On the Issue of Zircaloy Ductility during a Reactivity-Initiated Accident

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Abstract

During reactor exposure, Zircaloy cladding undergoes various microstructural changes including irradiation damage, oxidation, and hydrogen pick-up. There is a concern that the combination of these changes in high burnup cladding will cause failure during a reactivity-initiated accident (RIA) at an energy deposition level significantly lower than that of fresh cladding. In RIA conditions, the cladding must withstand loading at high strain rates and under deformation paths close to transverse plane-strain extension. Thus to assess cladding failure it is necessary to examine the failure mechanism of unirradiated Zircaloy cladding under RIA-like loading conditions. We present here a theoretical analysis of a possible failure mode of Zircaloy cladding due to localized necking. The results of the analysis suggest that high-burnup cladding is susceptible to pronounced losses of ductility under a combination of plane strain loading deformation and the presence of thickness imperfections. Such imperfections may be caused by hydride embrittlement of the cladding or non-uniform oxidation such that an axial thickness change is created.

1. Introduction.

In today's nuclear power reactors, zirconium alloys are commonly used for fuel cladding and structural components because of their combination of low neutron absorption, superior high temperature corrosion resistance, and adequate mechanical properties. There is a substantial body of in-reactor experience with these alloys, especially Zircaloy-2 and Zircaloy-4 used in BWRs and PWRs, respectively. The in-reactor performance of these alloys has been good, allowing the cladding to fulfill its primary function of containing the fission products and providing structural integrity to the fuel rods under normal operating conditions [1].

In an effort to increase the capacity factor of nuclear power plants, utilities and fuel vendors are currently proposing to increase fuel burnup from the current 30 GWd/ton to 45 and 60 GWd/ton, while also increasing the fuel cycle from 12 to 18 and 24 months. These changes will help reduce fuel outages and allow nuclear energy to be more competitive. However, the proposed burnup increase will double the exposure time of the cladding in the reactor environment, where it is exposed to concomitant neutron irradiation, oxidation and hydriding. It is mandatory that the fuel continue to operate safely under these more demanding conditions, both under normal operation and accident conditions.

One of the postulated analysis accidents is the reactivity-insertion accident (RIA), caused by a control rod drop or control-rod ejection. Recent full-scale simulations conducted at the CABRI reactor in France, the NSSR reactor in Japan and at the Kurchatov Institute in Russia [2] appear to show that, as the burnup increases above 45 and 50 GWd/ton, there is a decrease in the ability of the cladding to withstand the large energy deposition attendant upon an RIA. If the cladding fails during an RIA, it is possible that fuel dispersal could cause flow blockage and an unacceptable rise in fuel temperature. This raises concerns about the extension of fuel burnup levels beyond current limits.

In order to properly evaluate the possible consequences of this accident on reactor cladding, it is necessary to understand the behavior of Zircaloy under RIA-like loading conditions (plane-strain loading, high strain rate), on a cladding that has suffered oxidation and hydriding, in addition to radiation damage. We report here a preliminary analysis of a localized necking instability as a possible failure mode during an RIA event. It is well known that sheet metal ductility is usually limited by a localized necking instability. During localized necking, through-thickness slip is concentrated in a narrow band of material such that the adjoining material ceases to deform. Therefore, the overall ductility of the material may be severely limited even though the fracture occurs in a ductile manner. This type of instability can be triggered by small imperfections in thickness, caused for example by non-uniform oxidation or by the failure of a thin rim of embrittled material at small strains. High burn-up thin-wall cladding and exhibiting low work-hardening should be especially sensitive to this form of instability, especially if forced to deform under plane-strain conditions. Hence, this model is very relevant to loading and deformation of high burnup fuel cladding due to a RIA event.

2. Failure of High-burnup Zircaloy Cladding under RIA conditions.

One of the consequences of a reactivity-initiated accident is a large instantaneous energy deposition in the fuel. The resulting increase in fuel and cladding temperature can lead to fuel rod failure and, in more severe cases, to fuel dispersion and loss of coolable geometry. Based on simulation experiments conducted in the 1970's in the US [3], regulatory guidelines were established that set energy deposition limits to avoid fuel rod failure and loss of coolable configuration (due to fuel dispersion). The thresholds for failure and for fuel dispersion were established at 170 and 280 cal/g UO_2, respectively.

Recent results appear to indicate that the fuel failure threshold decreases significantly as the burnup increases [2]. In experiments performed in the CABRI facility in France [4], 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 to the above, for the HBO experiments conducted in the NSSR facility in Japan, presented a different failure mode [5]. Occurring at lower temperatures (~50° C) and much lower overall hydrogen content (~190 ppm average), failure in the HBO-1 test is in some ways of greater safety concern. First, cladding failure occurred despite some plasticity of the cladding and despite cladding wall thinning. Second, failure of the cladding has a much different fracture profile than the CABRI experiment. Post irradiation examination showed the cladding has many surface cracks (< 0.1 mm long) which arrested within the ductile cladding. Unlike the CABRI failure, most of the fracture surface involves shear on planes often inclined ± 45° through the thickness. Thus, most of the HBO-1 cladding has good crack growth resistance with failure apparently dominated by plane stress and through-thickness slip. Hydrides are evident only near the outer surface (within ~50μm) of the cladding.

It is significant that a second test performed at the NSSR facility, (HBO-3), was performed under almost identical conditions but did not fail. The Zircaloy cladding in the HBO-3 test had a slightly thinner oxide thickness than HBO-1 (20 to 25 μm compared to 38 to 48 μm) and a lower average hydrogen content (187 ppm vs. 148 ppm). Importantly, the outer rim of cladding which contained excessive hydrides was significantly thicker in HBO-1 than in HBO-3 (approximately 50 μm as compared to 25 μm in HBO-3). Under test conditions, HBO-1 failed with less than 1% hoop strain, while the cladding in the HBO-3 test exhibited approximately 1.5% residual hoop strain without failing. Thus small differences in material conditions resulted in large differences in failure responses between these two tests.

In the present paper, we draw attention to a consequence of sheet metal plasticity in which a localized necking instability can occur under certain conditions. Once triggered, such an instability can limit ductility and cause failure at very small strains, even though the failure process itself is ductile fracture characterized by through-thickness shear on a plane inclined roughly 45 degrees through the cladding thickness. As the analysis below indicates, the onset of localized necking is very sensitive to the deformation path imposed on the cladding during an RIA event, the deformation behavior of the cladding (specifically its strain-hardening characteristics), and the presence of any thickness imperfections.

Given the small thickness of Zircaloy cladding (~610 μm), a relatively thin strip (~50 μm) of hydrided material which cracks (or fails at small strains) can create an imperfection which can trigger a localized necking phenomenon, resulting in cladding failure at small macroscopic strains. We believe that the presence of a thin rim of hydrided cladding which fails at very small strains can in effect create the type of thickness imperfection which can trigger a localized necking instability which severely curtails any hoop extension of the cladding. With regard to the differing behavior of the HBO-1 and HB0-3 tests, the analysis suggests that the thickness of an embrittled, hydrided rim is a critical factor which determines whether or not cladding will fail in a RIA event.
We now show how such a mechanism can limit the ductility of Zircaloy cladding.

3. Influence of an Embrittled Rim on Zircaloy Ductility

The failure of Zircaloy cladding during pellet-cladding mechanical interaction resulting from an RIA, is usually attributed to either crack growth leading to brittle failure, or to the global accumulation of damage throughout the cladding thickness, leading to a general yielding type of failure. However, it is well known that the ductility of sheet metal is frequently limited by a plastic instability known as localized necking [6-9]. During localized necking, through-thickness slip deformation is concentrated 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 if no other failure mechanism 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.

In this work, we present analysis which suggests that a thin layer of embrittled material, which fails at small strain, creates a thickness imperfection that seriously degrades the ductility of otherwise undamaged cladding. We assume that the cladding contains a layer of metal which has been embrittled due to a combination of radiation damage, hydriding and oxidation. Once this material has failed there are two regions, A and B, with thicknesses t_A^o and t_B^o, as shown in figure 1. If localized necking controls the cladding failure strain, $\bar{\varepsilon}_f$, then

$$\bar{\varepsilon}_f = \bar{\varepsilon}_e + \Delta\bar{\varepsilon}_\gamma \qquad (1)$$

where $\Delta\bar{\varepsilon}_\gamma$ is the equivalent strain increment to initiate localized necking once the embrittled layer fails at $\bar{\varepsilon}_e$. To estimate $\Delta\bar{\varepsilon}_\gamma$ we assume the material obeys the power law hardening relationship.

$$\bar{\sigma} = K\,\bar{\varepsilon}^n\,\dot{\bar{\varepsilon}}^m \qquad (2)$$

where $\bar{\varepsilon}$ is the equivalent strain, $\dot{\bar{\varepsilon}}$ is the equivalent strain rate, n is the strain hardening exponent, m the strain rate hardening exponent, and K the strength coefficient. Furthermore, we assume that the cladding deforms in plane-strain tension such that its axial extension is zero (i.e. $\varepsilon_{ZZ} = 0$ in figure 1). 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 [7] expected to be present in this case.

From equilibrium we can write

$$\sigma_A\, t_A = \sigma_B\, t_B \qquad (3)$$

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 [8]

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

where ε_1^A and ε_1^B are the principal strains in the A and B regions. 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 non-uniformity of the brittle section of the wall. Note that the limit strain will not be dependent on the strength coefficient K. Once localized necking occurs, the ductility of the cladding is that of the "limit" strain, which is the far-field strain at the onset of necking.

Applying the analysis above to Zircaloy, using n = 0.01 and m = 0.02 typical of irradiated Zircaloy [10] we found that even a very small imperfection (f = 0.03) can trigger a necking instability, which reduces the limit strain to 0.01. 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%, even though the material would normally exhibit higher strains at failure. This is shown in Figure 2 where the strain in the necked region is plotted against the strain in the far-field, for various values of the flaw size f, assuming the strain hardening exponent n= 0.02 and strain rate hardening exponent m=0.02, typical of irradiated Zircaloy cladding. The limit strains are indicated by arrows in figure 2. The limit strain for f=0.03 in this case is only 0.017. It should be mentioned that not considered in the present analysis is any flow softening due to dislocation channeling induced by irradiation damage, nor any effect of the high strain rates present during an RIA. Both of these effects should accelerate the localized necking development.

Finally, we note that the above analysis assumed plane-strain deformation of the cladding in which there was no axial extension of the cladding. Previous analyses have shown that the onset of localized necking is quite sensitive to deformation path [6]. Specifically, localized necking is inhibited by deformation paths other than plane-strain tension. For example, deformation in uniaxial tension delays localized necking to strains approximately twice those in plane tension. Thus for cladding whose ductility is limited by the localized necking phenomenon, tests of cladding in uniaxial tension will result in erroneously high indications of cladding ductility if it is subjected to plane-strain tension in service . As a result, we strongly believe that the use of ductility data generated from axial tension tests of cladding can be very misleading in predicting the performance of cladding during a RIA event in which the cladding is likely to be subjected to plane-strain tension. We also note that there may be a significant difference in material behavior between failure due to hoop extension and that after axial extension; for a RIA event, hoop extension is the operative failure path.

4. Discussion

The determination of the mode of failure prevalent in high burnup cladding during an RIA is essential to understanding the phenomenon and developing scientific-based licensing regulations that ensure safety in using high burnup fuel. To predict failures during an RIA, it is necessary to consider all possible modes of failure, including those made possible by the damaged state of the cladding after long reactor exposure. Then more realistic fuel failure limits can be developed.

The mode of failure proposed in this paper had been observed previously by Pettersson et al. [11] who conducted burst tests in Zircaloy cladding irradiated to up to 10.8×10^{24} $n.m^{-2}$ ($E > 1$ MeV). The tests revealed that small surface flaws considerably limited the ductility of Zircaloy cladding when loaded under plane strain conditions. This sensitivity to defects was absent in similar tests the same authors conducted in ring specimens, where the loading is uniaxial [12]. The authors concluded that the combination of plane strain loading and the small amount of strain hardening in the irradiated materials led to the high degree of defect sensitivity observed.

In addition, Lee and Adamson [13] have shown that the load-carrying capacity of irradiated Zircaloy cladding is severely degraded under plane-strain loading conditions when compared with axial tension . They also emphasize that the continuum modeling of the deformation behavior of irradiated Zircaloy cladding may be inadequate because of the localized nature of the deformation. As mentioned above, localized deformation induced by dislocation channeling would further reduce the ductility of the irradiated material.

Our analysis and the experimental evidence above indicated a sensitivity of cladding ductility to a combination of small surface flaws, small work hardening exponent, (associated with reactor exposure) and plane-strain deformation path which can severely limit cladding ductility. The flaw size calculated in section 3 can readily appear in irradiated cladding especially after high burnup. One possibility is uneven cladding oxidation. After 45 GWd/ton, oxide thicknesses of 70-100 μm can exist in some types of cladding [14]. In that case a 20% fluctuation in the oxide thickness could trigger the above mentioned instability. Another possibility is that a hydride rim or blister forms, possibly aided by oxide spalling. The fracture resistance of this rim would be highly dependent on the local hydride distribution, but a hydrided rim of about 50 μm seen in the HBO-1 experiment [5] may well be sufficient to cause cladding failure by localized necking. In contrast, the 25 μm hydride rim in HBO-3 may be an insufficient imperfection to cause failure prior to the 1.5% hoop extension during an RIA.

5. Summary

The failure of Zircaloy cladding during an RIA event has been examined from the standpoint of the occurrence of a localized necking instability limiting cladding ductility. Ignoring the influence of high strain rates (which are also likely to reduce ductility), our analysis suggests that high burn-up cladding may be susceptible to pronounced losses of ductility under a combination of plane-strain deformation and the presence of thickness "imperfections", such as may be caused by a rim of hydrided material or non-uniform oxidation of the cladding. Applied to Zircaloy cladding with deformation behavior expected of high burn-up fuel, the analysis suggests that a 18 μm flaw, possibly originating from cracking within a thin rim of heavily hydrided material, could cause failure of the cladding at hoop strain levels of less than 1% strain after flaw initiation. The features and general characteristics of this failure mode appear to correspond well the cladding failure observed in the NSSR tests, but not with the CABRI REP Na-1 failure.

Acknowledgments

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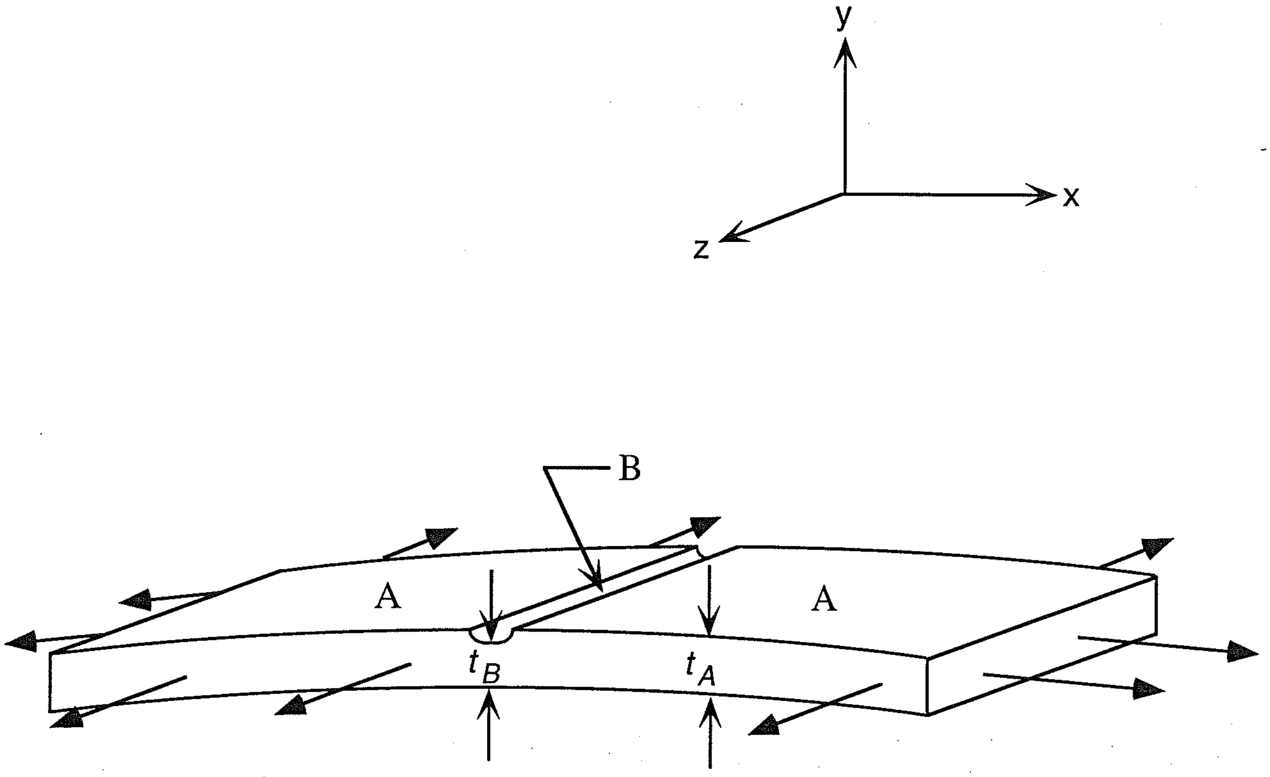

Figure 1: A schematic drawing indicating an imperfection reducing the thickness of the sheet from t_A to t_B. Plane-strain conditions are indicated during sheet extension.

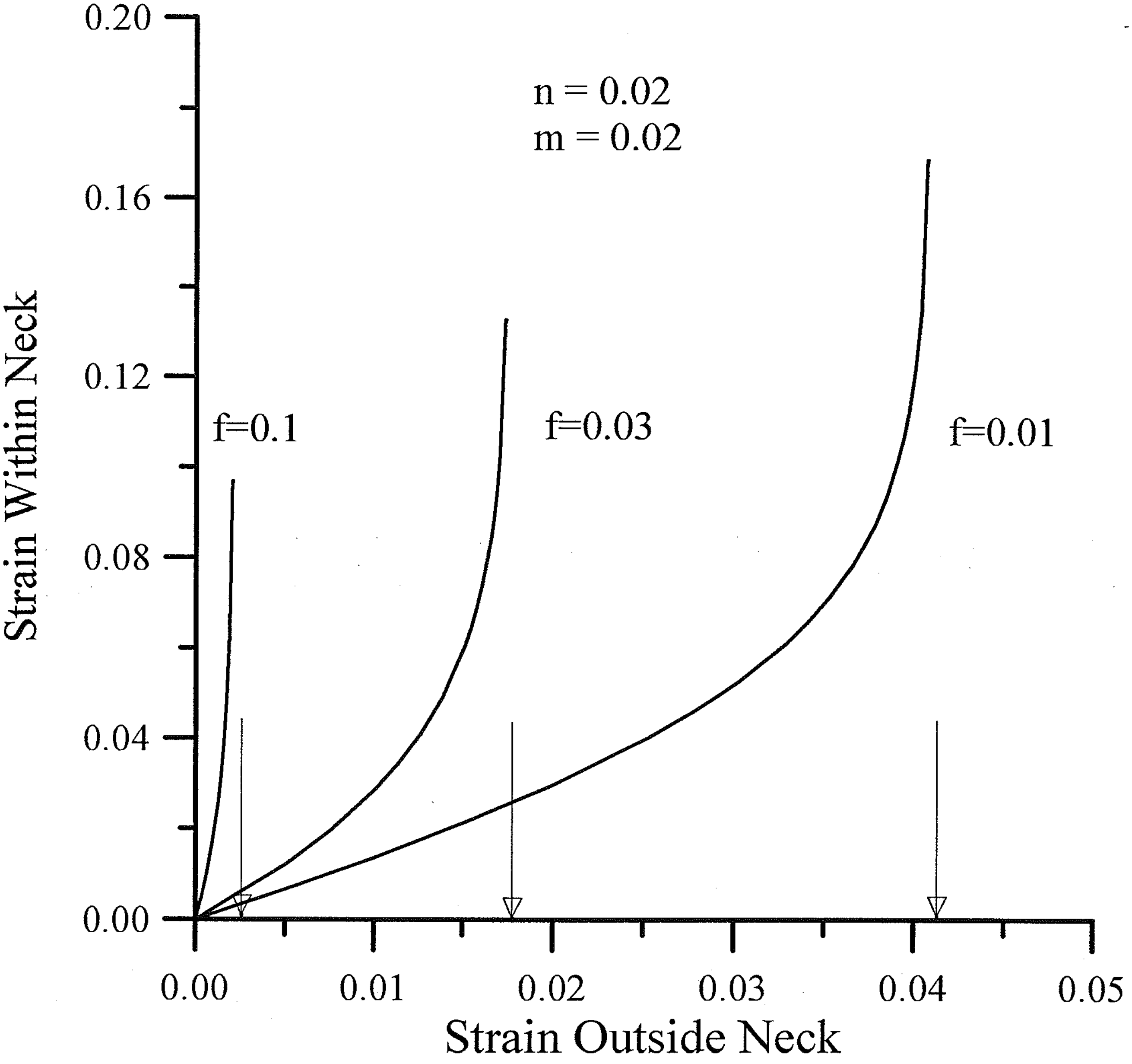

Figure 2. The development of a localized necking instability for the plane-strain deformation of cladding containing imperfections of differing severities *f*. Strains are maximum principal strain values and arrows denote the limit strain for each case.