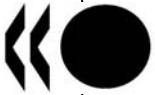


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English - Or. English

**NUCLEAR ENERGY AGENCY
COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS**

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REVIEW OF HIGH BURN-UP RIA AND LOCA DATABASE AND CRITERIA

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CSNI constitutes a forum for the exchange of technical information and for collaboration between organisations which can contribute, from their respective backgrounds in research, development, engineering or regulation, to these activities and to the definition of its programme of work. It also reviews the state of knowledge on selected topics of nuclear safety technology and safety assessment, including operating experience. It initiates and conducts programmes identified by these reviews and assessments in order to overcome discrepancies, develop improvements and reach international consensus in different projects and International Standard Problems, and assists in the feedback of the results to participating organisations. Full use is also made of traditional methods of co-operation, such as information exchanges, establishment of working groups and organisation of conferences and specialist meeting.

The greater part of CSNI's current programme of work is concerned with safety technology of water reactors. The principal areas covered are operating experience and the human factor, reactor coolant system behaviour, various aspects of reactor component integrity, the phenomenology of radioactive releases in reactor accidents and their confinement, containment performance, risk assessment and severe accidents. The Committee also studies the safety of the fuel cycle, conducts periodic surveys of reactor safety research programmes and operates an international mechanism for exchanging reports on nuclear power plant incidents.

In implementing its programme, CSNI establishes co-operative mechanisms with NEA's Committee on Nuclear Regulatory Activities (CNRA), responsible for the activities of the Agency concerning the regulation, licensing and inspection of nuclear installations with regard to safety. It also co-operates with NEA's Committee on Radiation Protection and Public Health and NEA's Radioactive Waste Management Committee on matters of common interest.

FOREWORD

This document is intended to provide regulators, their technical support organizations and industry with a concise review of existing fuel experimental data at RIA and LOCA conditions and considerations on how these data affect fuel safety criteria at increasing burn-up. It mostly addresses experimental results relevant to BWR and PWR fuel and it encompasses several contributions from the various experts that participated in the CSNI SEGFSM activities. It also covers the information presented at the joint CSNI/CNRA Topical Discussion on high burn-up fuel issues that took place on this subject in December 2004.

The document was assembled by C. Vitanza and M. Hrehor upon suggestion from the CSNI chair. It was presented at the SEGFSM meeting in April 2005. Comments made by the SEGFSM members have been incorporated in the final version of the document. The document content does not necessarily embody the opinion of the organizations to which each Group member belongs.

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1. INTRODUCTION AND MAIN CONCLUSIONS

This paper follows a discussion that took place in a joint CSNI-CNRA session in December 2004. The objective was to review the database gathered through testing in RIA and LOCA conditions and to assess the status of development of new criteria taking into account the effect of burn-up. During the discussion, which was moderated and led by Mr. J.M. Conde of the Spanish CSN, presentations were made by the following authors:

- K. Valtonen of STUK, Finland, on the outcome of typical RIA enthalpy depositions and LOCA cladding temperature transients at high burn-up for BWR and PWR cores
- J. Papin of IRSN, France, on the database, main results and plans of the Cabri test programme on RIA
- T. Fuketa of JAERI, Japan, on the database, results and plans of the NSRR RIA programme and on the outcome and plans of the LOCA quench tests at JAERI
- F. Eltawila of the USNRC on the status, main results and plans of the LOCA programme conducted at the US Argonne National Laboratory

An overview of the correlations published in the literature for the RIA failure threshold vs. burn-up was presented by C. Vitanza of the NEA secretariat. On LOCA he presented a provisional LOCA criterion based on ductility tests done with pre-hydrided, non-irradiated cladding specimens. The above presentations are available at the NEA secretariat.

In preparing this paper, it was decided to organise and present the database from various laboratories in a consistent manner, in order to facilitate the comparison of data from different sources. While recognising that valuable results have been obtained also by other laboratories, the focus of this paper is on those tests carried out in NEA member countries which are directly applicable to PWR and BWR fuels. For what concerns PWRs, most of the current data concern Zr-4 cladding. The limited available data for Zirlo, M5 and E110 cladding are also presented here.

After the CSNI-CNRA discussion of December 2004, new results have been generated especially in the LOCA area. These refer in particular to the ductility tests carried out at Argonne on high burn-up cladding – the first high burn-up ductility data produced so far. Because of these recent results, the conclusion of this paper on LOCA is slightly different from the conclusion of the CSNI-CNRA discussion.

The conclusions are as follows:

- On RIA, there is a well-established testing method and a significant and relatively consistent database from NSRR and Cabri tests, especially on high burn-up Zr-2 and Zr-4 cladding. It is encouraging that several correlations have been proposed for the RIA fuel failure threshold. Their predictions are compared and discussed in this paper for a representative PWR case.
- On LOCA, there are two different test methods, one based on ductility determinations and the other based on “integral” quench tests. The LOCA database at high burn-up is limited to both testing methods. Ductility tests carried out with pre-hydrided non-irradiated cladding show a pronounced hydrogen effect. Data for actual high burn-up specimens are being gathered in various laboratories and will form the basis for a burn-up dependent LOCA limit. A provisional burn-up dependent criterion is discussed in the paper.

2. THE CABRIA DATABASE

The CABRIA database consists of 14 tests, which were carried out in the decade 1993-2002. The test conditions and range of most important parameters and fuel variants are given in the following table:

Coolant	Liquid sodium
Coolant initial temperature	280°C
Coolant flow rate	4 m/s
Coolant pressure	~3 bar
Test fuel	Re-fabricated segments, ~56 cm active length
Fuel origin	PWR reactors
Fuel type	UO ₂ (10 tests) and MOX (4 rods)
Cladding type	Zr-4 (11 tests), M5 (2 tests), ZIRLO (1 test)
Pulse width	Variable, typically 9, 30 and 75 ms
Fuel enthalpy (at peak position)	~200 cal/g at 30 MWd/kg, ~100 cal/g at 60 MWd/kg for UO ₂
Axial power shape	Peaked at mid height, peak factor=~1.2-1.3
Burn-up range	30-75 MWd/kg
Oxide thickness	10-130 µm

Fuel cladding failure occurred in four tests, whereas no failure was registered in the remaining ten tests. Of the four tests that resulted in fuel failure, three had UO₂ fuel and one MOX fuel. All failures were with Zr-4 cladding, whereas no failure occurred in the three tests with M5 or ZIRLO cladding. The three UO₂ failures occurred at enthalpy below 80 cal/g and on fuel that had a burn-up of ~60 MWd/kg and significant corrosion (80 to 130 µm). However, several other CABRIA tests were run at comparable burn-up and level of corrosion, without producing failure. Oxide spalling appears to be the distinctive element that separates the failed and non-failed fuel in CABRIA UO₂ tests, in that all three UO₂ fuel rods that failed had spalling, whereas the rods that didn't fail had uniform non-spalled oxide.

One MOX test out of four resulted in fuel failure. The failure occurred on a fuel rod that had burn-up of 55 MWd/kg and moderate corrosion, 50 µm (without oxide spalling). This is the only CABRIA failure that took place on a non-spalled cladding. However, failure in this case occurred at 113 cal/g, which is a rather high enthalpy level, where a failure may not be surprising, considering that this level is near the upper envelope of all CABRIA data.

Failures were not associated to a particular pulse width. The three UO₂ fuel rod failures occurred at all three pulse widths that have been used in CABRIA, i.e. 9, 30 and 75 ms.

A summary of the main parameters and outcome of the CABRIA tests are listed in Table 1. The achieved enthalpy – or the failure enthalpy for the test fuel rods that failed – is plotted as a function of burn-up in Fig. 1.

3. THE NSRR DATABASE

The NSRR database consists of 26 tests of PWR fuel, 16 tests of BWR fuel and 28 additional tests with non-commercial or ATR (MOX) fuel. The tests were carried out in the period 1989-2003. The test conditions and the range of most important parameters is given in the following table:

Coolant	Water
Coolant initial temperature	20-85°C
Coolant flow rate	Stagnant
Coolant pressure	1 bar
Test fuel	Re-fabricated segments, ~10 cm active length
Fuel origin	PWRs, BWRs, ATR and JMTR (test reactor)
Fuel type	UO ₂ , MOX in 6 cases
Cladding type	Zr-4, MDA (1 test), ZIRLO (1 test) for PWR, Zr-2 for BWR fuel
Pulse width	4-5 ms in most cases
Fuel enthalpy (at peak position)	~200 cal/g at 30 MWd/kg, ~120 cal/g at 60 MWd/kg
Axial power shape	Uniform
Burn-up range	0-65 MWd/kg
Oxide thickness	0-60 µm

Failure was registered in 5 out of 26 tests with PWR fuel and in 5 out of 16 tests with BWR fuel. The five BWR failures belong to one specific set of fuel. Seven failures were registered in the other 28 tests. Failures appear to start at the cladding outer surface and propagate in a brittle mode up to ~40% of the wall thickness and then continue to propagate in a ductile manner [1].

There is currently no clear-cut explanation as to why some fuel rods failed and others didn't at apparently similar conditions. In particular, there is one set of BWR rods that failed at 61 MWd/kg - 5 out of 5 for enthalpy between 70 and 86 cal/g [2] -, whereas the same fuel in the 40-56 MWd/kg range did not fail even at 130-145 cal/g.

The NSRR failures occurred at moderate enthalpy both for Zr-2 and Zr-4, at enthalpy typically in the range 60-80 cal/g. Failures occurred also for cladding that had low corrosion and hydrogen pick-up and that had no spalling, suggesting that at low coolant temperature as in the NSRR, the cladding may become brittle even for a small amount of absorbed hydrogen. None of the MOX fuel rods tested in NSRR failed, but burn-up was low in these cases [3]. A ZIRLO cladding with UO₂ fuel failed, but enthalpy was very high for the burn-up level of that test (120 cal/g at 58 MWd/kg) [4].

A summary of the main parameters and outcome of the NSRR PWR and BWR tests are presented in Table 2. The achieved enthalpy – or failure enthalpy for fuel that failed – is plotted versus burn-up in Fig. 2. (Additional NSRR tests were run after this note was written).

4. OTHER DATABASES

RIA tests at cold coolant conditions were performed in the SPERT and PBF facilities (USA) in the 1968-70 and 1978-80 time-periods respectively. RIA tests on VVER fuel were also carried out in the Russian IGR, BGR and Gidra reactors (1984-2000)¹.

There were six fuel failures in the SPERT tests, three of which occurred for burn-up of 1-4 MWd/kg and for enthalpy higher than 190cal/g, which is not surprising. However, a fourth low burn-up failure occurred at a lower enthalpy, i.e. 147cal/g, a point that will be briefly commented below. Only two SPERT tests were made at appreciable burn-up, i.e. 32 MWd/kg, resulting in fuel failures at <143 cal/g and at 85 cal/g respectively.

Within the PBF experimental series there are only six tests which are relevant for fuel failure threshold assessment. They were run with fuel having burn-up between 0 and 5 MWd/kg. No failure was registered in five tests run at high enthalpy, i.e. 185 cal/g or higher, whereas failure occurred in one test at 140 cal/g.

These databases can thus be summarised as follows:

- There were two failures, one in the SPERT and one in the PBF series, which occurred for practically fresh fuel at enthalpies of 140 and 147cal/g. This contrasts with 11 other SPERT and PBF tests, 18 NSRR tests and 2 CABRI tests – altogether 31 tests showing a higher failure level than in the above two tests.
- More importantly, there was a SPERT failure at 85 cal/g for fuel burn-up of 32 MWd/kg, which is a very low failure enthalpy compared with NSRR and CABRI data at similar burn-up. A peculiarity of this test (and of the other SPERT tests at the same burn-up level that failed below 143cal/g) is that corrosion was as large as 65µm, which is very high compared with modern BWR fuel. The commercial BWR fuel tested in NSRR, for instance, had oxide thickness of 20-30µm at 60 MWd/kg.

The SPERT fuel was non-commercial, specially fabricated BWR fuel (with reduced cladding wall thickness in some cases). The burn-up accumulation didn't take place in a power plant but in a test reactor. Details of the base irradiation, e.g. on water chemistry control, are not known. The fuel was fabricated ~40 years ago, i.e. with the technology and quality available at that time, and as already mentioned, the corrosion was abnormally high compared with current BWR fuel. Bearing in mind the importance of corrosion for failure occurrence, consideration should be given to the above remarks when using these data for assessing failure thresholds of modern fuel.

VVER fuel failures in the IGR and BGR tests occurred at very high enthalpy, i.e. >~160 cal/g in pressurised fuel rods at zero burn-up and in the burn-up range 47 - 49 MWd/kg. At 60 MWd/kg, conservative failure thresholds of 130 and 140 cal/g have been derived for pressurised and respectively un-pressurised rods.

In accordance with [38], there is no concern with reduced failure enthalpy vs. burn-up in VVER fuel as long as the cladding corrosion and hydriding remain low. Further, the specific fuel pellet design, e.g. the pellet central hole also has an effect on cladding strain and rupture threshold.

¹ See additional Russian references number [36, 37, 38]

5. RIA FAILURE THRESHOLDS

Data renditions, correlations or models have been proposed by different authors for the RIA failure threshold. They are illustrated in Fig.3 and briefly described below.

The Japanese NSC threshold was introduced in 1998 [5] as a data rendition for the NSRR, CABRI, SPERT and PBF failures plotted vs. burn-up. The threshold is placed below all fuel failure data, except REPNa-1. No distinction is made between different fuels or between hot and cold RIA. The NSC criteria also consider a fuel coolability limit, which is considerably higher than the failure threshold (and which thus constitutes the actual RIA limit).

The fuel failure threshold proposed in [6] was developed primarily on the basis of CABRI data, but it was well shown to also reproduce the NSRR data. It makes a distinction between ductile and brittle cladding, with the latter having appreciably lower failure enthalpy. Oxide spalling (for Zr-4) and low coolant temperature (for Zr-2 or Zr-4 cladding) are considered as important causes of cladding brittleness. Burn-up and corrosion affect the threshold. There is no difference between UO₂ and MOX fuel or between BWR and PWR fuel. The differences between hot and cold coolant conditions are the lower initial enthalpy and the lower cladding ductility at cold conditions (for Zr-2 or Zr-4 cladding).

The KAERI correlation given in [7] is a fit of the CABRI, NSRR and SPERT failure data (non-failed data were not considered). Burn-up and corrosion are the most important parameters, whereas there is only a weak dependency on pulse width.

The fuel failure threshold proposed by the Swiss HSK makes a distinction between hot and cold coolant conditions and between UO₂ and MOX fuel [8]. The thresholds are based on the calculations performed with the FALCON code. For the HZP PWR case, the HSK threshold is practically the same as the one considered by EPRI. It is expressed vs. burn-up and is valid for corrosion up to 130µm. The CZP (Cold Zero Power) BWR case and the MOX fuel case have significantly lower thresholds than the HZP (Hot Zero Power) UO₂ case. The HSK criteria also have a fuel coolability limit, which is appreciably higher than the failure threshold.

The threshold proposed by SKI and other Swedish organisations [9] is derived on the basis of FRAPCON-SCANAIR calculations. The calculations give different thresholds depending on transient conditions such as pulse width and coolant temperature. For PWR HZP transients, the threshold depends on both burn-up and corrosion.

The Battelle threshold is based on calculations with the upgraded FRAPTRAN code [10]. As in the previous case, the threshold depends on pulse and temperature conditions. Burn-up and especially corrosion affect the threshold level.

The threshold proposed in the 2004 Research Information Letter (RIL) is expressed as a function of the corrosion level [11]. The threshold constitutes the lower envelope of the fuel failures from the NSRR, CABRI (except REPNa-1), PBF and SPERT data. The RIL threshold envisages a slight difference between hot and cold RIA conditions.

IRSN note: For IRSN, the existing database (mainly CABRI and NSRR tests) pointed out the inadequacy of the existing (i.e. zero burn-up) criteria for high burn-up fuel. Waiting for the establishment of suitable ones, the IRSN's current approach to justify the safety of new fuel

managements or cladding materials is based on a comparison of PWRs RIA transient and rod characteristics, compared to the existing data base (including CABRI REP Na1). A temporary "safety domain", based on peak fuel enthalpy, pulse width, oxide thickness and oxide spalling (which is considered as a major cause of cladding brittleness) has been used for safety assessment.

6. COMPARISON OF FAILURE THRESHOLDS FOR THE HZP CASE

The failure thresholds outlined above have been compared with each other for a PWR HZP case. In order to have a common basis for the comparison, the same oxide thickness vs. burn-up is assumed for all thresholds. The result of the comparison is shown in Fig.3. The oxide thickness considered is plotted versus burn-up in the upper part of the same figure.

All correlations predict a decrease of failure threshold with burn-up. In most cases the decrease is of roughly 100-150 cal/g, but in the HSK (EPRI) case the decrease is smaller, approximately 50 cal/g.

At low burn-up, i.e. up to ~30 MWd/kg, most thresholds are in the range of 170-200 cal/g. The RIL and NSC thresholds are however appreciably lower than that, typically from 100 to 150 cal/g between 0 and 30 MWd/kg. The NSC correlation threshold [5] is a data rendition that also includes the old SPERT data, particularly the failure data point at 32 MWd/kg and $\Delta H=85$ cal/g (i.e. $H=103$ cal/g for a HZP transient). None of the other correlations, except the RIL threshold, accounts for this particular data point.

At high burn-up, the thresholds range from 60 to 100 cal/g in most cases. For the correlations that acknowledge the oxide thickness dependence, a lower corrosion would result in a higher failure threshold. The HSK correlation is the one that gives, for large corrosion at high burn-up, the highest threshold for HZP RIA. However, no HZP tests have been carried out so far beyond enthalpy of 100 cal/g with highly corroded fuel (>80 μm) at high burn-up. This is partly because it is difficult to achieve a high enthalpy at high burn-up due to fissile depletion in the fuel. The CIP0-1 test in CABRI, for instance, achieved 92cal/g at 75MWd/kg. (A NSRR test with 78MWd/kg fuel and 81 μm corrosion was carried out while this report was being completed. The fuel failed at 61cal/g). As a final remark on predicted thresholds, no correlation accounts for the reported REPNa-1 failure enthalpy.

The failure thresholds are generally above the calculated enthalpies for RIA cases in actual LWR cores. The shaded areas shown in Fig.4 give the conservative upper envelope of enthalpy calculated for HZP RIA in PWRs [12] at low and high burn-up. As seen, there is a generally good margin at low burn-up, whereas at high burn-up, the calculated enthalpies are very close or slightly exceed the lower failure thresholds. Since the failure threshold is expected to increase for ductile and low corroded claddings, limiting corrosion and retaining ductility (i.e. avoiding spalling) are important measures to maintain a sufficiently large failure margin at high burn-up.

7. LOCA DATABASE, DUCTILITY TESTS

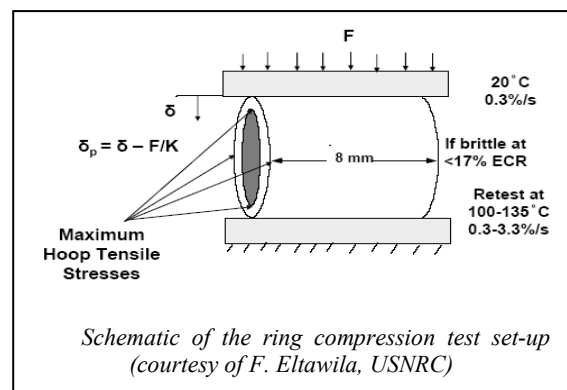
Hobson's data, non-irradiated Zr-4

In most countries, the current LOCA safety limits are based on ductility tests. The most relevant results were produced by Hobson and Rittenhouse [13a, b] using test specimens cut from Zircaloy-4 cladding tubes. Prior to cutting, the open tubes were subjected to two-side steam oxidation at various temperature levels and for a varied amount of time. Two-side oxidation was used in order to be representative of ballooned and burst tubes. After the high temperature oxidation phase, the sample residual ductility was determined by ring compression tests. The ring compression results were separated into two categories, i.e. those showing some residual ductility and those showing brittle behaviour. As expected, the "brittle" category consisted of those specimens that experienced longer exposures and/or higher temperature in the steam oxidation phase. The Hobson and Rittenhouse test conditions and specimen characteristics were as follows:

Test type	Slow compression of ring specimens
Crosshead speed	2.5 mm/min
Maximum deflection	3.8 mm
Test temperature	23 - 150oC
Criterion for "failure"	Zero ductility
Specimen type	Un-irradiated Zircaloy-4 rings cut from oxidised tubes
Ring length	6.35 mm (1/4 in.)
Ring outer diameter	10.72 mm
Ring thickness	0.686 mm
Specimen conditions	Tubes oxidised in high temperature and steam environment
Oxidation temperature	Constant, in the range 900 - 1300oC
Oxidation time	From 120 to 3600 s
Steam pressure	Atmospheric
Surface conditions	Two-side oxidation

The outcome of the Hobson-Rittenhouse tests and evaluations was that zero-ductility at 135°C is reached when the fractional thickness of combined [oxide + α -phase] layers relative to the wall thickness attains the value of 0.44. As said earlier, this occurs when the temperature is sufficiently high and the time-duration is sufficiently long. It was found that the temperature-time limit for zero-ductility could be expressed by the Baker-Just correlation for oxidation, where the Equivalent Cladding Reacted (ECR_{BJ}) was set equal to 17%. This limit is thus given by²

$$(1762/W) \cdot \exp(-Q/T_K) \cdot \sqrt{\tau} = 17 \quad \text{where } \tau \text{ is time in sec. and } T_K \text{ is temperature in } ^\circ\text{K.} \quad /1/$$



² The Baker-Just equation for two-side oxidation is: $ECR = (1762/W) \cdot \exp(-Q/T_K) \cdot \sqrt{\tau}$, where ECR is the percentage of metal wall thickness that has oxidized, W is clad thickness (mm), τ is the oxidation time (s) and T_K is the oxidation temperature in °K. $Q = 11450^\circ\text{K}$, although the value 11500 has also been used.

The additional database from ductility tests is presented in the following.

Hungarian data (AEKI), non irradiated Zr-4 and E-110

Ring compression tests have been carried out in VVER countries, including comparative assessments of the Zr-4 and VVER E110 alloy. Hungarian data include approximately 50 ring compression tests performed on Zr-4 and E-110 specimens oxidised at temperatures between 900 and 1200°C for time intervals ranging from 10 to 10000 seconds [14, 15]. The results are presented in Table 3. By setting the zero-ductility limit in correspondence with a crosshead displacement limit at a rupture of $\Delta l/d < 8\%$, and by plotting the brittle and ductile data on a time – temperature diagram (i.e. $\text{Log}t$ vs. $1/T_K$), one obtains the plot shown in Fig. 5a and 5b. As one can observe; the Zr-4 data are reasonably well separated by the 17% Baker-Just (BJ) limit, whereas for the E110 data; the limit is $\sim 100^\circ\text{C}$ lower and corresponds to a ECR_{BJ} of 8%.

Czech data (UJP), non irradiated Zr-4 and E-110, including pre-hydriding

Similar experiments were made in the Czech Republic [16, 17]. Also in this case there was a comparison between Zr-4 and E-110, but including a simulation of base irradiation corrosion made by pre-oxidising some of the specimens in autoclave. The data are shown in Table 4. With a ductile-brittle transition set at displacement $\Delta l/d \leq 8\%$, the results plotted in Fig. 6 are obtained (for as-received samples). One can again observe that the Zr-4 brittle and ductile data are delimited by the 17% BJ line, whereas for E-110 the limit is $\sim 8\%$. Specimen pre-oxidation resulted in a considerable drop of the ductility limit. This is clearly shown by the data in Table 4. According to the received information, however, pre-oxidizing in autoclave led to much higher hydrogen uptake than in actual reactor conditions.

US data (ANL) for non-irradiated Zr-4, ZIRLO and M5 (including pre-hydriding)

The ring compression tests performed at the Argonne National Laboratory (ANL) include non-irradiated Zr-4, ZIRLO and M5 cladding [18-21]. A total of ~ 60 tests were made at temperatures of 1000, 1100 and 1200°C. The results are listed in Table 5. Ductility was measured using the so-called “offset strain” as an indicator. Specimens with $< 2\%$ offset strain were classified as brittle. The outcome of the ANL test is depicted in Fig. 7a, b, c. The data show practically no difference between the three alloy types in terms of zero-ductility limit³.

One can observe from Fig. 7 that the $\text{ECR}_{\text{BJ}} = 17\%$ remains a valid and actually conservative limit at 1000 and 1100°C, whereas there is evidence that brittleness (at RT) can occur somewhat below that limit for the 1200°C case. At 1200°C, in fact, the ANL data show a ductile-brittle transition after ~ 100 seconds ($\text{ECR}_{\text{BJ}} = 12\%$). However, further ANL tests - as well as Hobson’s tests - showed that ductility increases significantly when ring compression is carried out at 100°C or 135°C instead of RT, even for $\text{ECR}_{\text{BJ}} > 17\%$ at 1200°C [31].

³ When the offset strain is plotted as a function of time for each of the three temperature levels, one can actually infer that ZIRLO and M5 behave generally better than Zr-4 in that they retain more ductility – on the assumption that offset strain is a reliable indicator of ductility. This has been shown for instance in [22].

Ring compression tests (at 135°C) of pre-hydrided, non-irradiated samples were also carried out at ANL. The main outcome was that [31]:

- Ductile-brittle transition occurs at $ECR_{BJ} = 6.5\%$ for $H \geq 600\text{ppm}$ (15x15 Zr-4)
- Ductile-brittle transition occurs at $ECR_{BJ} = 9.5\%$ for $H \geq 400\text{ppm}$ (15x15 Zr-4)
- For 17x17 Zr-4, ductile-brittle transition occurs at $ECR_{BJ} < 13\%$ for $H \geq 300\text{ppm}$

French data (CEA) for non-irradiated Zr-4 and M5 cladding, including pre-hydriding

In addition to the Czech and ANL data described above, the French CEA has also carried out experiments with non-irradiated, pre-hydrided cladding [23]. Both Zr-4 and M5 cladding were tested using various methods to deduct the post-oxidation residual ductility, including the ring compression method. Hydrogen concentration was 600ppm in Zr-4 and 200ppm in M5, reflecting the different level of corrosion expected for these two alloys at high burn-up. The outcome of the CEA tests is shown in Fig. 8. As-received, Zr-4 and M5 exhibited similar behaviour. Hydrogen addition had a marked effect on Zr-4 and a more moderate effect on M5 due to the lower H-content. Zero-ductility occurred at ECR_{BJ} between 5 and 10% for Zr-4 with 600ppm hydrogen⁴ and between 10-15% for M5 with 200ppm hydrogen. One can infer from this that the zero-ductility ECR drops roughly by a factor of 2.5 at 600 ppm ($17 \rightarrow \sim 7\%$) and by a factor of ~ 1.3 ($17 \rightarrow 13\%$) at 200ppm hydrogen. This can be expressed as follows:

$$ECR_{BJ}(\text{zero-ductility}) = 17\% \cdot [1 - H/1000] \quad (\text{from pre-hydrided Zr-4 and M5 samples, } H \leq 600\text{ppm}). \quad /2/$$

US data (ANL) for high burn-up Zr-4 cladding

While this note was being compiled, the NRC published new ring compression results for high burn-up Zr-4 cladding [24]. These are the first ductility data obtained from actual fuel cladding material. Ring compression was performed at 135°C. The fuel was retrieved from the H.B. Robinson reactor at high burn-up (~ 64 MWd/kg) and had a base-irradiation oxide layer of $\sim 70\mu\text{m}$ and H-content of $\sim 700\text{ppm}$. The cladding was de-fuelled and subjected to two-side oxidation, according to the time-temperature scheme shown in the upper left diagram of Fig. 9⁵. The corresponding effective temperature and time⁶ are given in the table on the same figure, together with the residual ductility results. The data, plotted on the usual time-temperature diagram as shown at the bottom-right part of Fig. 9, indicate that the ductile-brittle transition is somewhat below the 17% line, i.e. at about 11% BJ, which in terms of temperature means a $\sim 80^\circ\text{C}$ lower cladding temperature limit at high burn-up. It should be noticed that these first ANL tests do not account for oxide cracking occurring in the ballooning region, which might reduce the oxide protective effect. Future ANL tests intend to address this point. Based on these data the LOCA limit decrease would be:

$$ECR_{BJ}(\text{zero-ductility}) = 17\% \cdot [1 - 0.5 \cdot H/1000] \quad (\text{Based on H.B. Robinson cladding, } H = \sim 700\text{ppm}) \quad /2/$$

⁴ Fig 8 plots the results in term of measured ECR and not the conventional Baker-Just ECR. Further, the time-duration of the transient is not provided.

⁵ The upper right diagram shows that the burn-up effect is comparable with the ductility change from 135°C to RT for non-irradiated specimens.

⁶ The effective temperature and time correspond to a constant temperature transient, equivalent to the actual transient in terms of final ECR.

*Russian data for Zr1%Nb cladding (fresh and high burn-up)*⁷

Studies such as those conducted at the Russian Kurchatov Institute on the understanding of high temperature oxidation of Zr-Nb alloys have given important contributions to the understanding of the VVER E-110 alloy as compared to other Zr-1%Nb alloys [32]. As stated in [33]; the following general observations may be made on the basis of Russian experimental data:

- The irradiation inhibits the breakaway oxidation tendency on the outer surface of the E110 cladding;
- The tendency towards oxide spalling during high temperature oxidation is supplemented by the tendency towards an increase in oxidation rate on the cladding inner surface. In accordance with these data, it may be assumed that the contamination of the cladding inner surface by fission products is responsible for these effects.

The database characterising the residual ductility micro-hardness and hydrogen content in the oxidized irradiated cladding as a function of ECR has shown that:

- In accordance with the postulated relationship between the oxygen concentration in the prior β -phase, micro-hardness, and the oxygen induced zero ductility threshold, this corresponds to 8.3% measured ECR (i.e. 13% calculated with a Russian conservative correlation);
- The combination of the oxygen induced and hydrogen induced embrittlement of the E110 irradiated cladding leads to the reduction of the zero ductility threshold down to 6.5% measured ECR at the fast/fast combination of heating and cooling rates.

The investigations performed demonstrated that the oxidation behaviour of niobium-bearing alloys is very sensitive not only to alloying components but also to impurity composition. The analysis of the E110 problems in this context has shown that different methods are used for PWR and VVER zirconium alloy production. In fact, sponge Zirconium is used to produce the PWR alloys Zr-4, M5 and ZIRLO, whereas a mixture of iodide and electrolytic Zirconium is used for the fabrication of the VVER E110 alloy. This difference leads to differences in impurity composition.

Ring compression tests carried out after steam oxidation at 1100°C on modified E110 cladding produced from sponge Zr confirmed that the zero ductility threshold increased up to 19% ECR [33], due to low hydrogen uptake. The post-oxidation zero-ductility threshold of sponge E110 alloy was comparable to the ANL data. These results have prompted the Russian fuel manufacturer to move towards sponge zirconium alloy, as well as to modify the finish of the cladding surface (two-side polish and no etching).

The performed research enabled to make the conclusion that the oxidation behaviour and residual ductility of the E110 alloy are very sensitive to cladding micro-chemical composition and surface finishing. The use of sponge zirconium for the cladding fabrication results in a significant reduction of oxidation rate especially in the range 900-1000°C and in an increase of the zero ductility threshold. However, an increase of oxidation rate of sponge vs. standard E110 has been registered at 1200°C.

⁷ The text of this subsection was provided by experts of the Kurchatov Institute.

Summary of ductility tests

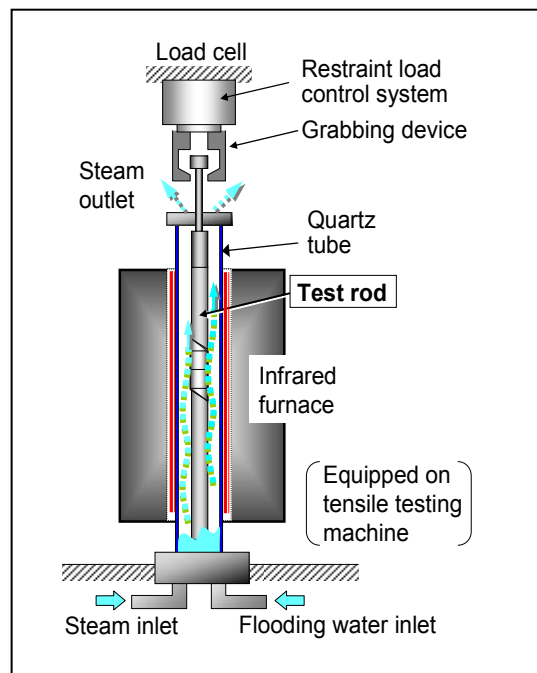
- ANL results show no significant difference between as received Zr-4, ZIRLO and M5
- CEA data show the same for Zr-4 and M5
- AEKI and UJP tests show a lower E-110 limit compared with Zr-4 ($ECR_{BJ} = 8\%$ for E-110 vs. 17% for Zr-4; i.e. a $\sim 100^\circ\text{C}$ lower tolerable temperature)
- UJP, CEA and ANL non-irradiated samples exhibit a significant hydrogen effect. ANL data at 135°C show a ductile-brittle transition at $ECR_{BJ} = 6-7\%$ for $H \geq 600\text{ppm}$ (15×15 Zr-4) and at $< 13\%$ for $H \geq 300\text{ppm}$ (17×17 Zr-4)
- However, preliminary ANL high burn-up data show that the ductile-brittle transition occurs at a level that is only moderately below 17% (at $11 \pm 2\%$). The reason for the difference between these and pre-hydrided cladding results is currently being debated. The tests do not simulate the oxide cracking occurring in the ballooning area.

8. LOCA DATABASE, QUENCH TESTS

JAERI tests with as received Zr-4 (zero burn-up)

The experimental basis for the LOCA criteria in Japan is constituted by quench tests conducted on cladding tube segments. These tests were performed at JAERI in the 80s [25]. The tubes were ballooned, ruptured, subjected to high temperature two-side oxidation and then quenched. During quenching there was full constraint, i.e. axial contraction was prevented during cooling. Full constraint represented a bounding case for the actual fuel assembly condition, since a reliable assessment of fuel-grid interaction and of consequent loading was difficult to make.

Approximately 50 experiments were carried out on non-irradiated Zr-4 cladding tubes, which served as a basis for the Japanese LOCA criteria. The results were divided into two categories, depending on whether the tube had survived the quenching or not. Fig. 10 provides a plot of the experimental results, showing the data points for the failed and non-failed tubes on a time vs. temperature diagram. As one can observe, the Baker-Just 15 % ECR line is a conservative rendition of the results. As most phenomena under LOCA conditions, including rupture, secondary hydriding from inner surface and axial loading⁸ during the quench, are taken into account in the quench tests, the 15% ECR criteria was adopted as the LOCA failure and coolability criterion in Japan.



Schematic of the quench test set-up
(courtesy of T. Fuketa, JAERI)

⁸ Considerations have however been expressed regarding the possible presence of additional transversal loads.

JAERI tests with non-irradiated, pre-hydrided cladding

Quench tests with pre-hydrided cladding have been carried out at JAERI with the purpose of determining the separate effect of hydrogen on quench failure. Tests carried out with H-content up to 600-800 ppm and full constraint show that the quench failure limit decreases substantially, typically from 17 to 7% [26]. This is consistent with the CEA ductility tests with pre-hydrided cladding. On the other hand, JAERI results show that the hydrogen effect is significantly smaller when the constraint is limited to 50 kg [27]. The difference between the full constraint and the 50 kg constraint is shown in Fig. 11. As for the case with a 50 kg constraint, it should be noticed that the scale is very sensitive and that all failure data points (as well as the two lines drawn in the figure) are above the BJ 17% limit.

JAERI and ANL tests with high burn-up cladding

Quench tests with high burn-up BWR and PWR fuel have recently been carried out at JAERI and ANL. At JAERI, three Zr-4 cladding tubes were cut from a PWR fuel rod having a burn-up of 48MWd/kg and moderate corrosion, i.e. 15-25 μ m. The hydrogen content was in the range of 120-210ppm. The tubes de-fuelled and filled with alumina pellets. They were then pressurized and subjected to steam oxidation at temperature between 1030 and 1180°C for a time ranging from 120 to 2195 seconds. Ballooning and burst occurred during the heat up phase. After holding at high temperature, the tubes were quenched while maintaining an axial tensile load of about 50 kg. Two of the tubes failed with a transversal crack during quenching, whereas the other specimens survived the quenching [27]. The choice of a 50 kg axial load in JAERI tests is based on evaluations and is still discussed [28, 29].

At ANL, two quench tests were done on a fuelled BWR rod segment having burn-up of 56 MWd/kg and 10 μ m oxide (~70 ppm H). There was no constraint on the segment during the quench test. The fuel segment was kept at 1204°C for 5 minutes. The cladding did not fail during quench, but the sample that was not embedded in epoxy failed during handling [30].

The results of the JAERI and ANL tests described above are summarized in Table 7 and in Fig.12. The latter clearly shows that the JAERI results are very well reproduced by the 17% ECR_{BJ} limit, in that the two failed tests are above this limit whereas the 4 non-failed ones are below it. The ANL data are just above the BJ 17% ECR line, but there was no failure during quenching in these two cases. One should however notice that these ANL tests were done without constraint - and that there was subsequent failure during handling at cold conditions in one case.

Russian tests

Considerable amount of quench tests has been done on both non-irradiated and irradiated VVER fuel in the Russian Federation [31]. These quench tests showed that the BJ 17% ECR limit holds also for the E-110 alloy. More general information on VVER fuel safety criteria can also be found in [34].

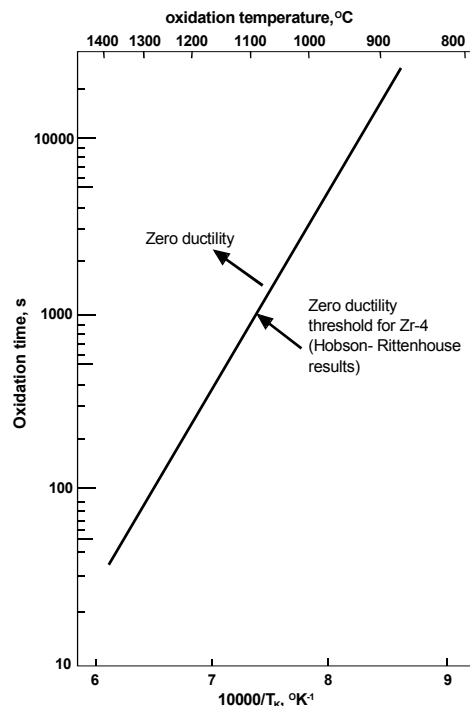
Summary of quench tests

- For as received Zr-4 samples, JAERI quench tests with full constraint give similar LOCA limit as the ductility tests (ECR_{BJ} =15% compared with 17%)
- Russian quench tests without constraint show that the BJ 17% limit holds also for the E-110 alloy, which contrasts with ductility test results (at RT)
- JAERI quench tests with full constraint show a failure limit reduction by a factor of 2.5 (17→7%) due to pre-hydriding, which is consistent with ductility test results
- JAERI (50 kg constraint) and ANL (no constraint) quench tests with high burn-up cladding show no quench failure below 17% ECR_{BJ}. Consensus on constraint level would be desirable.

9. LOCA SAFETY LIMIT

As shown in the previous sections, a straight line in a temperature-time chart ($1/T$ vs. Log Time) expresses the current LOCA limit. In most countries this limit is defined by the so-called *zero-ductility* boundary as derived from ring compression tests, which are done at cold conditions after the high temperature steam corrosion phase. The boundary separating the ductile from the brittle cladding data points defines the zero-ductility-based LOCA limit.

In other countries and notably in Japan the limit is defined by a different criterion, i.e. by whether fuel simulators *survive or fail during quenching* after the high temperature corrosion phase. The simulators are constituted of fuel segments or cladding tubes filled with alumina pellets. The boundary separating the non-failed from the failed cladding data points defines the quenching-based LOCA limit.



It so happens that (at least for Zr-4) the two approaches lead to virtually coincidental limits at zero burn-up if the quenching tests are done under full constraint conditions. The Baker-Just (BJ) equation, which in the past was used to describe high temperature corrosion, was also found to be a convenient means of expressing both the zero-ductility limit derived from ring compression tests and the failure limit derived from quench tests, apart from a very minor difference in a parameter setting (15 vs.17%). These limits are given by the two equations:

$$17=(1762/W)\cdot\exp(-Q/T_K)\sqrt{\tau} \quad \text{“Zero-ductility limit”} \quad /3/$$

$$15=(1762/W)\cdot\exp(-Q/T_K)\sqrt{\tau} \quad \text{“Quench limit”} \quad /4/$$

The graphic form of the BJ 17% limit is given by the line in the diagram shown above (for Hobson’s specimens, i.e. $W=0.686\text{mm}$). The BJ 15% limit is only $\sim 20^\circ\text{C}$ below this line.

One should notice that the Zr-4 17% ECR criterion implies the use of the BJ equation, regardless of how good this correlation is for oxidation. The criterion doesn’t say that the *actual* cladding oxidation should be below 17%, but that *temperature and time condition* should be below those leading to a calculated 17% ECR if the BJ correlation was used. Had one chosen another equation instead of BJ to calculate ECR, the zero-ductility limit would not have been expressed by the 17% ECR, but by some other value⁹ (while the temperature-time limit for zero ductility would obviously remain what it is).

Using measured ECR or using different ECR correlations may bring confusion on what these various ECRs mean in terms of temperature and time (these being the real LOCA parameters),

⁹ Had one chosen the Cathcart-Pawel equation, for instance, the limit would have been 13% - in order to obtain the same time-temperature limit as the 17% BJ limit.

especially when different alloys having different oxidation and ductility properties are considered. Further, at high burn-up, base irradiation ECR and high temperature ECR can be improperly mixed up. The use of a commonly acknowledged reference, such as the BJ equation, is therefore advisable. Alternatively, one can express the LOCA limit directly in terms of temperature and time – which are unambiguous parameters - instead of ECR. The two following tables give the main outcome of the zero and high burn-up investigations.

Summary of the LOCA tests

ALLOY TYPE	ZERO BURN-UP, AS RECEIVED			ZERO BURN-UP, PRE-HYDRIDED			HIGH BURN-UP		
	Ductility Tests	Quench, full constr.	Quench, 0-50kg constr.	-	Quench, full constr.	Quench, 0-50kg constr.	Ductility Tests	Quench, full constr.	Quench, 0-50kg constr.
Zr-4	BJ 17%	BJ 15%	-----	BJ ~7% for H≥ 600ppm	BJ ~7% for H≥ 600ppm	BJ 17% for H≥ 600ppm 50 kg constr.	Moderate burn-up effect, limit at 11% BJ	-----	BJ 17% (0-50kg constraint)
ZIRLO	BJ 17%	-----	-----	-----	-----	-----	-----	-----	-----
M5	BJ 17%	-----	-----	BJ ~13% for H≥ 200ppm	-----	-----	-----	-----	-----
E-110 E-110sponge	(*) BJ 17%	-----	BJ 17%	-----	-----	-----	-----	-----	BJ 17%

Ductility tests reported here were done by means of ring compression. The database consists mostly of tests done at RT. ANL has observed a marked ductility enhancement for tests done at 100°C and 135°C

(*) BJ 13% from Russian data and 8% from [14-17]

Main conclusions of the LOCA tests

CONCLUSIONS	EXPERIMENTAL BASIS
- <u>No burn-up effect</u> <i>Limit is 17% regardless of burn-up</i>	- Quench tests, 50 kg constraint, pre-hydriding to ~1000ppm (JAERI) - Quench tests, 50 kg constraints, high burn-up cladding (JAERI) <i>Note: Only Zr-4 has been tested under these conditions</i>
- <u>Moderate burn-up effect</u> <i>Limit is 11% BJ at high burn-up</i>	- High burn-up fuel, ANL HBR tests at 1200°C), without oxide cracking <i>Note: Only Zr-4 has been tested under these conditions</i>
Worst case - <u>Large burn-up effect</u> <i>Limit is 17%·[1-H/1000] BJ H in ppm, max. H=600ppm</i>	- Ductility tests, pre-hydrided Zr-4 samples (UJP, CEA, ANL) - Quench tests, full constraints, pre-hydriding to ~1000ppm (JAERI) <i>Note: Mostly Zr-4 data</i>

10. PROVISIONAL BURN-UP DEPENDENT CRITERION FOR Zr-4

In [22] an attempt was made in order to show how existing data could form the basis for a provisional burn-up dependent LOCA criterion. This was, among other things, aiming to provide an initial answer to a question posed by the NEA Committee on Nuclear Regulatory Activities (CNRA) to CSNI on the issue of burn-up dependent fuel criteria. The main points of the proposed approach are reviewed below.

Irradiation damage. There are indications that the irradiation damage as such does not have important effects on post-LOCA ductility. Consequently, an irradiation dose 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 above. They were obtained by ring compression tests and quench tests of non-irradiated, pre-hydrided samples. An important hydrogen effect is shown by ring compression tests and by quench tests with full constraint, up to a hydrogen content of 600ppm. Up to this level, the LOCA limit expressed in terms of ECR_{BJ} is reduced from 17 to ~7%, i.e. by a factor of 2.5. No further reduction is apparent for hydrogen concentration exceeding 600ppm. This means that the LOCA limit expressed as the ECR during the high temperature oxidation is:

$$ECR_{LIMIT, BJ} = 17\% \cdot [1 - H/1000] \quad (\text{Worst case. } H < 600\text{ppm, for higher content use } H = 600). \quad /5/$$

where H is in ppm. This should be taken as the worst case, compared with the more moderate burn-up effect derived from actual high burn-up fuel cladding (but without oxide cracking).

Wall thickness reduction. The pre-existing corrosion reduces somewhat the cladding resistance to LOCA because the metal wall thickness is reduced. This is accounted for by the factor $[1 - ECR_{BASE}/100]$ entering as multiplier in the previous equation, where ECR_{BASE} is the oxidised metal as percentage of wall thickness. This term is much weaker than the previous H pick-up term, and can be neglected in most cases.

As mentioned earlier, the ECR reduction can be expressed as a temperature limit reduction, i.e. (for e.g. $W = 0.576\text{mm}$):

$$T_{300s} = Q / (8.04 - \Delta_{ox}) - 273. \quad /6/$$

where $\Delta_{ox} = \ln [1 - H/1000]$. The value T_{300s} is the temperature corresponding to the time 300 seconds on the LOCA limit line. In other words, T_{300s} is the maximum tolerable temperature for a high temperature oxidation phase lasting for 300 seconds, and this tolerable temperature decreases with corrosion and Hydrogen content as given by Eq/6/.¹⁰ It can easily be shown that the amount of temperature decrease vs. H (vs. burn-up) is practically invariant for any type of LOCA transient.

The predictions given by Eq./5/ and /6/, which are based on current data represent a “worst case” for burn-up effect, are shown in Fig 13 and 14. The plots of Fig.13 relate to a relatively large

¹⁰ This would be for an ideal transient where the temperature would be brought to a high value and kept constant for a certain time period, such as in most ductility tests. There are means to “convert” from an actual LOCA transient, where temperature varies, to an equivalent, constant temperature transient, as explained in [22]. For any cladding thickness the term $(7.49 - \ln W)$ should be used in /6/ instead of the term 8.04

corrosion vs. burn-up (also shown in the figures) and to a relatively large hydrogen pick up fraction, i.e. 10ppm H/ μm oxide. By comparison, also the “total oxide” criterion – where the LOCA limit is set as: $\text{ECR}_{\text{LIMIT, BJ}} = 17\% - \text{ECR}_{\text{BASE}}$ is also plotted. As one can notice, the “worst case” results in an ECR decrease from 17 to 6% BJ at high burn-up (for large corrosion). This corresponds to a temperature limit decrease by $\sim 160^\circ\text{C}$, which mostly occurs at 30-50 MWd/kg, as the plot of Fig 13 shows. In other words, the “worst case” criterion would imply a $\sim 160^\circ\text{C}$ lower tolerable temperature at high burn-up compared with zero burn-up. The effect of corrosion is shown in Fig.14. Not surprisingly, the LOCA limit vs. burn-up does not decrease much when corrosion is small.

11. MAIN CONCLUSIONS

- On RIA there is a well-established testing method and a significant and relatively consistent database from NSRR and Cabri tests, especially on high burn-up Zr-2 and Zr-4 cladding. As observed in the CSNI-CNRA discussion and in this note, it is questionable whether other very old data can be considered representative for modern claddings. It is encouraging that several correlations have been proposed for the RIA fuel failure threshold. Their predictions are compared and discussed in this paper for a representative PWR case.
- On LOCA there are two different test methods, one based on ductility determinations and the other based on “integral” quench tests. The LOCA database at high burn-up is limited for both testing methods. Ductility tests carried out with pre-hydrided non-irradiated cladding show a pronounced hydrogen effect. Tests with actual high burn-up specimens - but without oxide cracking - show a moderate burn-up effect. However, this should be taken with caution, because recent separate effect tests done at CEA indicate that cooling rate can affect the results. Provisional burn-up dependent criteria are discussed in this paper. However, more high burn-up data is needed in order to arrive to firmer conclusions on a burn-up dependent LOCA limit.

Table 1. The CABRI Database, Main Parameters

Test and date	Rod and Burn-up	Pulse ms	Energy dep. cal/g	Corrosion μm	Results and observations
Na-1 (11/93)	GRA 5 64 MWd/kg	9.5	110 (at 0.4 s)	80 spalled	Brittle failure at $H_F = 30$ cal/g. Fuel dispersal (6 g)
Na-2 (06/94)	BR3 33 MWd/kg	9.1	211 (at 0.4 s)	4	No failure $H_{MAX} = 199$ cal/g Max. strain: 3.5%, FGR: 5.5%
Na-3 (10/94)	GRA 5 53 MWd/kg	9.5	120 (at 0.4 s)	40	No failure $H_{MAX} = 124$ cal/g Max. strain: 2% FGR: 13.7%
Na-4 (07/95)	GRA 5 62 MWd/kg	75	95 (at 1.2 s)	80 no spalling	No failure $H_{MAX} = 85$ cal/g Max. strain: 0.4% FGR: 8.3%
Na-5 (05/95)	GRA 5 64 MWd/kg	9.5	105 (at 0.4 s)	20	No failure $H_{MAX} = 108$ cal/g Max. strain: 1% FGR: 15.1%
Na 6 (03/96)	MOX 47 MWd/kg	35	125 at 0.66s 165 at 1.2 s	35	No failure $H_{MAX} = 133$ cal/g Max. strain: 3.2%, FGR: 21.6%
Na 7 (01/97)	MOX 55 MWd/kg	40	125 at 0.48s 175 at 1.20s	50	Failure at $H_F = 113$ cal/g Strong flow ejection
Na-8 (07/97)	GRA 5 60 MWd/kg	75	106 (at 0.4 s)	130 lim. spalling	Failure $H_F \leq 82$ cal/g, $H_{MAX} = 98$ cal/g, no fuel dispersal
Na-9 (04/97)	MOX 28 MWd/kg	34	197 at 0.5 s 241 at 1.2 s	< 20	No failure $H_{MAX} = 197$ cal/g Max. strain: 7.4%, FGR: ~34%
Na-10 (07/98)	GRA 5 62 MWd/kg	31	107 (at 1.2 s)	80 spalling	Failure at $H_F = 81$ cal/g, $H_{MAX} = 98$ cal/g, no fuel dispersal
Na-11 (06/00)	M5 63 MWd/kg	31	104	15	No failure $H_{MAX} = 93$ cal/g Max strain ~0.5%
Na-12 (12/00)	MOX 65 MWd/kg	62	106	80 no spalling	No failure $H_{MAX} = 103$ cal/g
CIP0-1 (11/02)	ZIRLO 75 MWd/kg	32	98	80 no spalling	No failure $H_{MAX} = 90$ cal/g
CIP0-2 (11/02)	M5 77 MWd/kg	28	89	20	No failure $H_{MAX} = 81$ cal/g

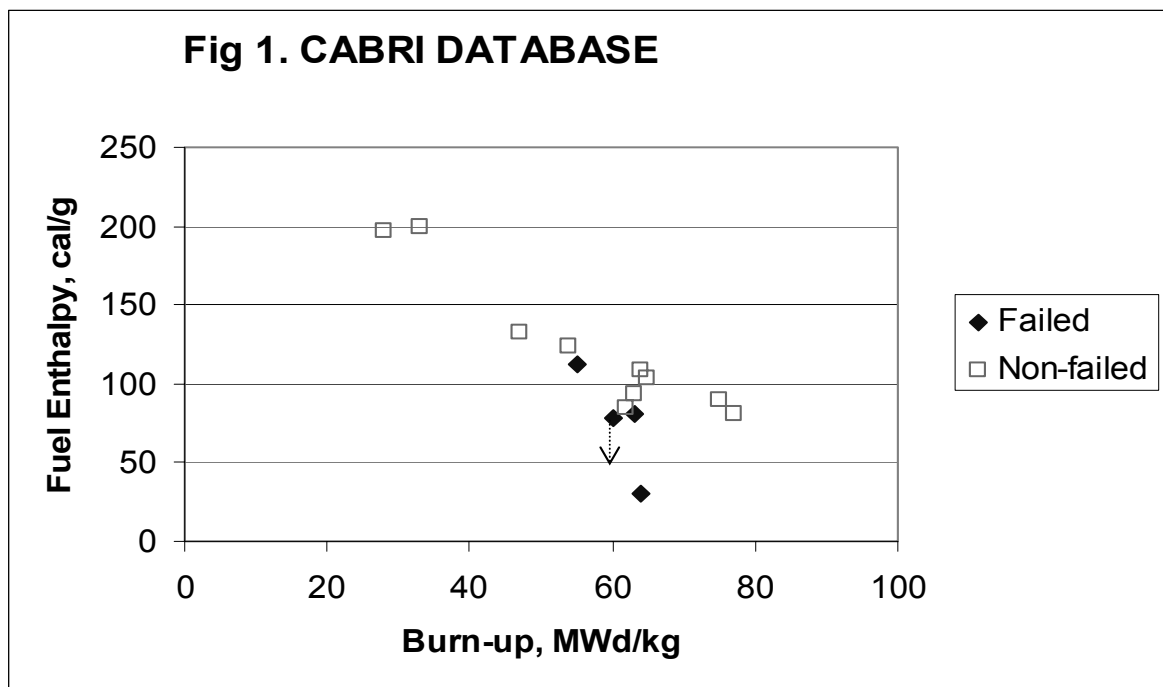


Table 2. The NSRR Database, Main Parameters¹¹

PWR fuel 1989-2003	Burn-up MWd/kg	Corrosion μm	Enthalpy cal/g	Failure enthalpy
MH-1	39	4	47	NF
MH-2	39	4	55	NF
MH-3	39	4	67	NF
GK-1	42	10	93	NF
GK-2	42	10	90	NF
OI-1	39	15	106	NF
OI-2	39	15	108	NF
HBO-1	50	43	73	60 cal/g
HBO-2	50	35	37	NF
HBO-3	50	23	74	NF
HBO-4	50	19	50	NF
HBO-5	44	60	80	77 cal/g
HBO-6	49	30	85	NF
HBO-7	49	45	88	NF
TK-1	38	7	126	NF
TK-2	48	35	107	60 cal/g
TK-3	50	10	99	NF
TK-4	50	15	98	NF
TK-5	48	20	101	NF
TK-6	38	15	125	NF
TK-7	50	30	95	86 cal/g
TK-8	50	10	65	NF
TK-9	50	10	99	NF
TK-10	46	10	86	NF
OI-10 ^{MDA}	60	27	104	NF
OI-11 ^{ZIRLO}	58	28	157	120 cal/g

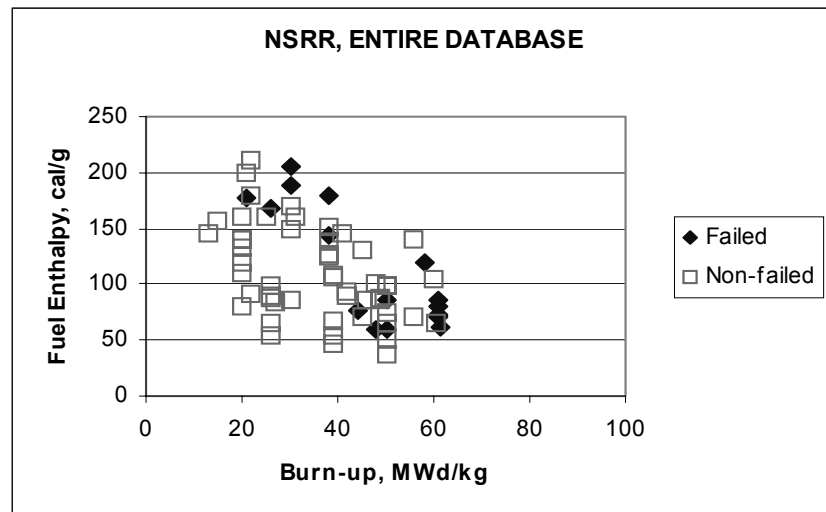
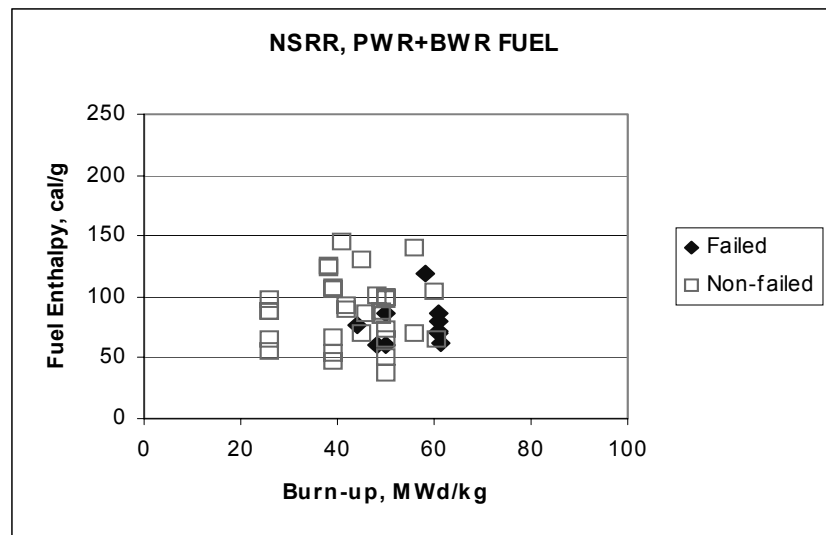
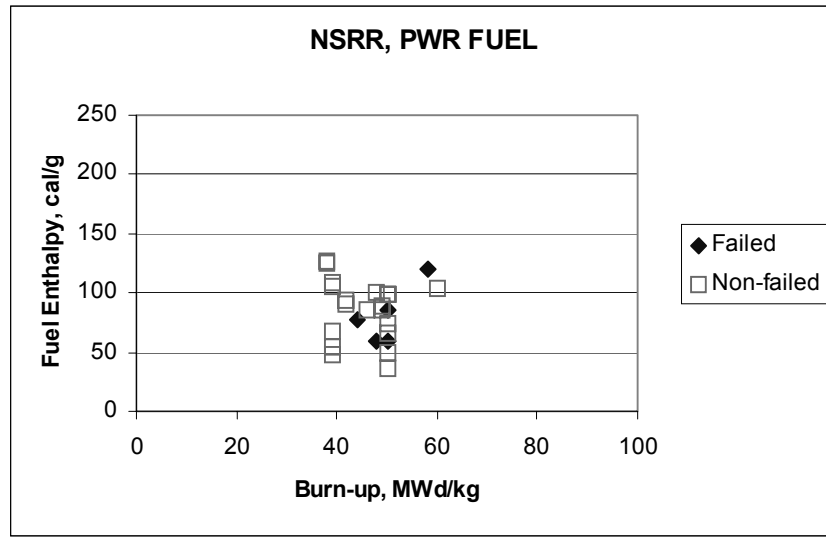
BWR fuel 1989-1998	Burn-up MWd/kg	Corrosion μm	Enthalpy cal/g	Failure enthalpy
TS-1	26	6	55	NF
TS-2	26	6	66	NF
TS-3	26	6	88	NF
TS-4	26	6	89	NF
TS-5	26	6	98	NF
FK-1	45	16	130	NF
FK-2	45	18	70	NF
FK-3	41	24	145	NF

JMTR fuel 1989-2003	Burn-up MWd/kg	Corrosion μm	Enthalpy cal/g	Failure (enthalpy)
JM-1	22	2	92	NF
JM-2	27	2	84	NF
JM-3	20	2	132	NF
JM-4	21	2	177	FAILURE
JM-5	26	2	167	FAILURE
JM-6	15	2	156	NF
JM-7	13	2	146	NF
JM-8	20	2	160	NF
JM-9	25	2	160	NF
JM-10	21	2	200	NF
JM-11	31	2	160	NF
JM-12	38	2	180	FAILURE
JM-13	38	2	150	NF
JM-14	38	2	160	144 cal/g
JM-15	30	2	150	NF
JM-16	38	2	140	NF
JMH-1 ⁽⁷⁾	22	2	160	NF
JMH-2	22	2	200	NF
JMH-3	30	2	220	205 cal/g
JMH-4	30	2	170	NF
JMH-5	30	2	220	189 cal/g
ATR-1	20	15	80	NF
ATR-2	20	15	110	NF
ATR-3	20	15	120	NF
ATR-4, -5	20	15	140	NF
ATR-6	30	-	85	NF

BWR fuel 1999-2002	Burn-up MWd/kg	Corrosion μm	Enthalpy cal/g	Failure enthalpy
FK-4	56	22	140	NF
FK-5	56	22	70	NF
FK-6	61	25	131	70 cal/g
FK-7	61	25	129	62 cal/g
FK-8	61	25	65	NF
FK-9	61	25	90	86 cal/g
FK-10	61	25	103	80 cal/g
FK-12	61	25	89	72 cal/g

¹¹ A fuel rod (JMN-1, 22 MWd/kg, 2 μm oxide) failed at 150 cal/g. This rod, however, had a reduced gap, 100 μm (as fabricated) instead of the normal design value of 190 μm like all other rods in the JM series

FIG. 2 NSRR DATABASE



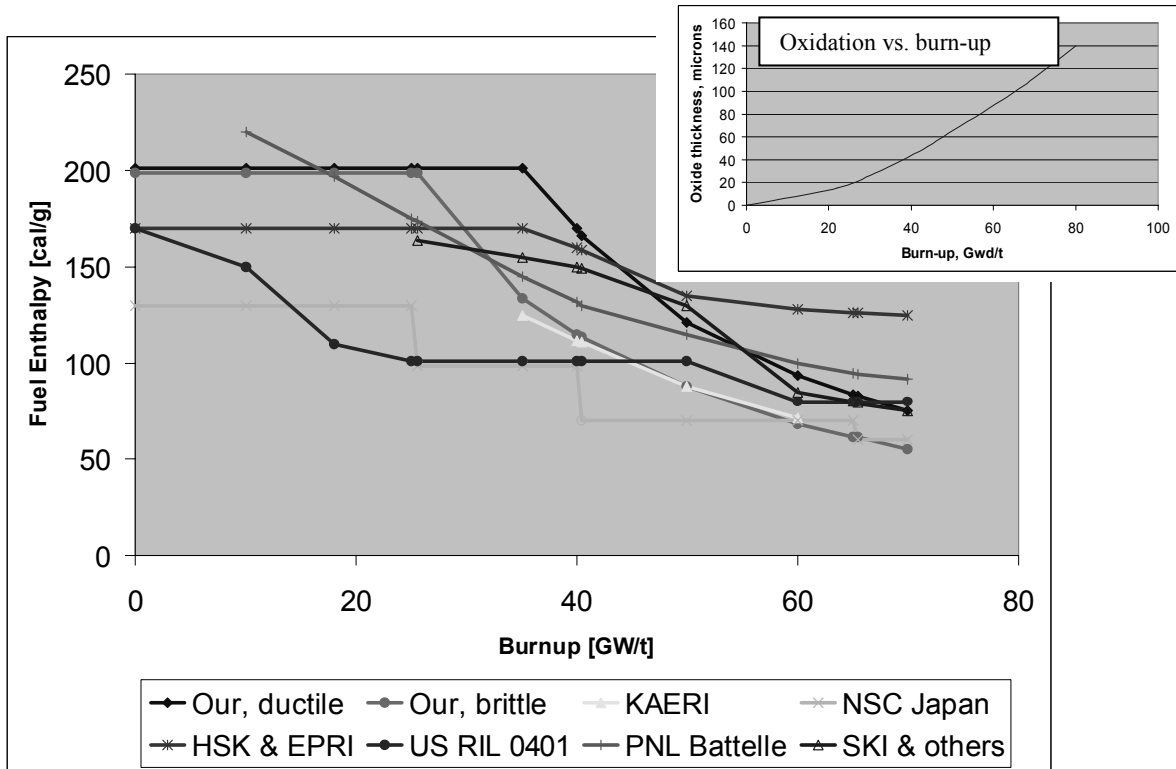


Fig.3. Comparison of various RIA failure thresholds for a PWR HZP case with relatively large oxidation. The assumed oxidation vs. burn-up is shown in the upper right figure.

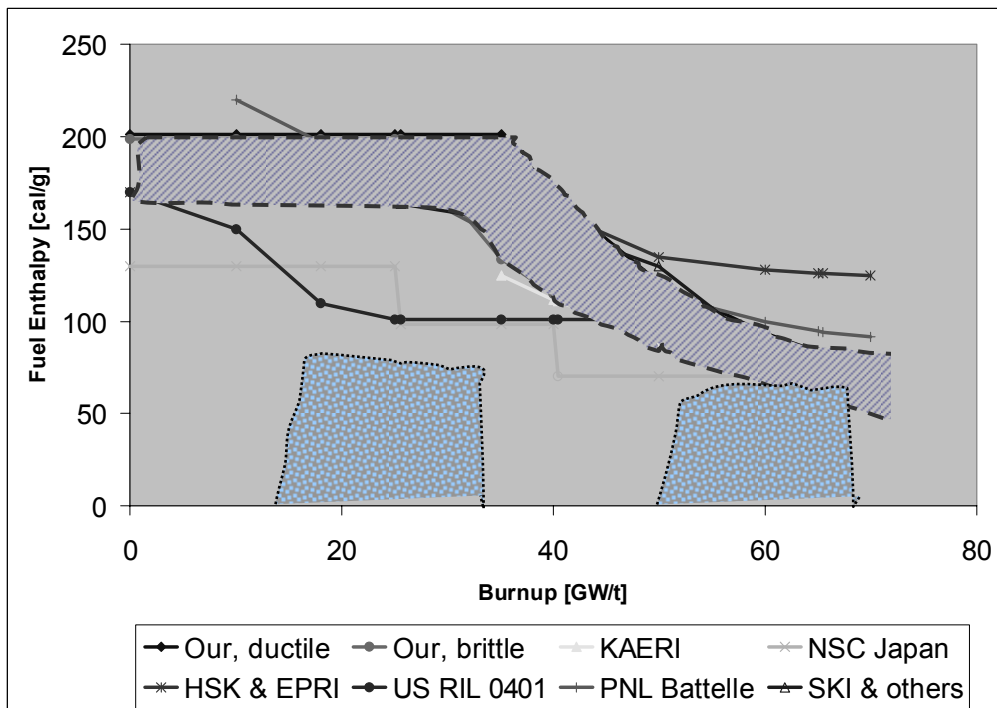


Fig.4. Most thresholds are within the upper shaded area. The two lower shaded zones represent the envelope of conservative calculations for actual PWR cores [12]. As seen, the core enthalpy can exceed the lower failure thresholds – i.e. for cases with large oxidation and brittle cladding.

Table 3. LOCA, HUNGARIAN DATABASE (AEKI) Ref. [14, 15]

No.	Zr-4 0.625mm					Zr 1%Nb 0.650mm				
	Temp.°C	Time s.	ECR [%]	H cont. [ppm]	$\Delta l/d$ [%]	Temp.°C	Time s.	ECR [%]	H cont. [ppm]	$\Delta l/d$ [%]
1	900	300	2.3	2.6	49.63	900	350	1.61	7.9	61.7
2	900	1000	3.5	2.3	40.7	900	1000	3.59	356	15.5
3	900	5000	5.9	1.6	15.4	900	3000	8.26	1325	3.8
4	900	11000	7	0.4	13.7	900	7000	13.11	2359	2.0
5	1000	87	2.9	1.2	45.34	900	11000	18.05	2896	2.3
6	1000	464	6	0.9	19.1	900	14000	18.58	2629	2.0
7	1000	2600	12.3	0.6	15.4	1000	100	1.94	1.3	54.2
8	1000	3300	15.2	8.0	5.83	1000	700	5.94	907	4.6
9	1000	4090	20.1	997.	4.33	1000	1200	8.97	1812	3.6
10	1000	7270	43.6	1854.	2.7	1000	1800	16.08	3135	2.0
11	1000	11360	77.3	110.	1.5	1000	3600	22.81	3274	1.6
12	1100	27	2.8	0.6	52.7	1000	6000	29.56	2330	1.6
13	1100	102	5.4	1.4	39.9	1100	19	1.56	17.6	56.7
14	1100	398	10.1	2.6	20.7	1100	133	4.55	598	3.5
15	1100	900	15.2	1.6	10	1100	704	11.16	907	3.5
16	1100	1500	19.5	2.1	6.4	1100	1500	16.62	678	2.58
17	1100	3000	26.8	5.5	4.65	1100	2400	21.58	704	2.6
18	1200	10	3.5	1.6	38.17	1100	5000	31.2	920	1.0
19	1200	40	5.8	0.7	17.07	1200	7	2.19	4.4	41.7
20	1200	163	10.5	0.9	8.46	1200	49	4.93	11.0	9.1
21	1200	367	15.4	1.4	5.42	1200	167	9.93	785	3.6
22	1200	790	21.9	4.9	3.75	1200	380	14.58	611	3.5
23	1200	1100	25.7	1.1	3.75	1200	646	19.19	549	3.1
24						1200	1205	26.41	580.3	2.36

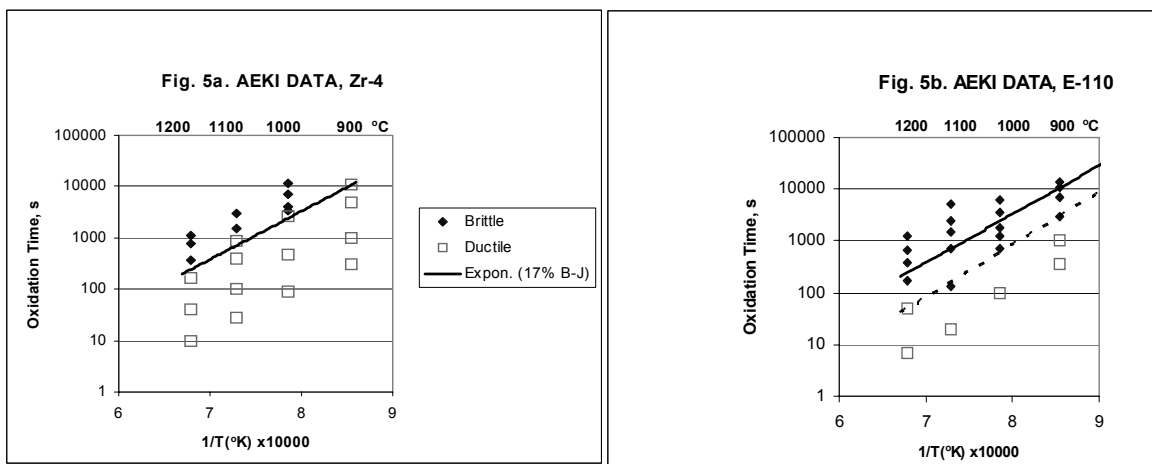


Fig.5. Temperature-time plot of the Hungarian data. The Zr-4 data fit with the BJ 17% criterion (solid line), whereas the E-110 limit is at BJ 8% (dashed line), i.e. at ~100°C lower temperature.

Table 4. CZECH DATABASE (UJP) Ref [16,17]

Zr-4 0.71 mm

Oxidation		Displacement, $\Delta L/D$, %		
Temp. °C	Time, sec.	As-received	2 μm oxide	10 μm oxide
1200	0	20.17	6.05	4.35
1200	90	20.17	5.74	4.66
1200	180	20.17	3.76	2.55
1200	270	9.93	3.29	1.98
1200	360	7.76	3.38	2.30
1200	600	5.12		
1200	720	3.41	3.58	1.87
1100	0		19.94	19.94
1100	90		19.94	1.5
1100	180	19.94	19.94	1.3
1100	360	19.94	5.7	0.4
1100	720	19.94	0.55	0.3
1100	1800	4.58	0.68	0
1100	3600	0.66	0.04	0.4
1100	180		6.46	3.1
1000	0	20.17	20.17	20.17
1000	360	17.07	20.17	20.17
1000	900	19.24	16.14	11.48
1000	1800	18.31	10.86	6.67
1000	1800		10.24	8.38
1000	3600	12.73	6.05	4.81
1000	7200	2.02	1.86	2.79
1000	10800	0.00	2.02	3.10
800	360	20.17	20.17	20.17
800	1800	20.17	20.17	20.17
800	3600	20.17	20.17	20.17
800	10800	20.17	20.17	20.17
800	18000		19.24	
800	25200	20.17	20.17	20.17

E-110 0.71mm

Displacement, $\Delta L/D$, %		
As-received	2 μm oxide	10 μm oxide
3.4	23.6	4.9
2.2	4.9	3.1
2.5	3.5	2.2
1.8	3.8	2.4
2.0	2.3	2.1
1.6		
2.0	1.4	1.1
	23.6	23.6
	23.6	0
23.6	23.6	0
23.6	0	0
0		0
0		0
0		0
23.6	23.6	23.6
2.9	23.6	23.6
4.5	23.6	13.4
3.8	22.3	3.4
	23.6	3.8
3.3	4.0	2.4
1.8	1.6	1.4
2.5	1.8	2.4
23.6	23.6	23.6
23.6	23.6	23.6
23.6	23.6	23.6
23.6	23.6	23.6
23.6	23.6	10.0
23.6	23.6	5.8

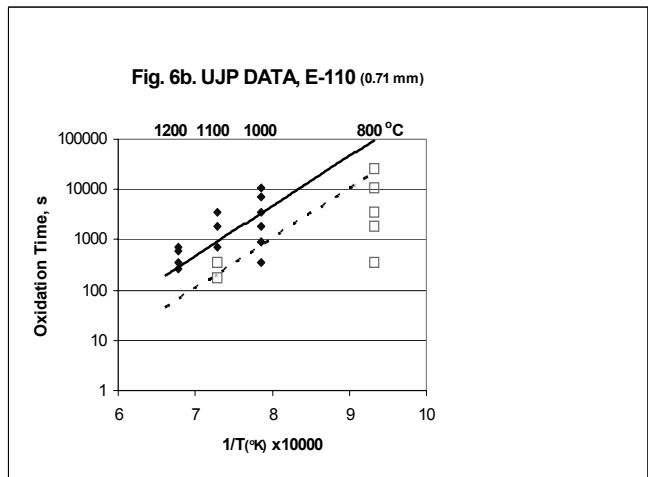
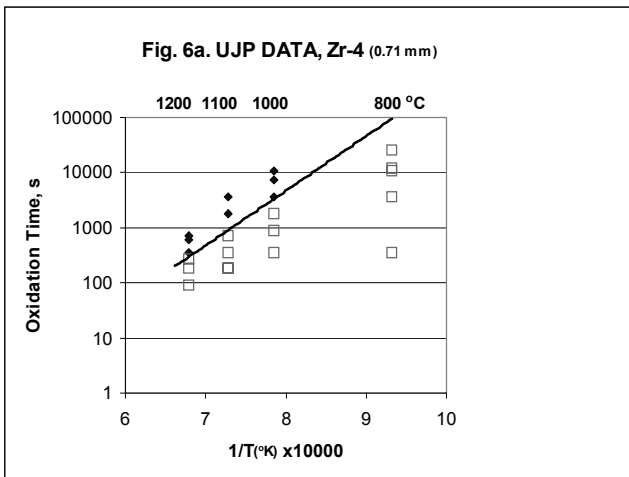


Fig. 6. Temperature-time plot of the Czech data. As for the AEKI data, the Zr-4 data fit with the BJ 17% criterion (solid line), whereas the E-110 limit is at BJ 8% (dashed line), or at ~100°C lower temperature.

Table 5. ANL DATA BASE Ref. [18-21] and [35]

(As-received samples, ring compression at RT. Ring compression temperature effect and pre-hydriding effect are summarised at the bottom of the table)

T, °C	Zr-4 0.57 mm		ZIRLO 0.57 mm		M5 0.61 mm	
	Time, s	Off. strain, %	Time, s	Off. strain, %	Time, s	Off. strain, %
1000	210	46	210	52	180	48
1000	840	29	840	25	780	51
1000	1900	7.5	1900	20	1700	40
1000	2430	5.1	2430	4.7	2100	19
1000	3360	3.2	3360	2.9	2980	3.2
1000			3560		3560	2.5
1100	67	58	67	52	60	57
1100	266	20	266	22	220	39
1100	600	5.4	600	5.1	520	7.5
1100	770	5	770	3.5	690	4.0
1100	1065	4.8	1065	3.3	930	1.8
1100	1100		1100		1100	3.2
1200	25	37	25	56	20	56
1200	65	4.0	65			
1200	100	1.0	100	2.8	85	3.4
1200	165	1.0	165	1.2	140	1.8
1200	240	0.8	240	1.9	190	1.7
1200	280	1.0	280	1.8	250	1.4
1200	400	0.5	400	1.2	350	2.0

- Significant ductility enhancement at 100-135C vs. RT, even for $ECR_{BJ} > 17\%$
- Pre-hydrided unirradiated 17x17 + 15x15 Zr-4 samples, oxidized at 1204C; ring-compression at 135C
 - Ductile-brittle transition at $ECR_{BJ} = 6.5\%$ for $H \geq 600\text{ppm}$ (15x15 Zr-4)
 - Ductile-brittle transition at $ECR_{BJ} = 9.5\%$ for $H \geq 400\text{ppm}$ (15x15 Zr-4)
 - Pre-hydrided 17x17 better than 15x15, but transition at $ECR_{BJ} < 13\%$ for $H \geq 300\text{ppm}$

Ref [35]

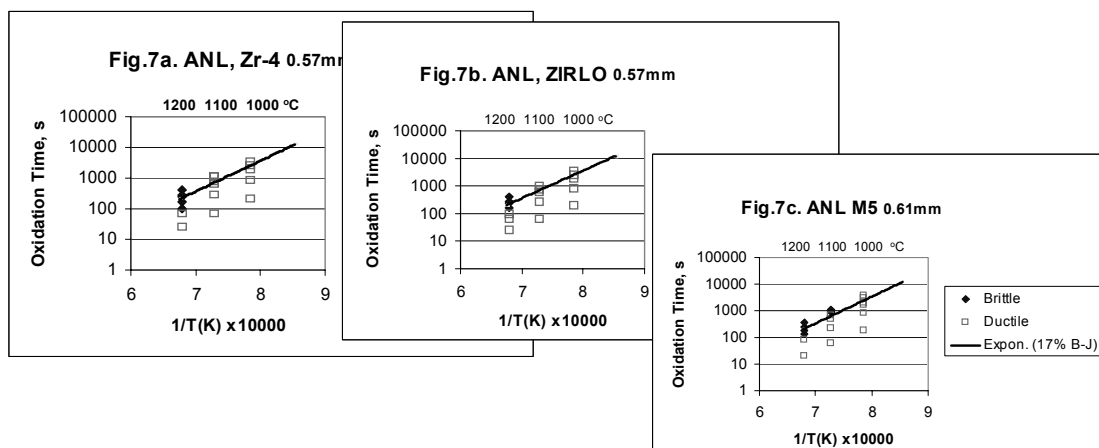


Fig.7. Argonne RT ductility data for Zr-4, ZIRLO and M5 cladding. The data generally fit with the 17% BJ limit (solid lines), but there is a tendency to lower ductility at 1200°C. However, ANL ring compression data at 135°C exhibit ductility beyond the 17% line also at 1200°C.

Ring Compression tests
(room temperature)
as-received vs. hydrided
materials, $T_{oxidation} = 1200^{\circ}C$

A marked decrease of the post-quench ductility is observed on hydrided materials, especially on Zy-4 + 600 ppm H even for the lower ECR value tested (i.e. for $ECR_{meas} \sim 3\% \Leftrightarrow BJ \sim 5\%$) while M5™ + 200 ppm H remains ductile for such an ECR

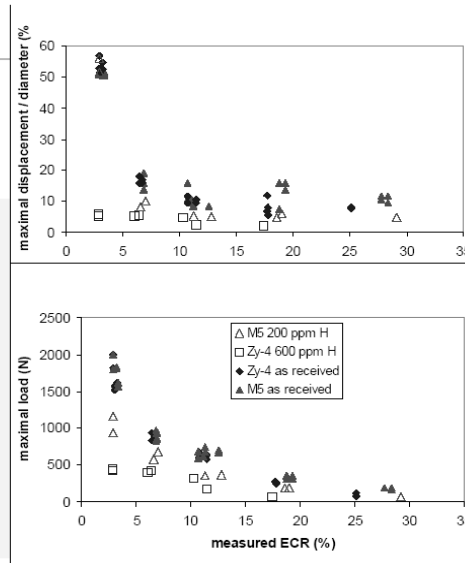


Fig.8. CEA data on ring compression tests of Zr-4 and M5 (RT) after steam oxidation at 1200°C [23]. The data show similar behaviour in terms of measured ECR (time not reported). The data also show a pronounced effect of pre-hydriding for both alloys.

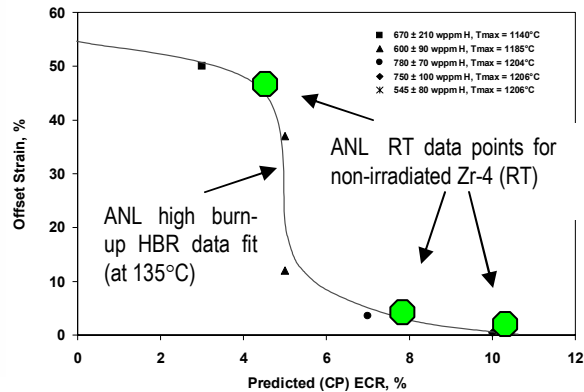
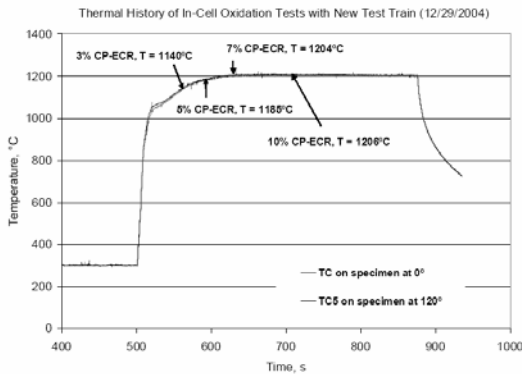


Table 6 Ref [24, 31]

SAMPLE BJ ECR, %	Equivalent temperature, °C	Equivalent time, seconds	Ductility at 350°C
4% [3%CP]	1100	48	Ductile
6.5% [5%CP]	1140	80	Ductile
6.5% [5%CP]	1140	80	Ductile
9% [7%CP]	1163	120 → 9% BJ	Ductile
13% [10%CP]	1193	180 → 13% BJ	Brittle

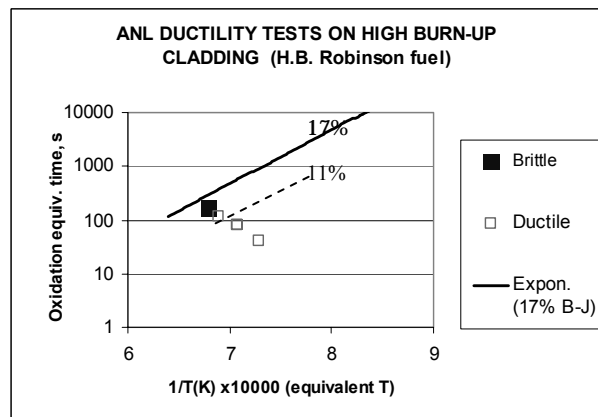


Fig.9. ANL ductility data from H.B Robinson fuel at high burn-up [24, 33]. The ECRBJ was calculated here from the temperature history (top left figure). Equivalent temperature and time give the same ECRBJ for a constant temperature transient. The results show a ductile-brittle transition somewhat below the 17% line, i.e. for a ECRBJ of ~11%. The upper right figure shows that the burn-up effect is comparable with the ductility change from 135 °C to RT. No oxide cracking is accounted for in these tests.

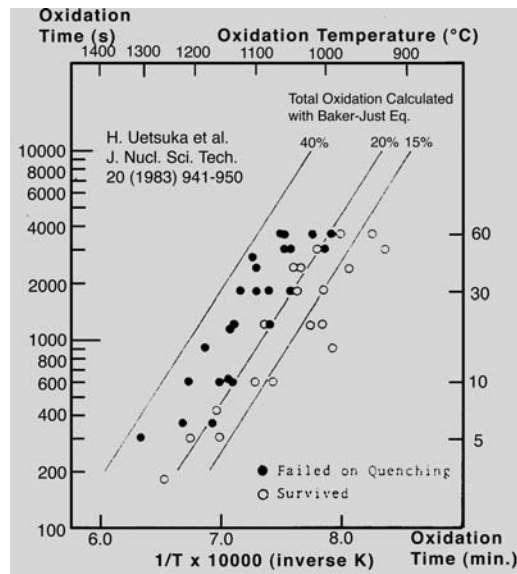


Fig.10. JAERI quench test database on as-received Zr-4 tubes subjected to high temperature oxidation and quench, under full constraint condition. Based on these data, the failure limit in Japan was set at ECRBJ=15% [25].

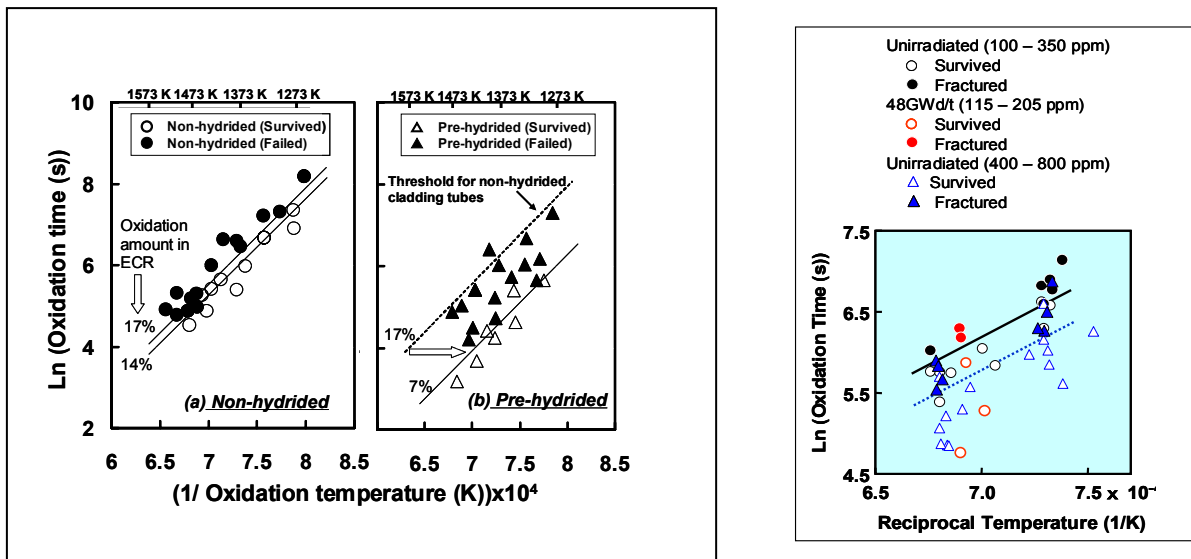


Fig.11. JAERI quench database on pre-hydrated cladding [26, 27]. The figure above is for full constraint, whereas that on the right hand side is for a 50 kg constraint. Under full constraint conditions, pre-hydrating causes a failure limit drop from 17 to 7% ECRBJ. The data for a 50 kg constraint, instead, show a significantly smaller pre-hydrating effect. (Note: the scale on the figure to the right is very sensitive and the 17% limit is below the two lines shown in the diagram).

Table 7. JAERI and ANL DATABASE: Quench tests with high burn-up cladding *Ref [27, 30]*
 (JAERI tests are with 50 kg constraint, whereas there was no constraints in the ANL tests)

Fuel Type	Burn-up, MWd/kg	Base Oxide thickness, μm	H content ppm	Oxid. Temper. $^{\circ}\text{C}$	Oxidation Time, s	Result	
PWR A3-1	44	20	170	1176	486	Failure	JAERI
PWR A1-2	44	25	210	1178	120	No failure	JAERI
PWR BL-3	39	18	140	1154	200	No failure	JAERI
PWR BI-3	41	18	140	1172	363	No failure	JAERI
PWR BI-5	41	15	120	1030	2195	No failure	JAERI
PWR BL-7	39	15	120	1177	543	Failure	JAERI
BWR	56	10	70	1204	300	No failure	ANL
BWR	56	10	70	1204	300	No failure	ANL

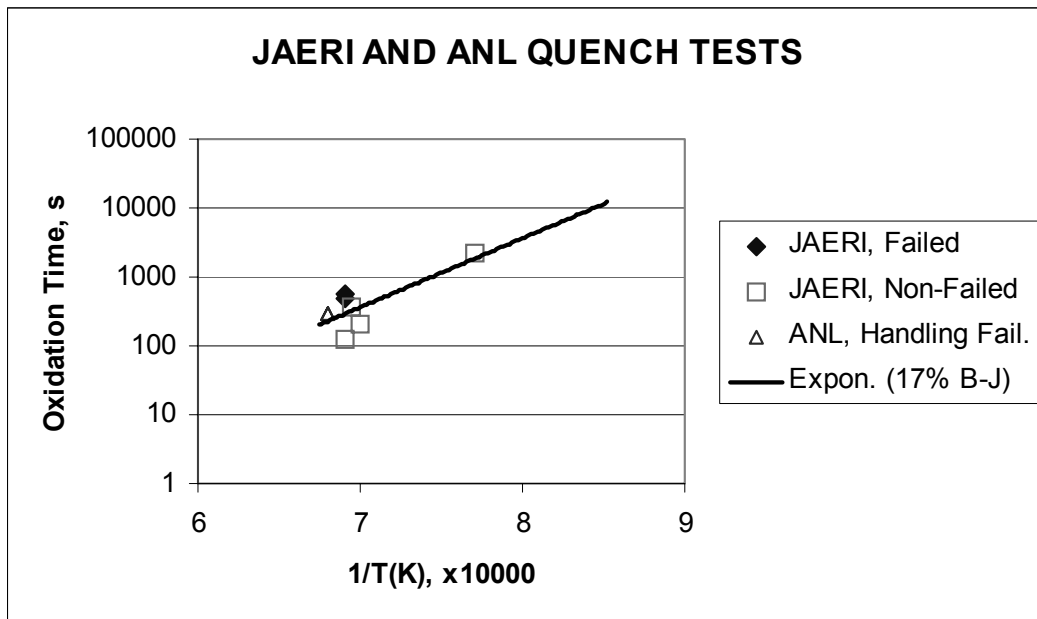


Fig.12. Plot of the JAERI and ANL data on quench tests of high burn-up cladding [27, 30]. The data fit well with the 17% ECRBJ limit. One should notice that there are two coincidental data points in the ANL case. The ANL tests were in fact not meant to provide a failure limit, and were run without constraints. In one case the sample failed during post-test handling. These results are shown here for completeness.

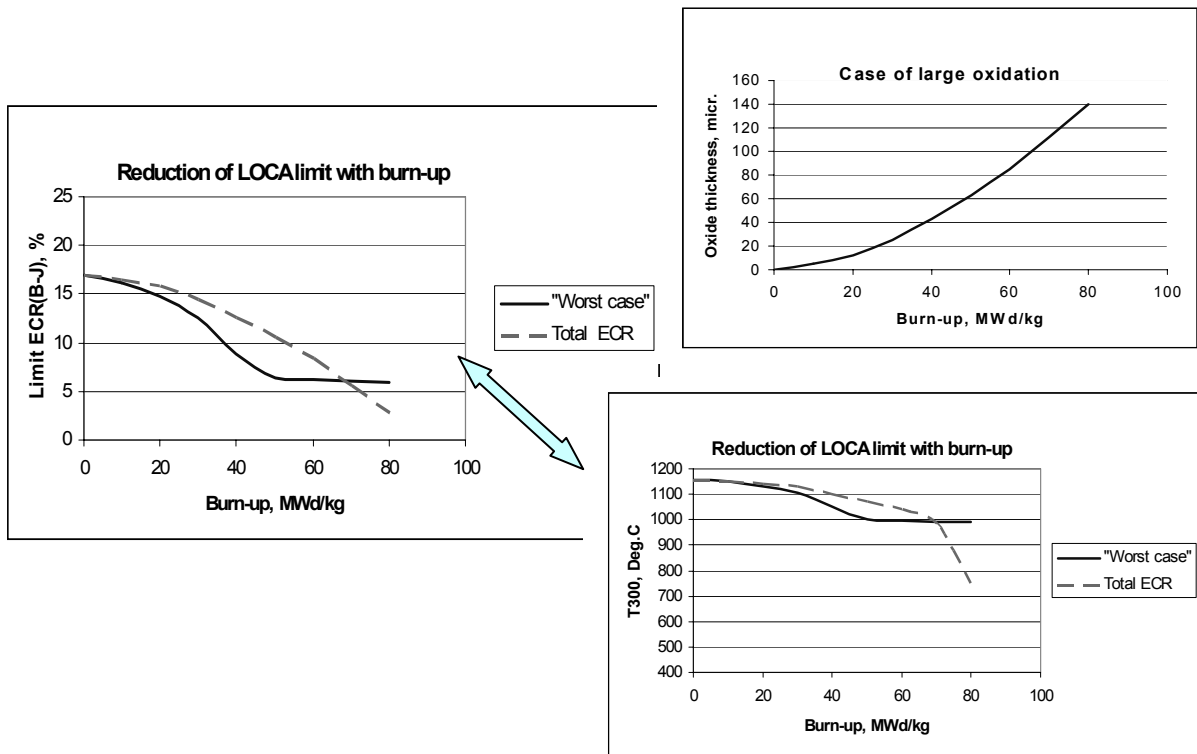


Fig.13. Calculated reduction of LOCA limit vs. burn-up in the “worst case” (Eq. /5/ and /6/). The limit is expressed in terms of ECR on the figure above, and as temperature limit (for 300s transient) on the right hand side figure. The reference corrosion vs. burn-up is given in the top-right figure. The “total ECR” criterion is also shown for comparison.

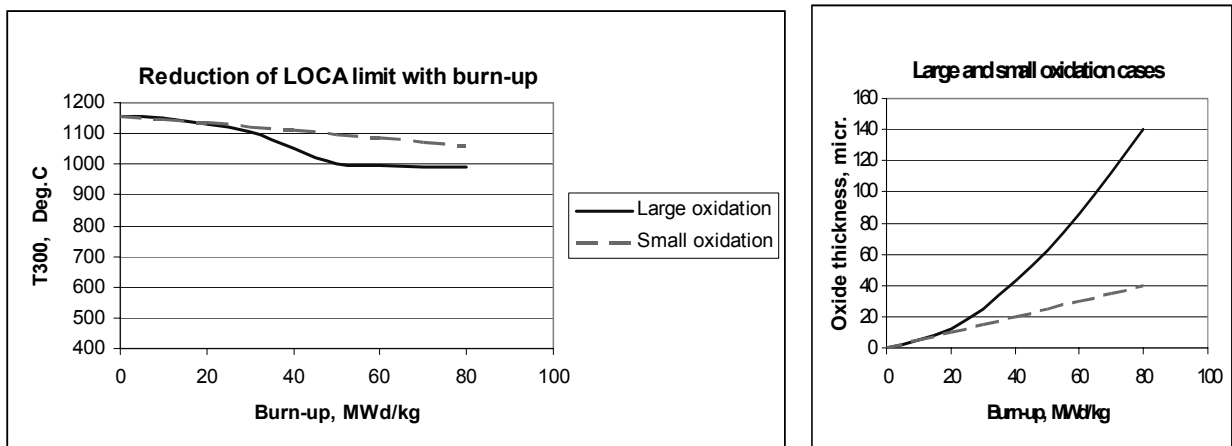


Fig.14. Comparison of the “worst case” burn-up effect (Eq/6/) for cladding with a large and small base irradiation corrosion. One can notice that the temperature limit decreases by ~160 °C for large corrosion, mostly in the 30-50 MWd/kg range. For low corrosion, the decrease is only ~60°C at 50 MWd/kg.

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