponent, m, must be known. For unirradiated, cold-worked Zircaloy 4, we have determined that in transverse tension as well as compression, n=0.052 and m=0.017. Data for annealed Zircaloy show that the presence of hydrides does not significantly change either strain hardening or strain-rate hardening, although ductility is reduced.[15] However, it is well known that irradiation decreases the n-value to about n=0.02.[13] As will be shown later, this decrease in work hardening has a pronounced effect in making the cladding more susceptible to localized necking failure.

The other parameter which is critical to the localized necking phenomena is the severity of the imperfection. We view the imperfection as a change in the wall thickness of the cladding with a linear

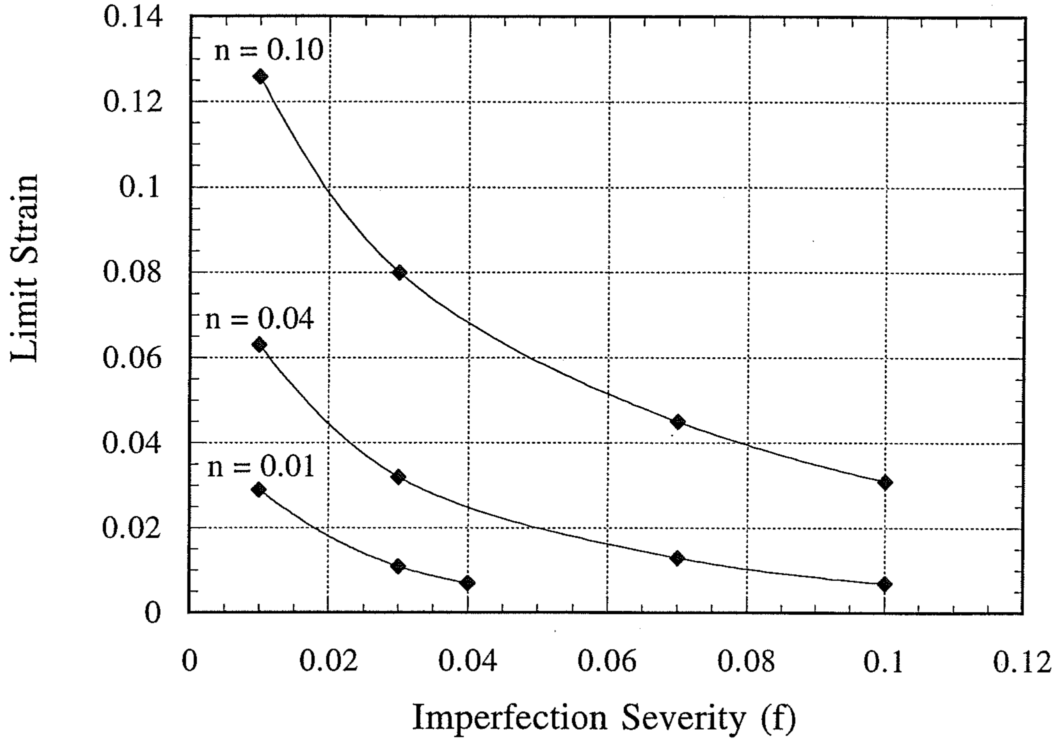

Figure 2. The dependence of the limit strain outside a localized neck on the imperfection severity and strain hardening exponent, n, of the cladding. Plane-strain deformation with m = 0.02 is assumed.

dimension, as in Figure 1. Thus, a blunted axial crack which forms due to the failure of an embrittled rim (due to excessive hydriding) would constitute an imperfection. The material outside the imperfection is assumed to remain load-bearing with similar stress-strain response to that of the thinner region within the imperfection. This assumption is supported by the stress-strain response of hydrided Zircaloy[15], mentioned above. Furthermore, the presence of adjacent cracks should have little influence on this analysis unless their density exceeds that level which reduces the load-bearing capacity of the rim, at which point the cracks would cease to form anyhow due to the decreased normal stresses. The fracture mechanics of parallel cracks indicates that mode 1 cracks spaced closer than about 4 times the crack depth will have significant interactions[16], suggesting that load shedding occurs to a level of approximately 10%. For a hydrided rim 50 μm thick, the above indicates that adjacent cracks would have to be within 200 μm of each other in order to have a large influence. We believe that the cladding will have failed due to localization near a particularly deep crack long before such a crack density develops.

Applying Equation 5 to the failure of Zircaloy cladding after an imperfection forms due to cracking of an embrittled rim (or possibly non-uniform oxidation which also results in a linear thinning of the cladding), we depict the development of localized necking instability in Figure 2. The strain-hardening values and strain-rate hardening values chosen are typical of the ranges for Zircaloy cladding before and after irradiation exposure. Most important is the influence of irradiation in decreasing the strain hardening capacity of the Zircaloy, as reported in MATPRO.[13] As seen in Figure 2, decreasing the n-value has a pronounced effect in making the cladding susceptible to failure due to localized necking soon after the formation of even a small imperfection.

The sensitivity of localized necking failure to the combination of strain hardening and flaw severity is shown in Figure 3. Clearly the far-field strain which can accumulate outside the flaw is severely limited by the combined presence of even a shallow flaw (for example, 3% of the cladding thickness) in a heavily irradiated cladding with a

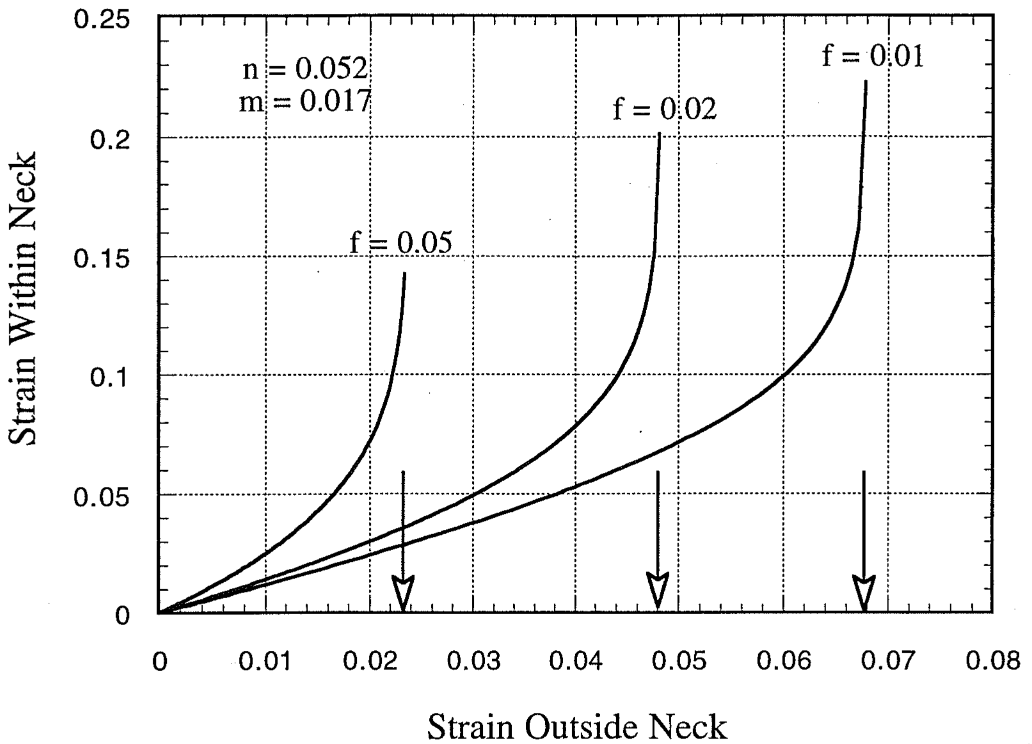

Figure 3. The development of a localized necking instability for the plane-strain deformation of cladding containing imperfections of differing severities, *f*. Strains are maximum principal strain values, and arrows denote the limit strain for each case.

small work hardening capacity. For example, using n=0.02 and m=0.02 typical of irradiated Zircaloy[13], the analysis predicts that such a small imperfection (*f* = 0.03) can trigger a necking instability which reduces the limit strain to 0.017. For cladding with a wall thickness of 0.6 mm, this means an embrittled rim of only 18 μm would limit the hoop strain at failure to only 1.7% in excess of the strain which fails the outer embrittled rim. The fractographic profile of such a failure sequence would consist of a fracture surface segment normal to the cladding surface for approximately 18 μm followed by a 45° through-thickness shear fracture. The features of this failure mode appear to correspond well with the failure observed in NSRR.[4] It should be recalled that ignored in the present analysis is any flow softening due to irradiation damage, which should accelerate the localized necking development.

The analysis presented above is also supported by the observations of Pettersson et al.[17] who studied the influence of irradiation on the ductility and defect sensitivity of recrystallized Zircaloy tubing. These researchers found that the ductility of the tubing subjected to plane-strain burst tests is much more sensitive to surface flaws in the irradiated condition than in the unirradiated state. Furthermore, the defect sensitivity is much reduced tests conducted on ring specimens where the loading is uniaxial.[18] The authors concluded that both the plane-strain deformation path and the low level of work hardening in the irradiated condition are important in determining defect sensitivity.[17]

V. ON THE INFLUENCE OF DEFORMATION PATH ON CLADDING FAILURE

Pettersson et al.[17,18] and others[19] have long recognized the importance of deformation path in influencing the ductility of Zircaloy cladding. As a result, burst tests are commonly used to determine failure strains. A main point of this paper is that cladding failure due to the onset of localized necking is also very sensitive to strain path. In particular, the near plane-strain deformation path imposed during a

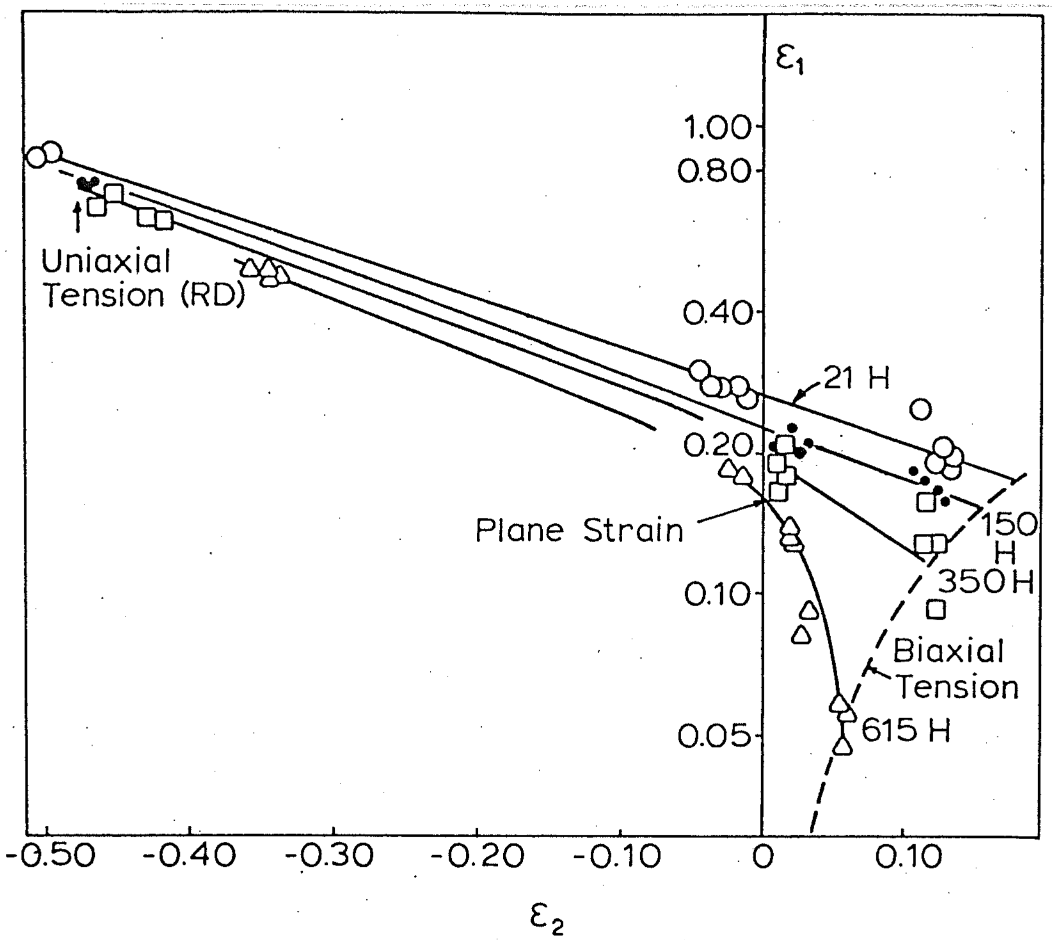

Figure 4. A fracture limit diagram for ZIRCALOY-2 sheet at four levels of hydrogen. The major, ε_1, and minor, ε_2, principal strains in the plane of the sheet at fracture are shown (after ref. 15).

burst test enhances the localized necking instability such that the failure strain is predicted to be approximately 50% of that in uniaxial tension for a plastically isotropic sheet.[12] This effect does <u>not</u> depend on the presence of an imperfection, as the original Hill analysis showed.[12]

For a sheet material with a strong degree of plastic anisotropy, the differences in failure strains between uniaxial and plane-strain tension can be even more dramatic than those described above. Figure 4 shows the influence of deformation path on the ductility of unirradiated, annealed but plastically anisotropic Zircaloy 2 sheet. The data in Figure 4 are presented in terms of the major and minor principal (true) strains, ε_1 and ε_2, in the plane of the sheet at fracture. Two conclusions, pertinent to the present discussion, can be readily made. First, there is a large decrease in extensional ductility between uniaxial and plane-strain tension for unhydrided sheet failing due to the formation of localized necking; for example, the data in Figure 4 indicate a failure strain decrease from ε_1=0.80 in uniaxial tension to ε_1=0.28 in plane-strain tension. This "ductility loss" can be predicted by the theory of localized necking failure for plastically anisotropic sheet[14], but it <u>cannot</u> be predicted by the SED failure criterion.[8] Second, for sheet containing hydrides, fracture due to hydride-induced damage (i.e., voids nucleated at cracked hydrides[15]) intervenes before localized necking can develop; the result is that ductility is further reduced, especially in multiaxial tension. The worst possible case is equal biaxial tension, such as would be the case if there were a very strong pellet-cladding bond or friction during the fuel expansion in an RIA. Such pellet-cladding bond is often found in high burnup fuel.

The implications of Figures 3 and 4 with regard to cladding containing a rim of hydrided material and undergoing a RIA event are significant. Figure 4 suggests that a rim of heavily hydrided cladding which is subjected to plane-strain (or even worse, equal biaxial tension) fails with relatively little plastic strain. The present analysis indicates that the ensuing crack can then act as an imperfection resulting in cladding failure with only very limited remnant ductility as suggested by Figures 2 and 3. Given the inherently low strain hardening of the irradiated cladding and the near-plane-strain path likely to be imposed during the RIA, the cladding thus deformed appears to be susceptible to failure at relatively small hoop extensions. Under such a failure sequence, a critical issue which must be addressed is the depth of any "heavily" hydrided rim layer. A thick rim of hydrided material (for example, 50 μm or more, such as HBO-1) on thin-wall, high burn-up cladding (with inherently low strain hardening) provides the very ingredients necessary for failure with only a minimal amount of cladding extension. Thus limiting the thickness of any rim of hydrided material becomes critical, which is perhaps why HBO-3, with a 25 μm rim of hydrides, did not fail during a recent RIA test.[4]

VI. SUMMARY

The basic premise to the analysis presented here is that thin-wall Zircaloy cladding obeys the deformation and failure behavior common to sheet metals. As such, the cladding is susceptible to failure as a result of a localized necking instability, which is known to be very sensitive to the presence of surface flaws. The flaw sensitivity issue has been examined in the context of a well-known "imperfection" analysis which is based on the influence of a linear imperfection in sheet thickness (or strength) on the onset of localized necking failure in sheet metal. The analysis predicts that the combination of a plane-strain deformation path and a low strain hardening capacity make the cladding especially sensitive to the presence of flaws. Both of these factors appear to be present and imposed on high burn-up cladding during a RIA event. Furthermore, existing failure criteria, such as the strain energy density parameter, are incapable of accurately predicting such failures. An accurate failure code should take into account the deformation behavior inherent in high burn-up cladding, its sensitivity deformation path as well as surface flaws, and the susceptibility of the cladding to the formation of surface flaws of a known depth.

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