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A creep rupture criterion for Zircaloy-4 fuel cladding under internal pressure

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Abstract

The usual criterion which limits the cladding strain to 0.01 to prevent the creep rupture under internal pressure seems too conservative for application to transport and interim storage. So we have analysed CEA's data on this subject for CWSR Zircaloy-4 in order to find a less conservative criterion. Temperatures between 350 and 470 °C were studied for stresses between 100 and 550 MPa, according to the irradiation level from 0 to 9.5×10^{25} n m^{-2}. Except for high stressed irradiated material (because of low ductility), the plastic instability appears as the major mechanism of rupture. For the unirradiated material, it is essentially due to the stress increase with strain. This instability is accelerated by annealing for the irradiated one at moderate or low stress. From these considerations, we propose a new rupture criterion for CWSR Zircaloy-4 cladding submitted to internal pressure, for both unirradiated and irradiated materials.

1. Introduction

In the framework of the management of nuclear wastes, research and development programs dealing with the long term behaviour of spent nuclear fuel in conditions of interim storage or final disposal have been undertaken. One major issue concerns the long term behaviour (50, 100, even 300 years) of the irradiated cladding, and its integrity, in dry storage conditions.

After irradiation in a Pressurized Water Reactor, the fuel rod contains a relative high amount of helium, introduced during fabrication or produced by α decay, and fission gases, produced and released during irradiation. Since the fuel rod is expected to undergo a temperature up to about 400 °C at the beginning of storage, the cladding is submitted to a relative high internal pressure due to these gases. The estimated circumferential stress (the maximal principal stress) ranges from about 70 to 120 MPa. Although temperature and stress will decrease during storage, long term creep is considered as the relevant deformation mechanism which could potentially lead to a cladding rupture [1–3].

Furthermore, during the few days transport from the power plant to the storage site, the temperature and the stress of the cladding might reach about 470 °C and 130 MPa in the worst cases. Such conditions may lead to partial annealing of irradiation defects and notable creep strain.

In order to predict the creep behaviour of the fuel cladding during storage, we have undertaken creep

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experiments on irradiated cladding samples and modelling work. The objective is to establish an adequate creep law and a rupture criterion through a stepwise approach from short term experiments at high stress (few days or less) to long term ones at low stress (few years). The experiments at high stress were previously carried out to study the irradiation transient Pellet-Cladding Mechanical Interaction (PCMI) [4]. The first step of the study of storage has consisted of medium term (few tens days) creep experiments [5,6] and the formulation of a first deformation law [5].

The criterion which is usually used to prevent the creep rupture of fuel cladding consists to limit the circumferential strain to 0.01. This value seems based on the minimum uniform elongation of fuel rods with a burnup less than 40000 MWd/tU submitted to burst tests [2]. Furthermore, no rupture of such fuel rods will occur if strain is limited to 0.01 during storage [3]. According to its first justification, the strain limit should decrease to about 0.005 with increasing burnup [2], at least for creep at high stress level. For long term creep during storage or creep at potential high temperature during transport, the cladding ductility could be higher because of the irradiation defects annealing. Effectively, rupture strains much higher than 0.01 were observed [5,6].

So, the purpose of this work is to analyse the creep rupture results for both unirradiated and irradiated cladding in order to determine the mechanism of rupture and to propose a new criterion.

2. Experiments

2.1. Material

The studied material is Cold Worked Stress Relieved (CWSR) Zircaloy-4 cladding. Before irradiation, its external diameter and thickness are 9.5 and 0.57 mm.

The unirradiated material and the irradiated one used for the PCMI study [4] was an optimized low tin (1.25–1.3%) Zircaloy-4. Creep tests on irradiated material were performed on two types of samples:

- cladding samples previously irradiated in NaK in the reactor OSIRIS, without fuel and stress, up to a fast neutron fluence (energy >1 MeV) of $2.2\times 10^{25}\ \mathrm{n\,m^{-2}}$,
- samples taken on claddings of fuel rods previously irradiated 2 or four annual cycles in a PWR.

For the first step of the study of interim storage [5,6], the material was a standard Zircaloy-4 with a chemical composition conforming to the ASTM specification. Creep samples were taken on claddings of fuel rods previously irradiated four cycles in a PWR.

2.2. Creep test procedure and interpretation

The cladding sample, previously closed at one end, is submitted to an internal pressure at constant temperature. Pressure is applied by the mean of oil [4] or argon [5,6]. In the first case, a regulation ensures a constant pressure. In the second one, the internal pressure decreases with increasing strain but in attenuated manner due to a buffer volume. So, according to the measurements, pressure can be considered as constant in most of the cases.

In the medium part of the test sample, where the diameter variations are measured, the mean values of the radial, circumferential and axial stresses: σ_{rr}, $\sigma_{\theta\theta}$ and σ_{zz}, verify the following relations (second order approximation with respect to e/D_m):

$$\sigma_{\theta\theta} - \sigma_{rr} = 2\cdot(\sigma_{zz} - \sigma_{rr}), \tag{1}$$

$$\sigma_{\theta\theta} - \sigma_{rr} \approx (p_\mathrm{i} - p_\mathrm{e})\frac{D_\mathrm{m}}{2e}, \tag{2}$$

where p_i and p_e are the internal and external pressures, D_m and e are the mean diameter and the thickness of the tube. To interpret the creep experiments, one must only consider the stress difference $\sigma_{\theta\theta} - \sigma_{rr}$ [5]. Its conventional (calculated with the initial geometry) and true values are respectively called by $\sigma'_{\theta\mathrm{c}}$ and σ'_θ.

The mean conventional and true circumferential strains in the tube thickness are respectively:

$$\varepsilon_{\theta\mathrm{c}} = \frac{\Delta D_\mathrm{m}}{D_\mathrm{m0}} \approx \frac{\Delta D_\mathrm{e}}{D_\mathrm{m0}}, \tag{3}$$

$$\varepsilon_{\theta\theta} = \ln(1+\varepsilon_{\theta\mathrm{c}}), \tag{4}$$

where D_m0 and ΔD_m are the initial value and the variation of the tube mean diameter. Because ΔD_m is not directly measurable it is approximated by the associated variation of the external diameter of the tube ΔD_e.

The creep experiments results are represented by the rupture time and the rupture conventional strain as function of the conventional stress $\sigma'_{\theta\mathrm{c}}$.

2.3. Results on unirradiated material

Creep tests up to rupture were realized in the temperature range from 350 to 400 °C, for stresses between 240 and 530 MPa. The rupture time varies between 100 s and 46 days (Table 1). At a given temperature, it concurs approximately with a decreasing exponential function of stress. The slope of straight line on semi-logarithmic graph appears independent of temperature (Fig. 1). The rupture strain decreases from about 0.4 to 0.08 as a function of stress (Fig. 2). In the studied range, it appears independent of temperature and

Table 1
Results of creep rupture under internal pressure for unirradiated CWSR Zircaloy-4

Temperature (°C)	Stress (MPa)	Rupture time (s)	Rupture strain
350	266	3.99E+06	0.307
	319	6.34E+05	0.180
	337	3.35E+05	0.210
	362	1.40E+05	0.148
	373	1.06E+05	0.206
	373	1.22E+05	0.173
	391	5.11E+04	0.130
	392	3.67E+04	0.103
	394	3.67E+04	
	414	3.31E+04	0.207
	414	3.62E+04	0.173
	414	3.28E+04	0.207
	426	1.72E+04	0.138
	445	6.12E+03	0.112
	445	4.32E+03	0.077
	447	4.32E+03	0.095
	477	2.77E+03	0.106
	479	1.40E+03	0.076
	479	1.44E+03	0.094
	479	1.51E+03	0.076
	480	1.30E+03	0.058
	480	1.08E+03	0.076
	529	1.08E+02	0.106
380	239	1.07E+06	0.396
	309	7.27E+04	0.237
	319	6.19E+04	0.254
	360	1.30E+04	0.174
	399	4.68E+03	0.174
	420	1.44E+03	0.130
	421	1.44E+03	0.130
400	250	1.62E+05	0.350
	301	2.84E+04	0.237
	350	4.68E+03	0.166
	400	7.20E+02	0.139
	401	7.20E+02	0.130

correlatively the rupture time is very sensitive to temperature.

2.4. Results on irradiated material

Experiments on high stressed samples (PCMI) were realized for fast neutron fluences between 4×10^{24} and 9.5×10^{25} n m^{-2}. Test temperature was 350 or 380 °C. Stress ranged from 350 to 550 MPa in the first case and from 300 to 420 MPa in the second one. In these conditions, the rupture time varies between 2 h and 3 days (Table 2). It increases until two cycles in PWR (4–5 $\times 10^{25}$ n m^{-2}) and remains almost constant after at a level from 10 to 1000 times the rupture time of the unirradiated material according to temperature and stress (Fig. 3). The rupture strain decreases regularly with increasing irradiation (Fig. 4). It is about 0.01 at four cycles for high stress.

Medium term creep experiments on cladding previously irradiated four cycles in a PWR have been realized within the temperature range from 380 to 470 °C, at stresses between 100 and 240 MPa, for durations between 10 and 150 days (Table 2). According to their conditions the tests have led or not to rupture. Every rupture strain is higher than 0.05 and the three not ruptured samples have reached clearly higher strains than 0.01 (Fig. 5). It shows the existence of significant annealing of irradiation damage. Nevertheless, the time reached by a sample which is not yet ruptured, at (380 °C, 240 MPa), tends to show that the beneficial effect of irradiation hardening on rupture time remains in spite of this annealing (Fig. 3).

2.5. Relation between stationary creep strain rate and rupture time

Fig. 6 shows the rupture time t_R as a function of the minimal creep strain rate v_s for all temperature and

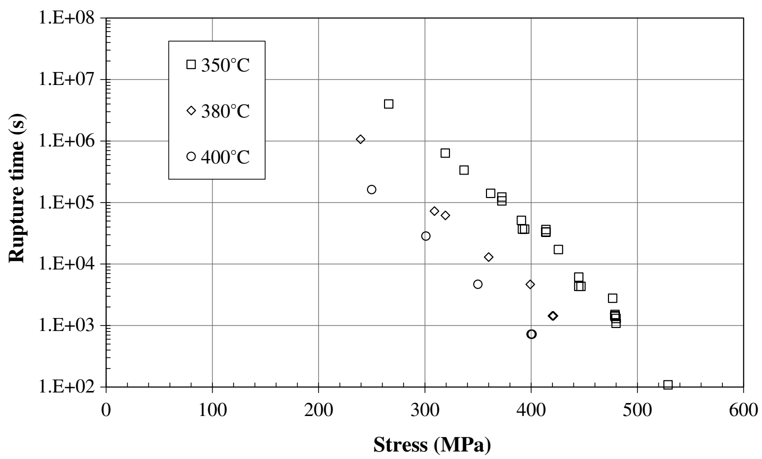

Fig. 1. Unirradiated CWSR Zircaloy-4 rupture time as a function of stress and temperature.

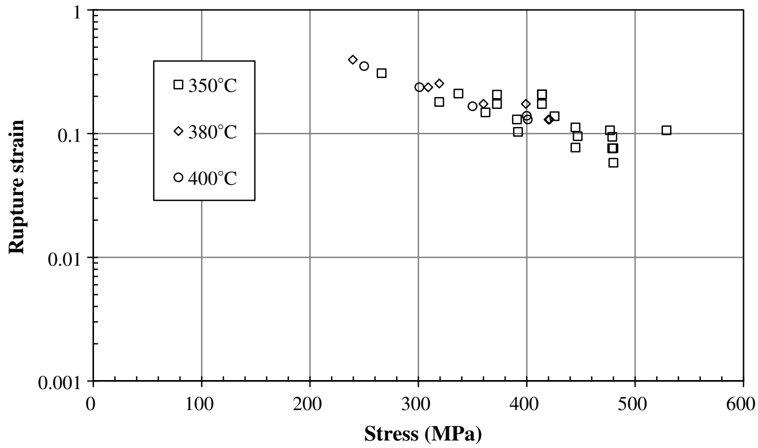

Fig. 2. Unirradiated CWSR Zircaloy-4 rupture strain as a function of stress and temperature.

Table 2
Results of creep rupture under internal pressure for irradiated CWSR Zircaloy-4

Fluence ($n\,m^{-2}$)	Temperature (°C)	Stress (MPa)	Rupture time (s)	Rupture strain
4.8E+24	350	360	2.02E+05	0.063
5.4E+24		386	1.19E+05	0.074
4.4E+24		445	2.21E+04	0.079
6.8E+24		397	1.55E+05	0.108
8.3E+24		445	3.96E+04	0.068
7.9E+24		517	7.20E+03	0.080
5.8E+24	380	309	9.72E+04	0.071
6.4E+24		333	5.76E+04	0.052
6.6E+24		358	3.06E+04	0.029
2.2E+25	350	396	2.34E+05	0.017
2.1E+25		442	1.01E+05	0.028
2.4E+25		516	3.60E+04	0.038
4.6E+25	350	545	7.56E+04	0.025
4.5E+25	380	360	1.87E+05	0.022
4.6E+25		385	9.36E+04	0.023
4.3E+25		418	5.04E+04	0.017
9.3E+25	350	494	1.87E+05	0.009
9.4E+25		539	6.12E+04	0.010
9.5E+25	380	381	1.48E+05	0.012
9.5E+25		413	7.56E+04	0.009
≈8.8E+25	383	239	>8.13E+06	>0.015
≈8.8E+25	399	196	>1.28E+07	>0.031
≈8.8E+25	420	215	3.65E+06	0.097
≈8.8E+25	471	100	>1.53E+06	>0.100
≈8.8E+25	469	119	1.68E+06	0.273
≈8.8E+25	472	120	9.31E+05	0.053

stress conditions, for both unirradiated and irradiated materials. The whole data lie within a factor of 5 from the law of Monkman and Grant [7]:

$$v_s^q t_R = C_{MG} \quad (5)$$

fitted to the results on the unirradiated material with the constant q equal to 1 (C_{MG} = 0.056).

3. Analysis of the rupture mechanism

3.1. Unirradiated material

Rupture of the unirradiated material is in every case preceded by a tertiary creep phase. The strain acceleration occurs when the effect of the stress increase, due

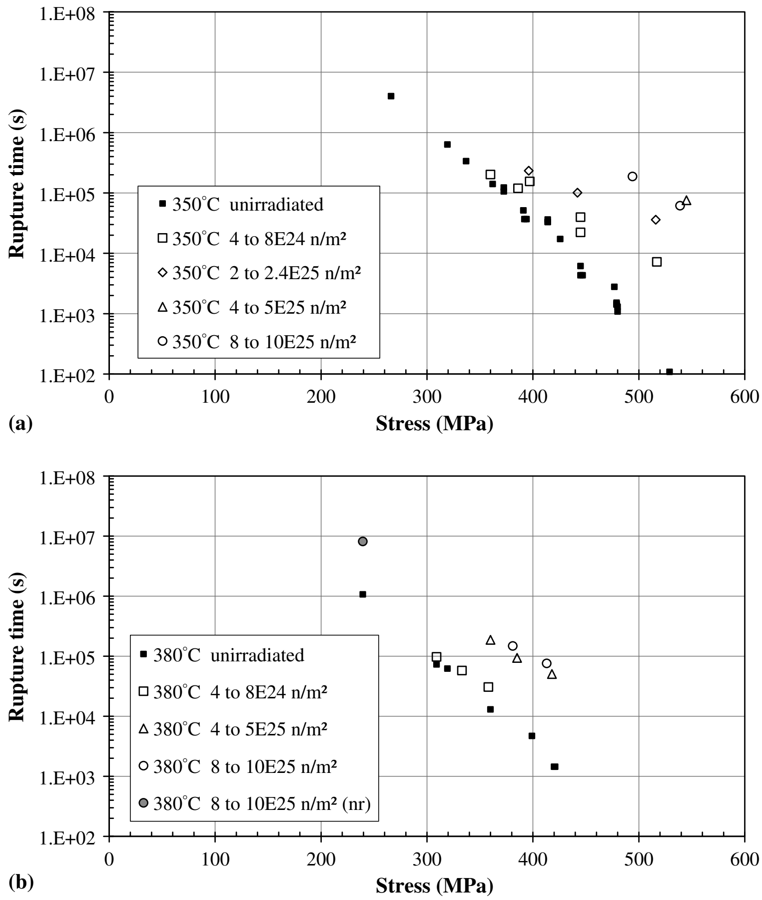

Fig. 3. (a) Irradiated CWSR Zircaloy-4 rupture time as a function of stress and fast neutron fluence at 350 °C. (b) Irradiated CWSR Zircaloy-4 rupture time as a function of stress and fast neutron fluence at 380 °C.

to the diameter increase and the thickness decrease during creep at constant pressure, exceeds that of apparent strain hardening, which may result from the true strain hardening, annealing and damage.

In order to know if the two last terms exist, we have calculated the sample deformation with the creep law of A. Soniak et al. (AS) [4], taking into account the stress increase with strain. It was done by a numerical method, with a code called MISTRAL (MaterIal STRAin Laws), on the assumption that the cladding radial, circumferential and axial directions are the principal directions of stresses and strains (as for an infinite tube). Results are presented on Fig. 7 for three experimental conditions.

For strain below 0.01, there is a good agreement between calculation and experiment. It was expected because the law of AS was founded on these results, within this strain range. The tertiary creep is over-estimated by calculation. It shows an under-estimation of strain hardening in this phase. Besides, strain hardening is almost zero for strain beyond 0.01 according to the law of AS. The over-estimation of the tertiary creep would be worst with annealing or damage.

Calculation predicts plastic instability: strain tends to infinite at one given time. It appears at a strain of the order of the rupture strain. So we have estimated the plastic instability time as a function of temperature and stress by such calculations. Calculated instability times are in rather good agreement with the measured rupture ones and even a little smaller in most of the cases (Fig. 8).

All these observations demonstrate there is practically neither annealing nor damage during tertiary creep phase, except probably just before rupture. Correlatively, plastic instability due to the stress increase with strain appears as the major mechanism of rupture.

These conclusions are in agreement with those of Mayuzumi and Onchy [1] for creep experiments on pre-pressurized specimens and the resistance of Zircaloy to cavity formation according to Zhou et al. [8].

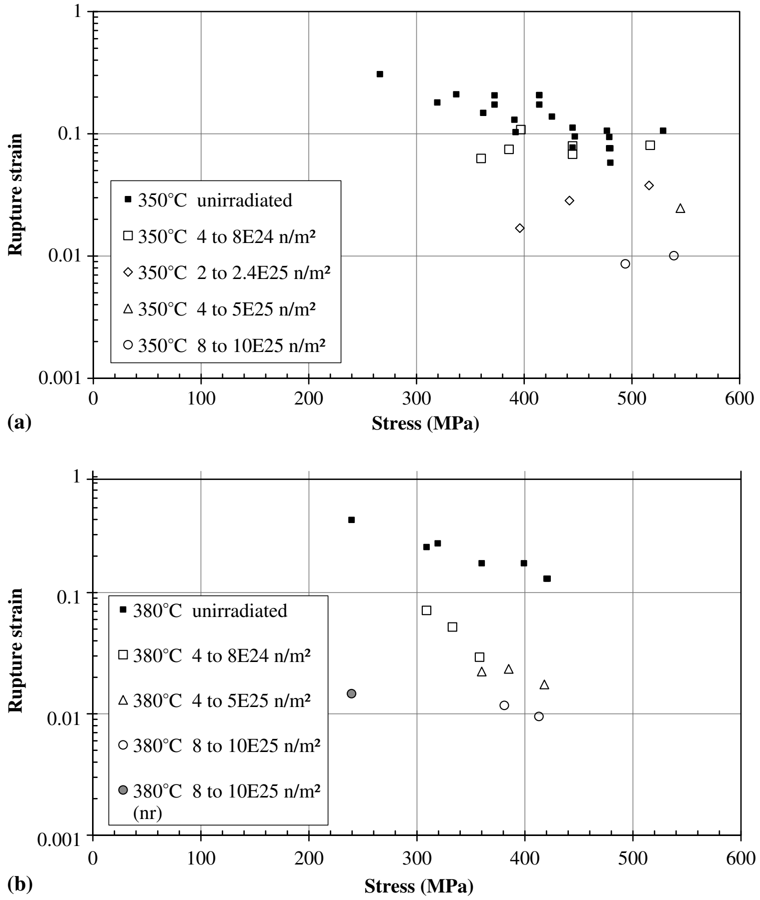

Fig. 4. (a) Irradiated CWSR Zircaloy-4 rupture strain as a function of stress and fast neutron fluence at 350 °C. (b) Irradiated CWSR Zircaloy-4 rupture strain as a function of stress and fast neutron fluence at 380 °C.

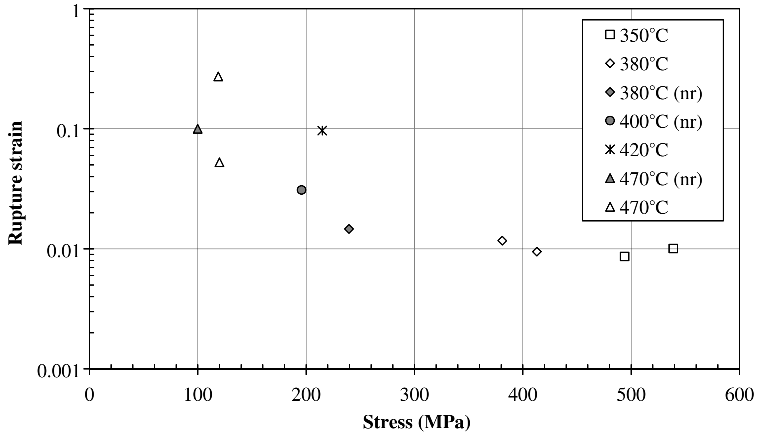

Fig. 5. Highly irradiated CWSR Zircaloy-4 rupture strain as a function of stress and temperature (nr): not ruptured.

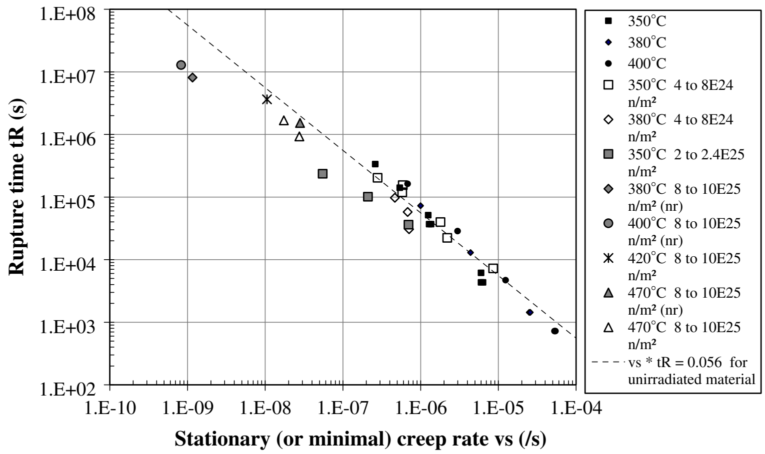

Fig. 6. Unirradiated and irradiated CWSR Zircaloy-4 rupture time as a function of the minimal creep strain rate.

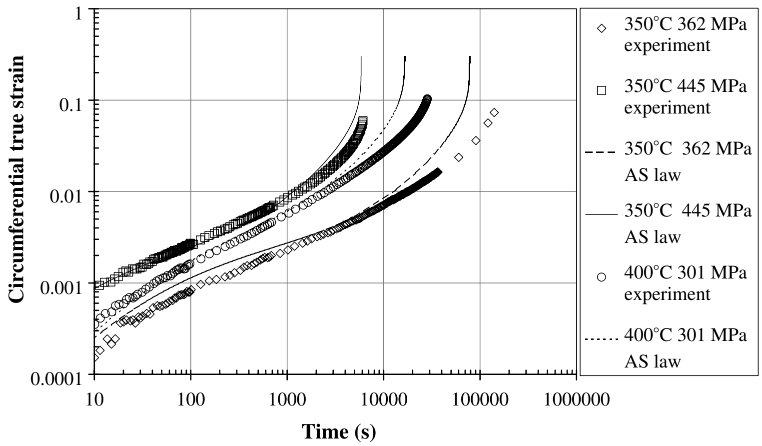

Fig. 7. Unirradiated CWSR Zircaloy-4: comparison of measured and calculated strains.

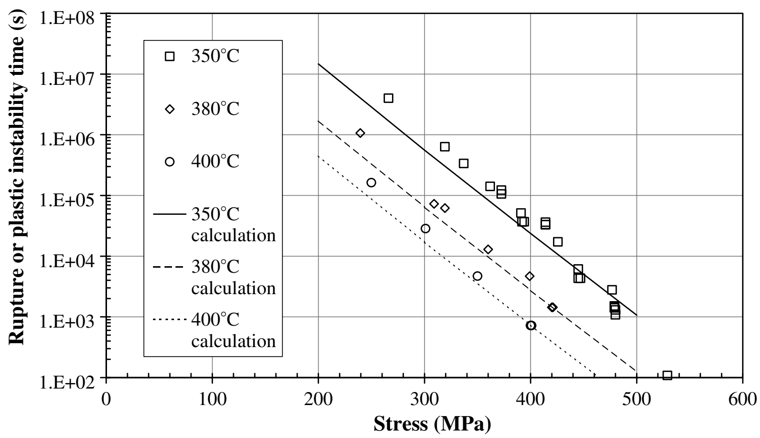

Fig. 8. Unirradiated CWSR Zircaloy-4: comparison of calculated plastic instability time to experimental rupture time.

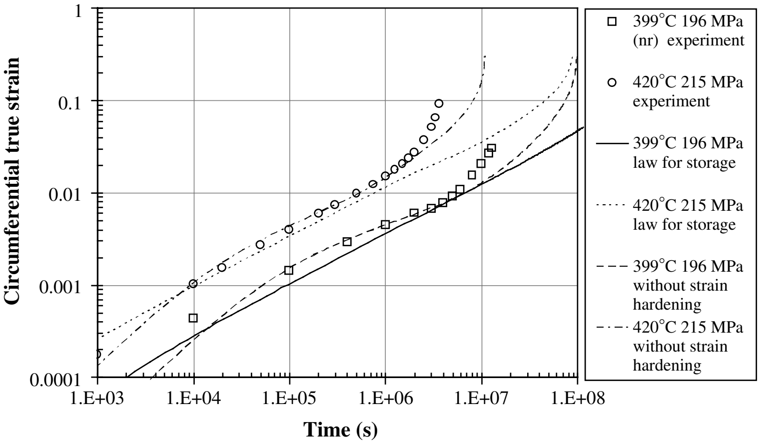

Fig. 9. Highly irradiated CWSR Zircaloy-4: comparison of measured and calculated strains.

3.2. Irradiated material submitted to high stress (PCMI)

Until at least two cycles in PWR (about 4.5×10^{25} $\mathrm{n\,m^{-2}}$) the irradiated material is ductile: rupture occurs after a tertiary creep phase or, in some cases at 350 °C, a long secondary creep phase. The stress increase due to strain is still an important mechanism for rupture, but this one occurs clearly before plastic instability except at very low fluence.

At four cycles ($8\text{–}10\times10^{25}$ $\mathrm{n\,m^{-2}}$) brittle aspects of rupture appear: rupture strain of about 0.01, no tertiary creep phase at 350 °C. Nevertheless, early tertiary creep appears at 380 °C; it is probably due to the appearance of annealing. The stress increase is not important for rupture because only small strains are reached.

3.3. Irradiated material submitted to moderate stress (transport and storage)

Due to annealing, the irradiated material is ductile at moderate stress level and it shows an important tertiary creep phase before rupture, according to the results of the five last creep experiments mentioned in Table 2.

For the two experiments in the range of validity of the creep law for storage [5], we have calculated the sample deformation, with this law, taking into account the true stress variation with strain as for the unirradiated material. The measured strain is largely under-estimated by calculation during tertiary creep and the calculated plastic instability time is more than 20 times the actual rupture time (Fig. 9).

These results demonstrate the over-estimation of strain hardening for strain beyond about 0.01 by the law, but not necessarily the existence of annealing. To assess this existence, we fitted a law to each experiment, for strains below 0.01, keeping the stress influence of the creep law for storage, but with the same strain hardening form that the law of AS [4] in order to have practically no strain hardening for strains beyond 0.01. For each creep experiment, calculation with its own law so determined leads still to clear under-estimation of tertiary creep strain. It confirms the existence of enough annealing to induce an apparent negative strain hardening.

Nevertheless, the actual rupture strain is of the order of the predicted instability strain. It tends to demonstrate that plastic instability is again the major mechanism of rupture but, unlike the unirradiated material, it is accelerated by irradiation defects annealing.

4. Analytic study of deformation due to internal pressurization

4.1. Studied case, hypothesis

The total strain is considered as the sum of an elastic strain and a plastic one (we call plastic every irreversible strain due to stress whatever it is (almost) instantaneous with threshold or viscoplastic). So $\varepsilon_\theta=\varepsilon_\theta^{\mathrm{el}}+\varepsilon_\theta^{\mathrm{pl}}$ for the mean circumferential strain. The plastic strain rate v_θ^{pl} is assumed to be a function of the true stress difference $\sigma'_\theta=\sigma_{\theta\theta}-\sigma_{rr}$ and the plastic true strain:

$$v_\theta^{\mathrm{pl}}=\frac{\mathrm{d}\varepsilon_\theta^{\mathrm{pl}}}{\mathrm{d}t}=V(\sigma'_\theta,\varepsilon_\theta^{\mathrm{pl}}). \tag{6}$$

It is a viscoplastic law with strain hardening and without explicit annealing.

4.2. Relation between the derivative of pressure and that of strain rate

From the expression (2) of stress we obtain the following relation for its differential:

$$\frac{\mathrm{d}\sigma'_\theta}{\sigma'_\theta} = \frac{\mathrm{d}\Delta P}{\Delta P} + \frac{\mathrm{d}D_\mathrm{m}}{D_\mathrm{m}} - \frac{\mathrm{d}e}{e} = \frac{\mathrm{d}\Delta P}{\Delta P} + (1-Q)\,\mathrm{d}\varepsilon_\theta \tag{7}$$

with $\Delta p = p_\mathrm{i} - p_\mathrm{e}$ and $Q = \frac{\mathrm{d}\varepsilon_r}{\mathrm{d}\varepsilon_\theta}$. For plastic strains, this ratio is equal to $(-\frac{\mathrm{d}\varepsilon_z}{\mathrm{d}\varepsilon_\theta} - 1)$. Q is therefore equal to -1 for isotropic material or almost for CWSR Zircaloy-4 in every condition [9].

The law of deformation and this relation lead together to the following expression of the strain rate derivative with respect to time t:

$$\frac{\mathrm{d}v_\theta^\mathrm{pl}}{\mathrm{d}t} = m \cdot \left(\frac{1}{\Delta P}\frac{\mathrm{d}\Delta P}{\mathrm{d}t} + (1-Q)v_\theta\right)v_\theta^\mathrm{pl} - \frac{p}{\varepsilon_\theta^\mathrm{pl}}v_\theta^{\mathrm{pl}^2} \tag{8}$$

with $m = \frac{\sigma'_\theta}{v_\theta^\mathrm{pl}}\frac{\partial v_\theta^\mathrm{pl}}{\partial \sigma'_\theta}$ and $p = -\frac{\varepsilon_\theta^\mathrm{pl}}{v_\theta^\mathrm{pl}}\frac{\partial v_\theta^\mathrm{pl}}{\partial \varepsilon_\theta^\mathrm{pl}}$ which represent respectively the relative sensitivities of strain rate to stress and strain.

In the precedent expression, the elastic strain rate is negligible beside the plastic one, therefore:

$$\frac{\mathrm{d}v_\theta^\mathrm{pl}}{\mathrm{d}t} \approx m\frac{1}{\Delta P}\frac{\mathrm{d}\Delta P}{\mathrm{d}t}v_\theta^\mathrm{pl} + \left(m\cdot(1-Q) - \frac{p}{\varepsilon_\theta^\mathrm{pl}}\right)v_\theta^{\mathrm{pl}^2} \tag{9}$$

with the coefficient Q relative to the plastic strains. If $1-Q$ is replaced with 1, this relation becomes equivalent to that of Hart [10] for axial tensile test.

By using the relation between the conventional and true strains (4) we can deduce the following expression of the derivative of the conventional strain rate $v_{\theta\mathrm{c}}^\mathrm{pl} = \frac{\mathrm{d}\varepsilon_{\theta\mathrm{c}}^\mathrm{pl}}{\mathrm{d}t}$:

$$\frac{\mathrm{d}v_{\theta\mathrm{c}}^\mathrm{pl}}{\mathrm{d}t} \approx \exp(\varepsilon_\theta^\mathrm{pl}) \cdot \left[m\frac{1}{\Delta P}\frac{\mathrm{d}\Delta P}{\mathrm{d}t}v_\theta^\mathrm{pl} + \left(1 + m\cdot(1-Q) - \frac{p}{\varepsilon_\theta^\mathrm{pl}}\right)v_\theta^{\mathrm{pl}^2}\right]. \tag{10}$$

4.3. Stationary creep rate, uniform elongation

For a creep test at constant pressure, relations (9) and (10) show that the true and conventional plastic strain rates are respectively minimal for the following true strains:

$$\varepsilon_{\theta\,\mathrm{min}}^\mathrm{pl} \approx \frac{p}{m\cdot(1-Q)} = \frac{n}{1-Q}, \tag{11}$$

$$\varepsilon_{\theta\,\mathrm{min(c)}}^\mathrm{pl} \approx \frac{p}{1+m\cdot(1-Q)} = \frac{n}{1-Q+\frac{1}{m}} \tag{12}$$

as a function of the strain hardening coefficient: $n = \frac{\varepsilon_\theta^\mathrm{pl}}{\sigma'_\theta}\left(\frac{\partial\sigma'_\theta}{\partial\varepsilon_\theta^\mathrm{pl}}\right)_{v_\theta^\mathrm{pl}}$. Beyond $\varepsilon_{\theta\,\mathrm{min}}^\mathrm{pl}$, the effect of stress increase on the strain rate exceeds that of strain hardening and so creep acceleration appears.

For a test at constant conventional or true strain rate, pressure would be maximal for the true strain $\varepsilon_{\theta\,\mathrm{min(c)}}^\mathrm{pl}$ or $\varepsilon_{\theta\,\mathrm{min}}^\mathrm{pl}$ respectively. So, the deformation at which the creep acceleration begins in a creep test at constant pressure corresponds directly to the uniform elongation as it is called in a burst test at constant strain rate. However, it is only true at the same stress level for both two tests because the coefficient m strongly depends on it. So, if the usual rupture criterion was based on burst tests at constant strain rate of the order of magnitude of $10^{-4}\,\mathrm{s}^{-1}$, it would correspond to the beginning of creep acceleration for creep tests at high constant stress but not for long term creep.

By replacing $1-Q$ with 1 in (11) and (12), we obtain the uniform elongation in axial tensile test for instantaneous plasticity ($m=\infty$ and n finite), according to Considere [11] (mentioned in [12]), and for viscoplasticity [12,13].

4.4. Tertiary creep, plastic instability

According to the relation (9), the more strain rate relative sensitivity to stress, and the less strain hardening, the more creep acceleration. So, the assumption $p=0$ beyond $\varepsilon_{\theta\,\mathrm{min}}^\mathrm{pl}$ will lead to a conservative estimation of the creep acceleration, at least for unirradiated material as seen in Section 3. In this case, at constant pressure this relation becomes:

$$\frac{1}{v_\theta^\mathrm{pl}}\frac{\mathrm{d}v_\theta^\mathrm{pl}}{\mathrm{d}t} \approx m\cdot(1-Q)v_\theta^\mathrm{pl}. \tag{13}$$

To integrate this equation, let Q and m be constant. It is almost true for the first coefficient but not for the second one which generally increases with stress. With the initial value of m, acceleration is under-estimated and, despite the assumption of zero strain hardening, solution will not necessarily be conservative. Nevertheless, it will give an order of magnitude. With the value of m which corresponds to the stress at the highest strain, solution will be conservative. With two integrations of Eq. (13) and transformations by exponential and logarithm functions, we obtain the following expression:

$$\varepsilon_\theta^\mathrm{pl} - \varepsilon_{\theta\,\mathrm{min}}^\mathrm{pl} \approx \frac{1}{m\cdot(1-Q)}\cdot\left[-\ln\left(1 - \frac{t-t_\mathrm{min}}{t_\mathrm{inst}-t_\mathrm{min}}\right)\right] \tag{14}$$

in which t_min is the time at which the creep rate is minimal and t_inst is defined by:

$$t_\mathrm{inst} - t_\mathrm{min} \approx \frac{1}{m\cdot(1-Q)v_{\theta\,\mathrm{min}}^\mathrm{pl}}. \tag{15}$$

The second term of the expression of $\varepsilon_\theta^\mathrm{pl} - \varepsilon_{\theta\,\mathrm{min}}^\mathrm{pl}$ tends to infinite when the time t tends to t_inst although it is only equal to 1 or 3 for fractions $-\frac{t-t_\mathrm{min}}{t_\mathrm{inst}-t_\mathrm{min}}$ respectively equal to 0.63 or 0.95. So plastic instability appears, and will lead to rupture, just before t_inst.

4.5. Order of magnitude of the rupture time

The rupture time t_R of unirradiated CWSR Zircaloy-4 can be estimated by calculating the plastic instability time t_{inst} (paragraph 3). More, according to experiments, the time t_{min} at which the creep rate is minimal ranges from about 1/6 to 1/3 of the rupture time. So, this one verify the following relation within about 50%:

$$v^{pl}_{\theta\,min} t_R \approx \frac{1}{m \cdot (1-Q)} \approx \frac{1}{2m}. \tag{16}$$

This relation looks like the Monkman–Grant's law. With the creep law of AS, it gives $0.034 \leqslant C_{MG} \leqslant 0.086$ for initial stress between 200 and 500 MPa ($5.8 \leqslant m \leqslant 14.5$) although the mean experimental value is $C_{MG} = 0.056$ (Section 2.5).

Such a formula (1 instead of $1-Q$) was established by Hoff [14] (according to [15]) for axial tensile creep test assuming a Norton type secondary creep law. According to [15], for most of metals this relation is not reliable. Nevertheless, the relation (16) may be reliable in the present case for the following reasons:

- damage do not easily develop in Zircaloy-4 according to [8] (Section 3.1),
- the time for this damage development under internal pressure is shorter than it is in tensile creep test because the stress increase rate due to strain is twice as important in the first case as it is in the second one ($1-Q \approx 2$),
- strain hardening is not very important for CWSR Zircaloy-4,
- we used a precise estimation of m as a function of stress.

4.6. Conservative estimation of the rupture strain

Beyond the creep rate minimum, we have of course the following relation:

$$\varepsilon^{pl}_{\theta} - \varepsilon^{pl}_{\theta\,min} \geqslant v^{pl}_{\theta\,min}(t - t_{min}), \tag{17}$$

therefore, according to the relation (15), the rupture strain $\varepsilon^{pl}_{\theta R}$ at the rupture time $t_R \approx t_{inst}$ verifies:

$$\varepsilon^{pl}_{\theta R} \geqslant \varepsilon^{pl}_{\theta\,min} + \frac{1}{m(1-Q)}. \tag{18}$$

According to its estimation (Section 4.4) $\varepsilon^{pl}_{\theta}$ reaches the right hand value at about 63% of the time from t_{min} to $t_R \approx t_{inst}$. So, the margin between this conservative estimation of the rupture strain and its actual value appears sufficient.

5. Proposition of a rupture criterion

5.1. Unirradiated material

The precedent analysis and thus the relation (18) apply to the unirradiated material because neither annealing nor damage appear and because rupture occurs practically at the beginning of the plastic instability (Section 3.1). Furthermore, for lack of precise enough knowledge of the strain hardening coefficient as a function of strain, we neglect $\varepsilon^{pl}_{\theta\,min}$ and propose the following conservative criterion to prevent the cladding rupture ($Q \approx -1$):

$$\varepsilon^{pl}_{\theta} \leqslant \frac{1}{2m} \quad \text{with } m = \frac{\sigma'_\theta}{v^{pl}_\theta}\frac{\partial v^{pl}_\theta}{\partial \sigma'_\theta}(\sigma'_\theta, \varepsilon^{pl}_\theta) \tag{19}$$

with the true values σ'_θ and ε^{pl}_θ of stress and plastic (or creep) strain at the considered time. We propose the use of this criterion for every conditions although it has only been justified for constant pressure.

The secondary creep term of the law of AS [4] is of the following form as a function of stress and absolute temperature T, as the creep law for storage [5]:

$$v^{pl}_\theta = V_0(\varepsilon^{pl}_\theta).\exp\left(-\frac{T_a}{T}\right).\left(sh\left(\frac{\sigma'_\theta}{\sigma_c}\right)\right)^{m_0}. \tag{20}$$

In this case, the following expression of the relative sensitivity of strain rate to stress can be deduced:

$$m = m_0 \frac{\sigma'_\theta}{\sigma_c} \Big/ th\left(\frac{\sigma'_\theta}{\sigma_c}\right). \tag{21}$$

So m_0 is the value of m for a zero stress and m is almost proportional to stress beyond $3\sigma_c$. For both of the two above mentioned laws, the values of the parameters are the following: $m_0 = 1$ and $\sigma_c = 34$ MPa.

The maximal allowed creep strain at constant pressure has been calculated as a function of the initial stress, with the relations (19) and (21) and these values of parameters, and taking into account the stress increase during creep ($\sigma'_\theta \approx \sigma'_{\theta c}\exp(2\varepsilon^{pl}_\theta)$). The maximal allowed creep strain at constant stress would be a little higher (Fig. 10). The ratio of the actual rupture strain to the maximal allowed strain, at constant pressure, is at least 1.7 and 3.5 in average (with true strains). Like the actual rupture strain, the maximal allowed strain decreases as a function of stress and is not dependent on temperature in the studied range (Fig. 10). According to the relations (19) and (21), it is due to the increase of m with stress and its independence of temperature.

This criterion should be confirmed at low stress. However the limit value of 0.5 or 0.65 for the true or conventional maximal allowed strain at zero stress is not aberrant compared with the measured rupture strain at the minimal considered stress (240 MPa).

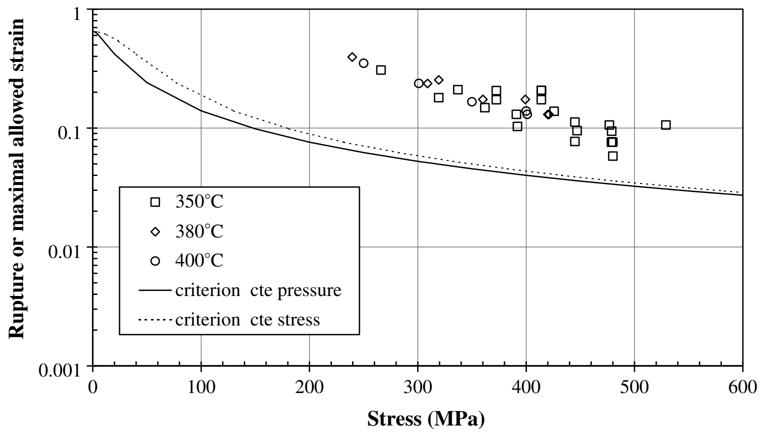

Fig. 10. Unirradiated CWSR Zircaloy-4: comparison of the proposed conservative criterion to the experimental results (conventional stress and strain).

5.2. Irradiated material

In this case, the analysis of Section 4 do not apply:

- at high stress, notably because rupture appears clearly before the plastic instability could occur,
- at low or moderate stress, although the plastic instability seems to be the determining factor of rupture, because of annealing.

So we have established an empirical rupture criterion for the irradiated material by multiplying the maximal allowed strain for the unirradiated one by a factor which follows the mean evolution of the rupture strain with the fast neutron fluence ϕ_t:

$$\varepsilon_\theta^{\mathrm{pl}} \leqslant \frac{1}{2m}\exp(-a\phi_t^b). \tag{22}$$

With fluence in $\mathrm{n\,m^{-2}}$, the parameters which fit the experimental results are: $a = 0.000011$ and $b = 0.2$.

The ratio of the rupture strain to the maximal allowed strain ranges from 1.3 to 10 for the whole data base (with true strains). Except below $2\times10^{25}\ \mathrm{n\,m^{-2}}$ at 350 °C and some cases at stress relevant of transport or storage, this ratio remains practically below 4. So the present criterion is effectively conservative but not overmuch. As it is shown on Fig. 11 for highly irradiated material, this criterion fairly follows the evolution of the rupture strain as a function of stress.

The limit of 0.1 for the maximal allowed strain when stress decreases is plausible because this strain is generally reached or passed before rupture for the stress level from 100 to 200 MPa. At stress of about 600 MPa, the maximal allowed strain is almost equal to the uniform elongation in burst test at high fluence [2]. At this level of stress and irradiation, the acceptance of higher strain

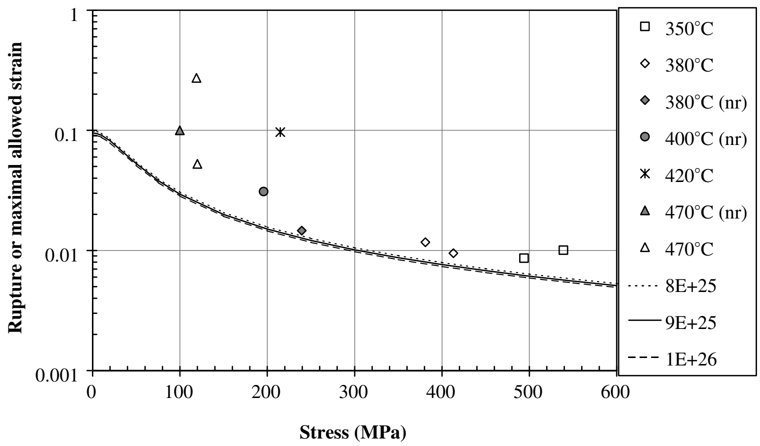

Fig. 11. CWSR Zircaloy-4 irradiated for four PWR cycles comparison of the conservative rupture criterion to the experimental results.

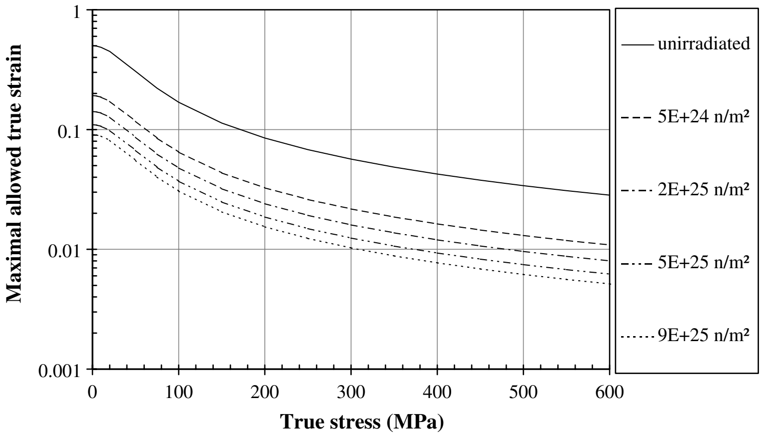

Fig. 12. CWSR Zircaloy-4 cladding maximal allowed creep strain under internal pressure as a function of stress and fast neutron fluence ($n\,m^{-2}$).

would be hazardous because of the brittle characters of rupture.

Last, as one can see on Fig. 12, the maximal allowed strain decreases very quickly up to an irradiation of about 2×10^{25} $n\,m^{-2}$ and slowly beyond.

6. Conclusion

Creep tests at constant internal pressure were realized up to rupture on CWSR Zircaloy-4 cladding. The rupture strain of the unirradiated material decreases from about 0.4 to 0.08 as a function of stress between 240 and 530 MPa, independently of temperature from 350 to 400 °C. At 350 or 380 °C, at stress between 300 and 550 MPa, the rupture strain decreases regularly with fast neutron fluence down to about 0.01 at $\approx 9 \times 10^{25}$ $n\,m^{-2}$. At this level of irradiation, for stresses between 100 and 240 MPa, the rupture strain is higher than 0.05 in the temperature range from 380 to 470 °C, because of annealing.

For the unirradiated material or the irradiated one submitted to moderate or low stress, the plastic instability appears as the major mechanism of rupture according to the comparison between calculation and experimental results. In the first case, it is essentially due the stress increase with strain although it is accelerated by the annealing of irradiation hardening in the second one. At high stress, the irradiated material bursts clearly before the plastic instability because of low ductility.

From analytical study in the hypothesis of zero strain hardening for the unirradiated material, and fit to the experimental results for the irradiated one, we propose a conservative creep rupture criterion for CWSR Zircaloy-4 submitted to internal pressure (relations (19), (21) and (22)). According to this criterion, the maximal allowed strain is a decreasing function of stress and fast neutron fluence and does not depend on temperature in the studied range.

For application to transport and interim storage, it is clearly less conservative than the usual criterion is. At high stress, it is more restrictive but in this case the application of the usual criterion appears hazardous.

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