Considerations on LOCA criteria including high burn-up effect

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Definition of the LOCA criterion parameter T_{300s}

The 17% ECR was intended as a way to represent the temperature-time limit beyond which the ductility of a given cladding type drops to zero.

In fact, the actual LOCA criterion is on the temperature-time condition for a given cladding to reach zero-ductility.

As discussed in Appendix 1, the 17% ECR is often misinterpreted. It has several disadvantages, such as

- It is an abstract oxidation as it does not represent the actual cladding oxidation
- It is a derived variable, the real LOCA variables are temperature and time
- It is impractical for expressing margins (e.g., what would a 3% ECR margin mean?)

Instead of the 17% ECR, another parameter, i.e. the "*300-second Temperature*" or T_{300s}, can be used to express the LOCA zero-ductility criterion.

The T_{300s} is the temperature that would lead the cladding to loose all ductility when exposed to steam for 300s.

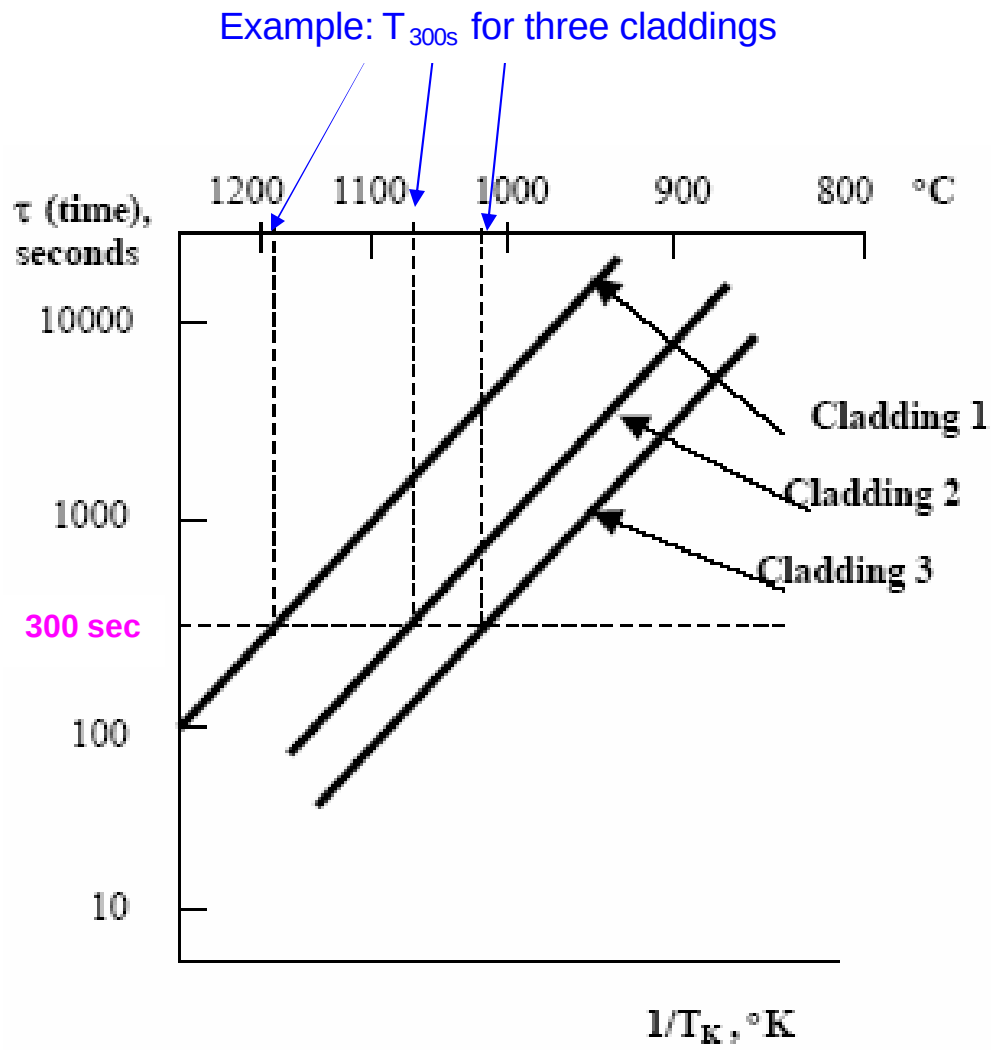


Fig 1. Zero-ductility limit for three different claddings (e.g. different thickness or material)

If one type of cladding had T_{300s}=1190ºC and another one T_{300s}=1060ºC as in Fig.1, one would have a direct information on LOCA time and temperature limit, in particular that the LOCA margin for Cladding 1 is ~130ºC larger than for Cladding 2.

Current failure criterion: the T_{300s} for un-irradiated Zircaloy-4

The current LOCA safety limits are based on the results obtained three decades ago. The tests were carried out by Hobson and Rittenhouse using test specimens cut from as-fabricated Zircaloy-4 cladding tubes.

In those tests, the aim was to establish the conditions under which the cladding looses its ductility and undergoes brittle failure.

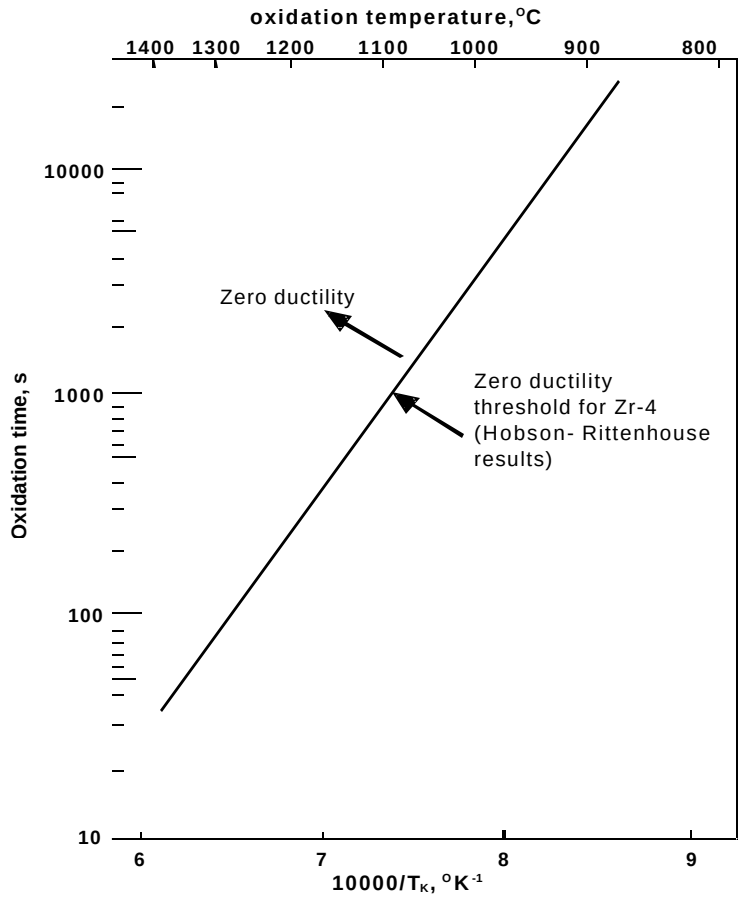
The loss of ductility was inferred by means of ring compression tests, on ring specimens cut from tubes that were previously subjected to high temperature oxidation – at constant temperature and in steam environment.

The oxidation was on open tubes, i.e. it was a two-side oxidation.

Summary of the Hobson and Rittenhouse test conditions

o Test type	Slow compression of ring specimens
o Crosshead speed	2.5 mm/min
o Maximum deflection	3.8 mm
o Test temperature	23 - 150°C
o Criterion for "failure"	Zero ductility
o Specimen type	Un-irradiated Zircaloy-4 rings
o Ring length	6.35 mm (1/4 in.)
o Ring outer diameter	10.72 mm
o Ring thickness	0.686 mm
o Specimen conditions	Tubes oxidised at high steam temperature
o Oxidation temperature	Constant, in the range 900 - 1300°C (1)
o Oxidation time	From 30 to 900 s
o Steam pressure	Atmospheric
o Surface conditions	Two-side oxidation

CURRENT CRITERION, UN-IRRADIATED Zr-4
HOBSON DATA



The zero-ductility threshold can be expressed by the equation[1]:

$$(1762/W) \cdot \exp(-11500/T_K) \cdot \nu t = 17 \qquad /1/$$

By re-arranging Eq/1/ and by setting the wall thickness W=0.686mm one gets:

$$T_K = 11500 / (4{,}64 - 2{,}3 \cdot \text{Log } W + 1{,}15 \cdot \text{Log } t)$$

$$T_K = 11500 / (5{,}02 + 1{,}15 \cdot \text{Log } t) \qquad /1'/$$

T_{300s}=1188°C

[1] The above is the so-called Baker-Just correlation, where the term 17 on the right hand side of Eq./1/ is the well known 17% ECR

EFFECT OF WALL THICKNESS

The effect of wall thickness for an un-irradiated Zircaloy-4 cladding can be evaluated by means of the equation

$$T_K = 11500 / (4{,}64 - 2{,}3 \cdot \text{Log } W + 1{,}5 \cdot \text{Log } t)$$

or

$$T_{300s} = 11500 / (7{,}49 - 2{,}3 \cdot \text{Log } W) - 273 \qquad /2/$$

The table bellow gives the T_{300s} as calculated for varying cladding thickness.

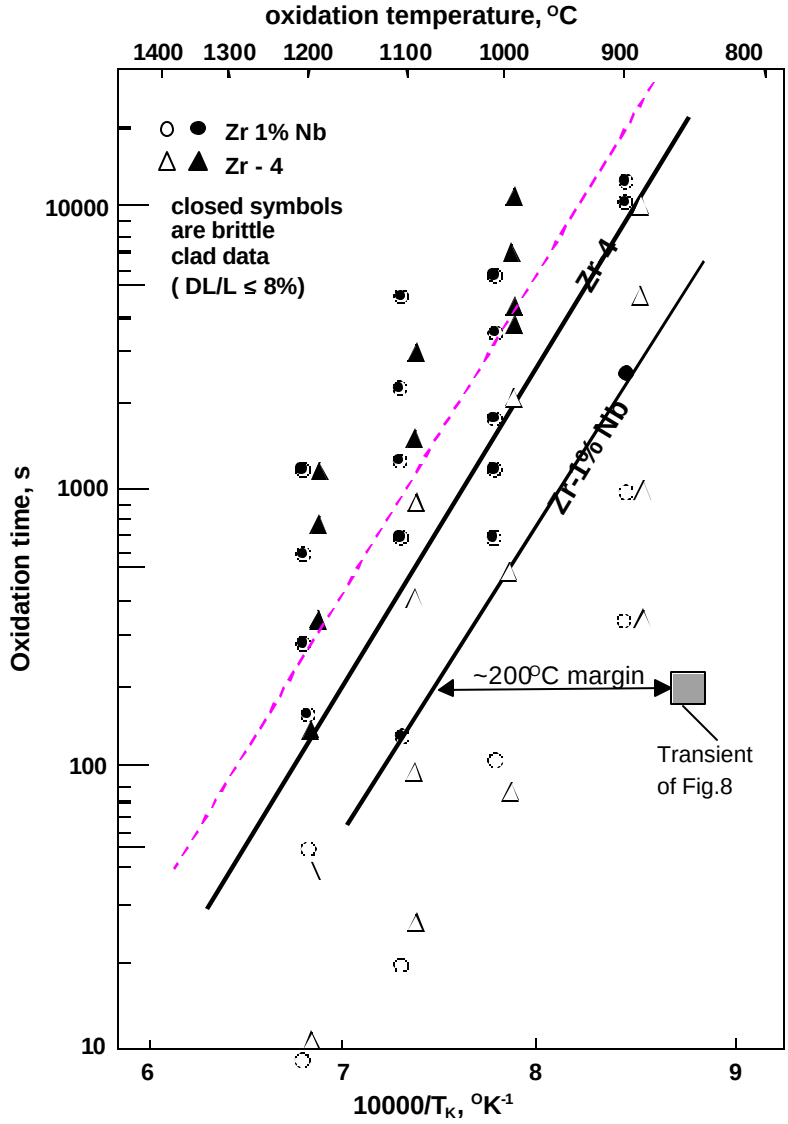
Wall thickness, mm	**T_{300s}, ºC** (Zircaloy-4)
0.686	1188
0.576	1157
0.500	1132
0.400	1095
0.300	1050

It shows that cladding with thinner wall is less resistant to LOCA, i.e. it would become brittle at somewhat lower temperature.

One can observe that the wall thickness effect is relatively small, in that one looses about 30-40ºC for a 0.1mm decrease in wall thickness.

UN-IRRADIATED E-110 vs. ZIRCALOY-4

Dotted line: Hobson-Rittenhouse



Studies have been conducted in various laboratories aiming to determine the behavior of the VVER Zr1%Nb alloy (E110) in LOCA-relevant conditions.

Results of ring compression tests performed at AEKI in Hungary [3][2]:

[2] In the AEKI ring compression tests the following geometry was used:
Zircaloy-4: 10.75mm OD, 0.625mm thick, 8mm long. E110: 9.1mm OD, 0.650mm thick, 8mm long.

The E110 line in Fig.3 corresponds to the following zero-ductility threshold:

T_K = 11500 / (5.91+1.15 . Log t) **/3/**

T_{300s}= 1140°C for Zircaloy-4
T_{300s}= 1040°C for E110

This shows that, the Zr-4 performs better than the E110, typically by ~100°C in terms of tolerable temperature [4].

One can observe that the T_{300s} for the Zircaloy-4 obtained in the Hungarian tests (T_{300s}= 1140°C) is somewhat lower than what one would expect based on the Hobson-Rittenhouse results (T_{300s}= 1172°C when one accounts for thickness).

One should notice that the conclusion that the E110 Zr-Nb alloy has a 100°C lower temperature-limit than Zircaloy-4 does not necessarily apply to any Zr-Nb cladding, but only to the specific one tested in Hungary.

Further, this difference does not necessarily mean that one alloy is "good" and the other one is "not good".

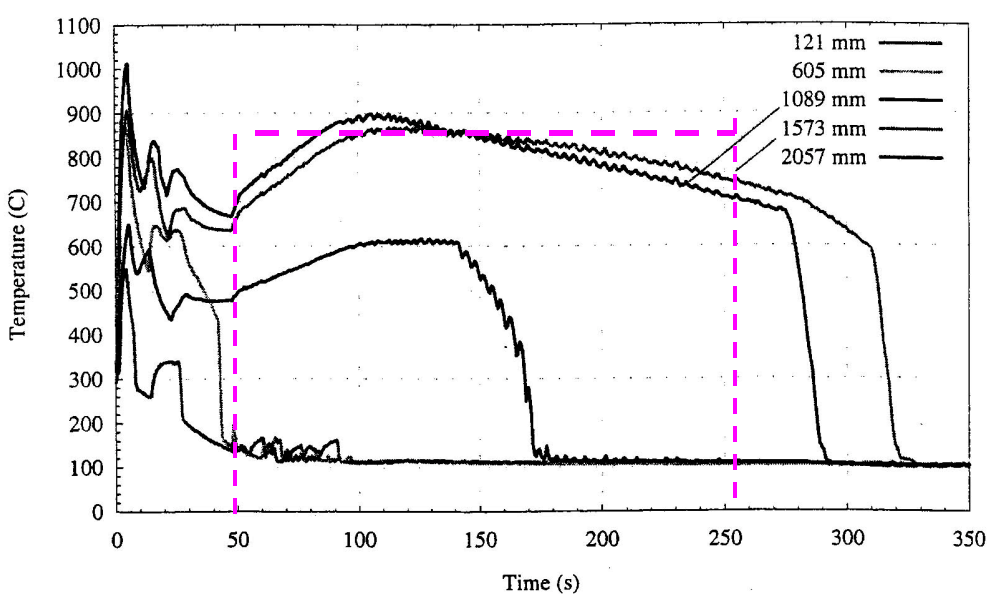
How good one alloy is must be related to the LOCA conditions in a given reactor type. If for instance LOCAs in VVERs result in lower cladding temperatures than in PWRs by 100°C or more, then the E110 cladding would have equal or greater safety margin in VVERs than Zircaloy-4 has in PWRs.

To illustrate this, the VVER LOCA transient shown in Fig. 4 has been considered. This transient results in an equivalent cladding temperature of ~850°C over an equivalent time period of ~200 seconds[3].

When the equivalent temperature-and-time point is plotted (square point) on the same diagram as in Fig. 3, it is clearly seen that, for the transient in question, the safety margin is still rather large, i.e. ~200°C in terms of cladding temperature. The advantage of using the 1/TK vs. Log t plot as in Fig. 3 is that the equivalent temperature and time calculated for the actual LOCA transient can be directly compared with the safety limit. The distance between the two provides a straightforward representation of the safety margin, as shown in the diagram of Fig.3.

Calculated LB LOCA temperatures for Paks NPP (ATHLET calculations)

Dashed line: equivalent temperature and time

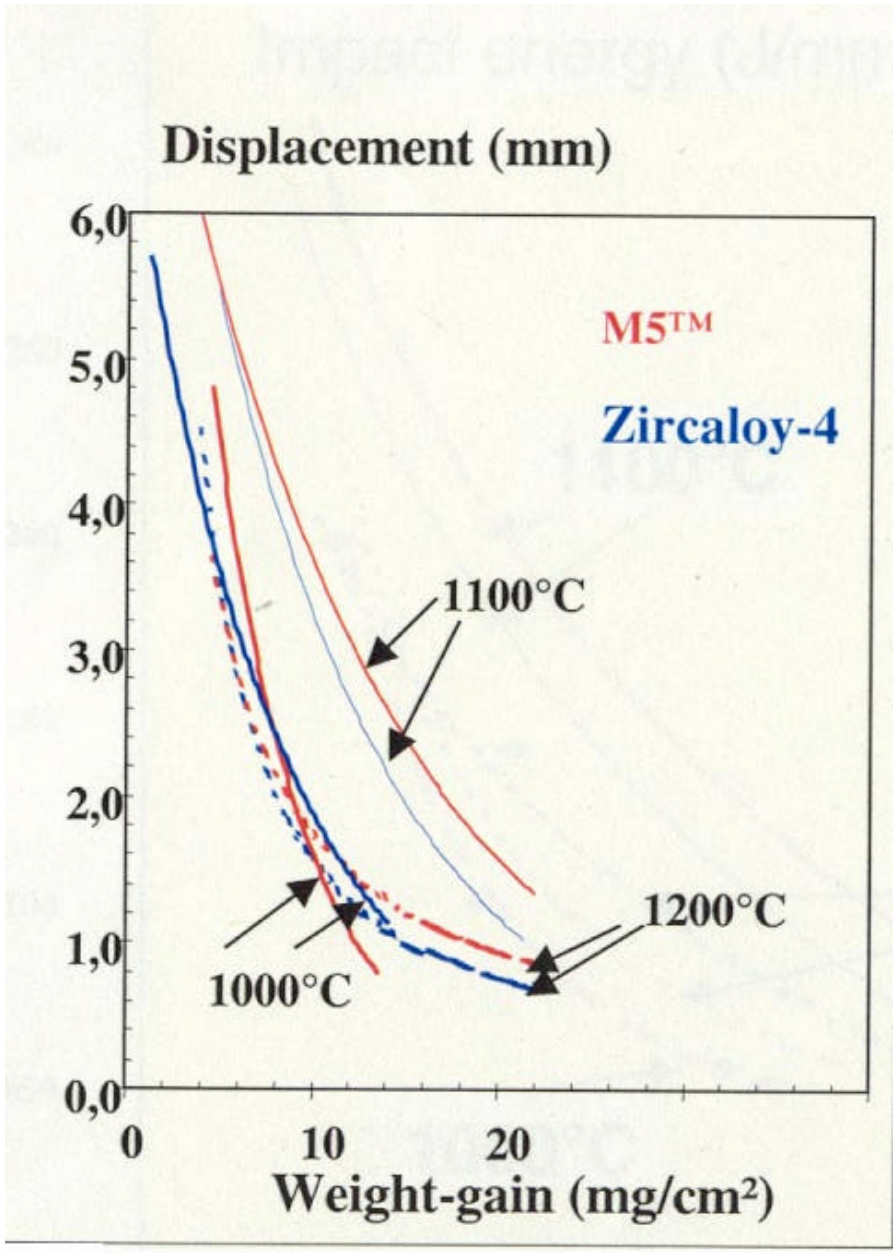


[3] The method to derive equivalent temperature and equivalent time is explained in Section 8.

The T_{300s} for the un-irradiated M5 cladding

The behavior of the M5 alloy in LOCA conditions as been studied in [6] by N. Waeckel. According to these results the M5 and the Zr-4 cladding behave similarly, in that they show virtually identical ductility reduction vs. corrosion. The latter is shown in Fig.5 [6]. This enables to conclude that the time-temperature relation for M5 zero-ductility is similar to that of Zr-4. One then gets:

T_{300s}= 1157°C for M5 (with W=0.576 mm, see table on Zr-4 in Section 3).



Effect of burn-up

Burn-up can affect the performance of the cladding at LOCA conditions, and in particular the residual ductility after the high temperature phase in steam, primarily because of the following reasons:

1. The cladding ductility is reduced due to irradiation damage during base irradiation

2. The cladding ductility is reduced due to hydrogen pick-up in steady state corrosion

3. The metal cladding wall thickness is reduced due to steady state corrosion. This reduces the cladding ability to withstand LOCA embrittlement (see e.g. Eq./1/)

These factors are discussed in the following. In doing that, the authors have relied on the experimental work carried out at JAERI, which has addressed the above separate effects through a fine experimental approach [7], which serves as basis for the provisional LOCA criterion set forth here.

Burn-up effect for Zircaloy-4

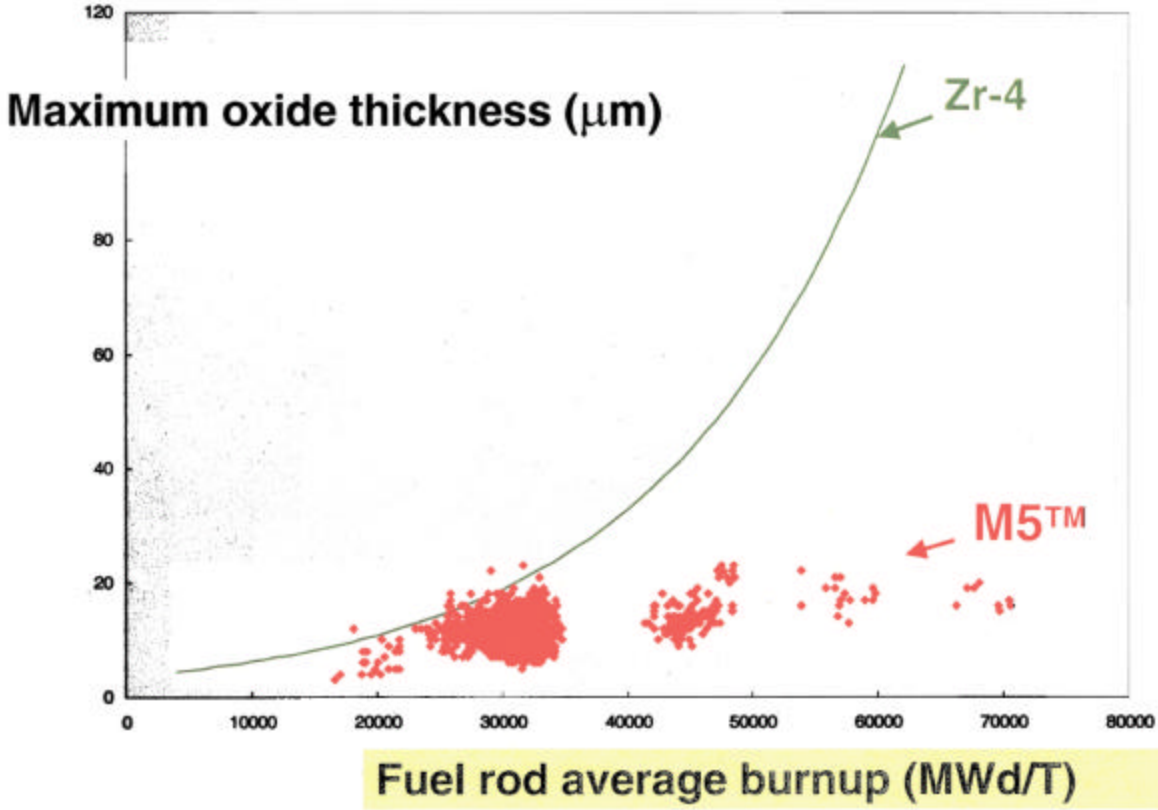
Irradiation damage. There is some evidence that the irradiation as such does not have important effects on post-LOCA ductility. The data presented in [7, 9] and in Appendix 2 show that the post-LOCA embrittlement is similar for un-irradiated and high burn-up cladding with low oxidation (48 MWd/kg, 20-30µm). For this reason, irradiation damage is not considered as a burn-up effect in this analysis, neither for Zr-4 nor for other alloys.

Hydrogen pick-up. Data on the separate effect of hydrogen have been presented in [7]. They were obtained by rod simulator tests, in which hydrogen had been forced into Zircaloy-4 cladding before the tests. The results show almost no hydrogen effect for unrestrained Zircaloy-4 tube specimens[4], whereas fully restrained tubes exhibited a relevant effect. In the latter case, the failure threshold, in terms of ECR, was reduced by a factor of 2.5 for hydrogen content up to 600ppm. However, there was no further effect for concentration exceeding 600ppm. Considering that 600ppm of hydrogen correspond to approximately 60µm of oxide, the hydrogen effect for Ox<60µm can conservatively be accounted for by the a multiplier [1-H/1000] or [1-Ox/100] to the right term of Eq./1/ (for Zr-4). For larger oxide thickness the multiplier is equal to 0.4. The term H is the Hydrogen content in ppm, Ox is the oxide thickness in µm.

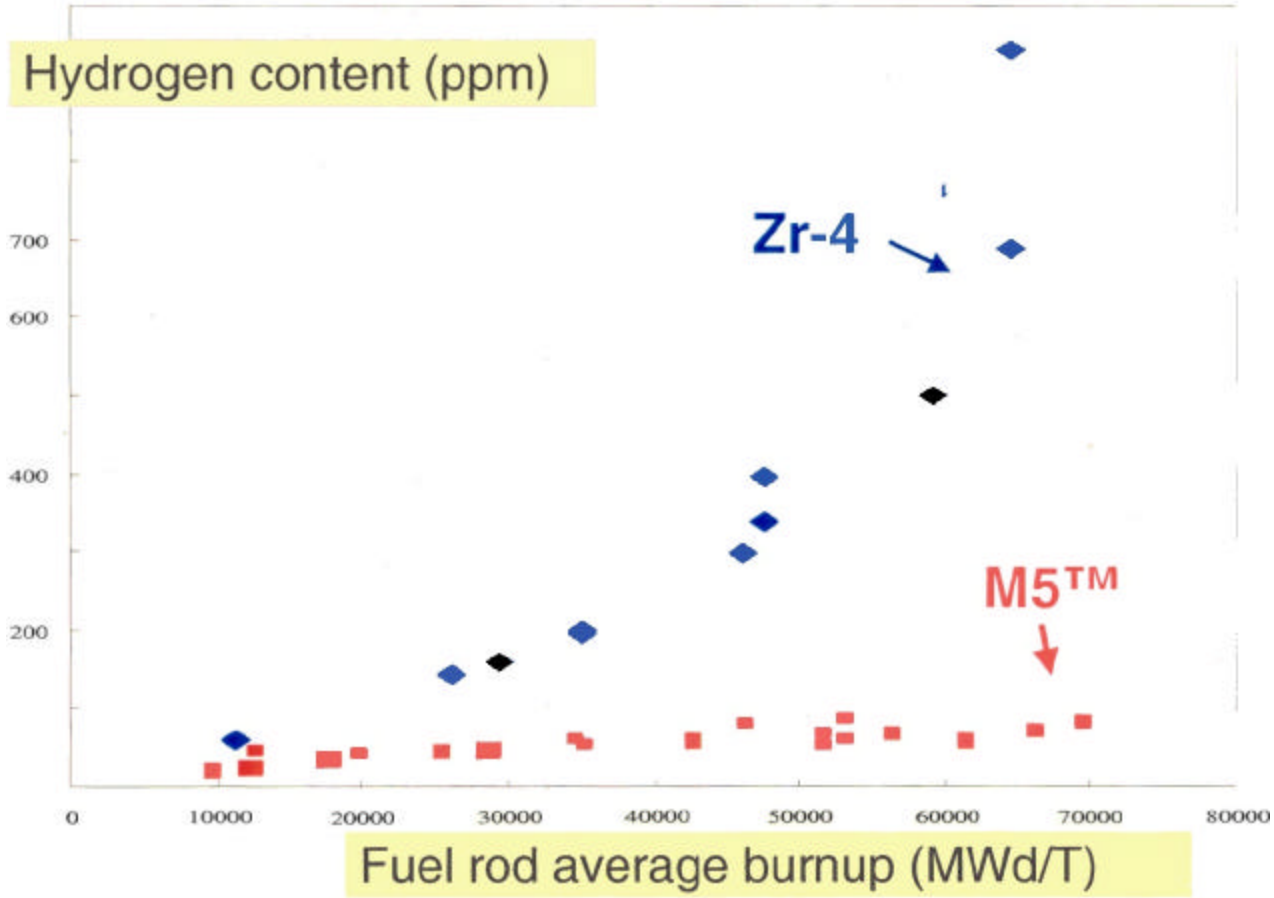
Wall thickness reduction. As found in [7], base irradiation oxidation does not "protect" against LOCA oxidation. On the other hand, the pre-existing corrosion weakens the cladding resistance to LOCA because the metal wall thickness is reduced. In the present evaluation, also the hydride rim at the cladding periphery has been subtracted. The wall reduction is thus accounted for by the term [1-1.7 .10^{-3} .Ox/W] as multiplier of the wall thickness W.

[4] This means for cladding tube specimens that were quenched without applying any constraint at the upper and lower end.

M5™ exhibits a much better in-reactor behavior than Zr-4



Hydrogen Uptake of M5™ is extremely low



Burn-up (oxide) dependent criterion

-1. Irradiation damage has negligible effect (JAERI data)

-2. Hydrogen up to 600 ppm can lower ductility limit (JAERI data)

-3. Base oxidation reduces wall thickness

Point 2 and 3 can be expressed as function of oxide thickness

Burn-up (oxide) dependent criterion

Zircaloy-4 (W=0.576mm)

$$T_{300s} = 11500 / (8.04 - ?'_{ox}) - 273$$

with $?'_{ox} = 2.3 \cdot \mathrm{Log}[(1 - 3.10^{-3}.Ox) \cdot (1-Ox/100)]$ for Ox<60µm

or $?'_{ox} = 2.3 \cdot \mathrm{Log}[(1 - 3.10^{-3}.Ox) \cdot 0.4]$ for Ox=60µm

M5 (W=0.576mm):

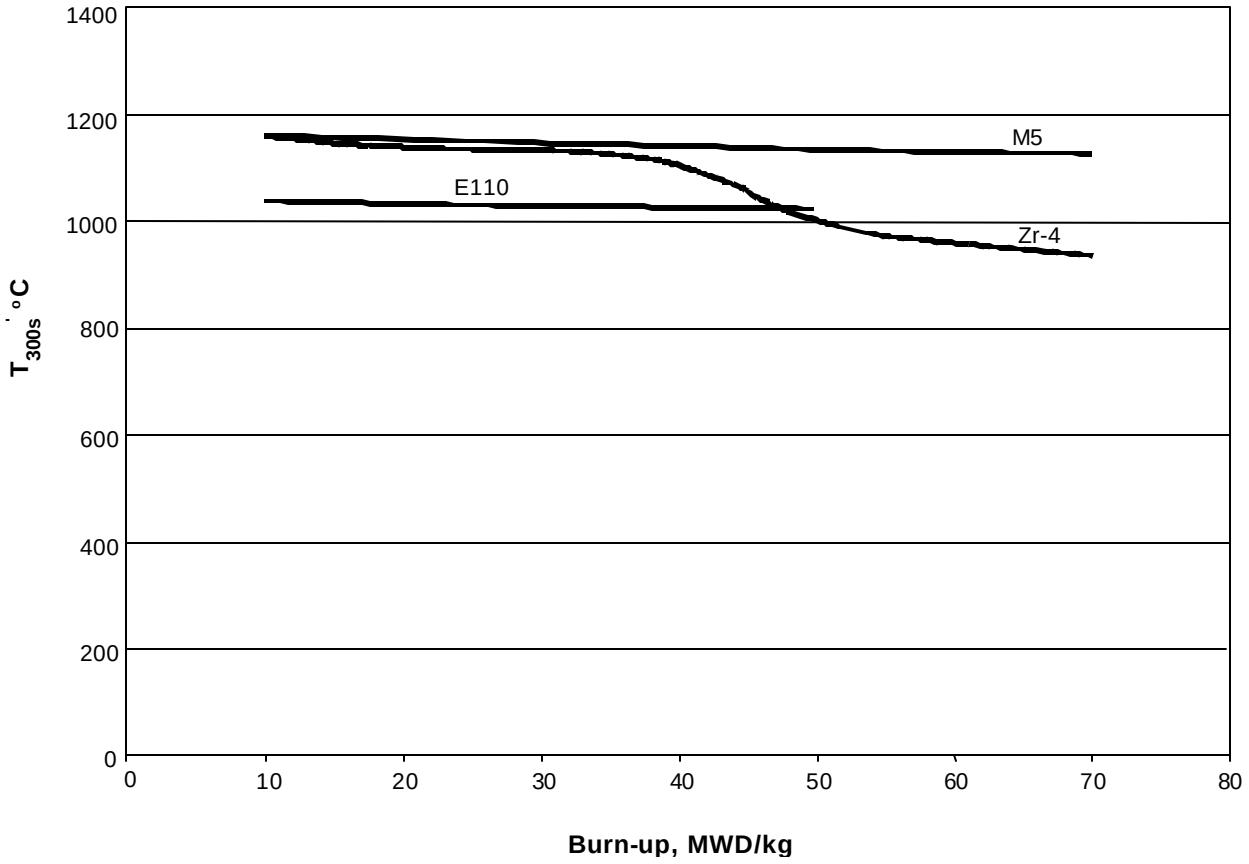
$$T_{300s} = 11500 / (8.04 - ?'_{ox}) - 273$$

with $?'_{ox} = 2.3 .\mathrm{Log}[(1 - 3 .10^{-3} .Ox) \cdot (1-Ox/200)]$ for x<120µm

E110 (W=0.650 mm)

$$T_{300s} = 11500 / (8.76 - ?'_{ox}) - 273$$

with $?'_{ox} = 2.3.\mathrm{Log}[(1 - 3.10^{-3}.Ox) \cdot (1-Ox/200)]$ for x<120µm

Result: No explicit dependence on burn-up, but dependence on oxide thickness

Effect of burn-up for Zr-4, M5 and E-110