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NUCLEAR ENERGY AGENCY
COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS

Proceedings of the Topical Meeting on
LOCA Fuel Safety Criteria

Aix-en-Provence, 22-23 March, 2001

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The mission of the NEA is:

- to assist its Member countries in maintaining and further developing, through international co-operation, the scientific, technological and legal bases required for a safe, environmentally friendly and economical use of nuclear energy for peaceful purposes, as well as
- to provide authoritative assessments and to forge common understandings on key issues, as input to government decisions on nuclear energy policy and to broader OECD policy analyses in areas such as energy and sustainable development.

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In these and related tasks, the NEA works in close collaboration with the International Atomic Energy Agency in Vienna, with which it has a Co-operation Agreement, as well as with other international organisations in the nuclear field.

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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 meetings.

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.
COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS
Special Expert Group on Fuel Safety Margins

PROCEEDINGS

OF THE TOPICAL MEETING ON LOCA FUEL SAFETY CRITERIA
Aix-en-Provence, 22-23 March, 2001
# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Highlight</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Highlights of the meeting</td>
<td>9</td>
</tr>
<tr>
<td>Conclusions and Recommendation of the LOCA Topical Meeting</td>
<td>11</td>
</tr>
<tr>
<td>1. The Rationale of the LOCA 10CFR50.46b Criteria for Zircaloy and Comparison With E110 Alloy</td>
<td>15</td>
</tr>
<tr>
<td>G. Hache, IPSN, France</td>
<td></td>
</tr>
<tr>
<td>The History of LOCA Embrittlement Criteria</td>
<td>37</td>
</tr>
<tr>
<td>G. Hache, IPSN, France</td>
<td></td>
</tr>
<tr>
<td>H.M. Chung, Argonne National Laboratory, USA</td>
<td></td>
</tr>
<tr>
<td>2. NRC Program for Addressing Effects of High Burnup and Cladding Alloy on LOCA Safety Assessment</td>
<td>65</td>
</tr>
<tr>
<td>Ralph Meyer, US NRC</td>
<td></td>
</tr>
<tr>
<td>3. Ring-Compression Test Results and Experiments Supporting LOCA PCT, Oxidation and Channel Blockage Criteria</td>
<td>75</td>
</tr>
<tr>
<td>Laszló Maróti, AEKI, Budapest, Hungary</td>
<td></td>
</tr>
<tr>
<td>4. Justification of the M5\textsuperscript{TM} Behaviour in LOCA</td>
<td>105</td>
</tr>
<tr>
<td>A. Le Bourhis, Framatome, France</td>
<td></td>
</tr>
<tr>
<td>5. Ductility Testing of Zircaloy-4 and ZIRLO\textsuperscript{TM} Cladding After High Temperature Oxidation in Steam</td>
<td>135</td>
</tr>
<tr>
<td>W. J. Leech, Westinghouse Electric Co., USA</td>
<td></td>
</tr>
<tr>
<td>6. Progress in ANL/USNRC/EPRI Program on LOCA</td>
<td>145</td>
</tr>
<tr>
<td>Hee Chung, R.V. Strain, T. Bray, M.C. Billone, Argonne Nat. Lab., USA</td>
<td></td>
</tr>
<tr>
<td>7. Thermomechanical Properties of Oxydized Zirconium based Alloy Claddings in LOCA Conditions</td>
<td>173</td>
</tr>
<tr>
<td>8. Progress in JAERI Program on High Burnup Fuel Behaviour under a LOCA Transient</td>
<td>197</td>
</tr>
<tr>
<td>Hiroshi Uetsuka, F. Nagase, JAERI, Japan</td>
<td></td>
</tr>
<tr>
<td>9. Evaluation of Fuel Rod Axial Forces During LOCA Quench</td>
<td>209</td>
</tr>
<tr>
<td>Nicolas Waeckel, Patrick Jacques, EDF SEPTEN</td>
<td></td>
</tr>
<tr>
<td>Rosa Yang, Robert Montgomery, EPRI</td>
<td></td>
</tr>
<tr>
<td>10. Synthesis of an EDF and FRAMATOME ANP Analysis on Fuel Relocation Impact in Large Break LOCA</td>
<td>231</td>
</tr>
<tr>
<td>Michel Lambert, Yann Le Hénaff, EDF/SEPTEN</td>
<td></td>
</tr>
<tr>
<td>Jean-Luc Gandrille, Framatome, ANP, France</td>
<td></td>
</tr>
</tbody>
</table>
11. High Burnup UO₂ Fuel LOCA Calculations to Evaluate the Possible Impact of Fuel Relocation After Burst
   C. Grandjean, G. Hache, C. Rongier
   Institut de Protection et de Sûreté Nucléaire
   CEN Cadarache, France

12. High Burnup Phenomena Affecting the Failure Mode of Fuel Rods During LOCA
   Hiroshi Hayashi, NUPEC, Japan

13. Results of the Experimental Research on High Burnup VVER-type Fuel Behaviour in LOCA Conditions
   Smirnov V.P. et al, RIAR, Russian Federation

14. IPSN Analysis of Experimental Needs Requested for Solving Pending LOCA Issues
   A. Mailliat, IPSN, France

List of Participants
HIGHLIGHTS OF THE MEETING

A topical meeting on LOCA Fuel Safety Criteria was held in Aix-en-Provence on 22-23 March, 2001. It was organised under the auspices of the CSNI and its Special Expert Group on Fuel Safety Margins in cooperation with IPSN Cadarache. The meeting was chaired by Dr. Georges Hache from IPSN.

In total 53 participants attended. Research and industry organisations from France, Hungary, Japan, the Russian Federation and the USA presented 14 papers in all. The papers covered three main issues: post-quench ductility; the impact of axial constraint; and fuel relocation.

Papers presented:

G. Hache, IPSN, France
The Rationale of the LOCA 10CFR50.46b Criteria for Zircaloy and Comparison With E110 Alloy

Ralph Meyer, US NRC
NRC Program for Addressing Effects of High Burnup and Cladding Alloy on LOCA Safety Assessment

Laszló Marótí, AEKI, Budapest, Hungary
Ring-Compression Test Results and Experiments Supporting LOCA PCT, Oxidation and Channel Blockage Criteria.

A. Le Bourhis, Framatome, France
Justification of the M5 behavior in LOCA

W.J. Leech, Westinghouse, USA
Westinghouse Ductility Testing of Zircaloy-4 and ZIRLO Cladding After High Temperature Oxidation in Steam

Hee Chung, R.V. Strain, T. Bray, M.C. Billone, Argonne Nat. Lab., USA
Progress in ANL/USNRC/EPRI Program on LOCA

Yu. K. Bibilashvili, N.B. Sokolov, L.N. Andreeva-Andrievskaya, at al. VNIINM, Russian Federation
Thermomechanical Properties of Oxydized Zirconium based Alloy Claddings in LOCA Conditions

Nicolas Waeckel, Patrick Jacques, EDF, France
Rosa Yang, Robert Montgomery EPRI, USA
Analysis of Fuel Rod Axial Forces During LOCA Quench

Hiroshi UETSUKA, F. Nagase, JAERI Japan
Progress in JAERI Program on High Burnup Fuel Behavior under a LOCA Transient
The LOCA Topical Meeting was closed by the discussion of Conclusions and Recommendations.
Conclusions and Recommendation of the LOCA Topical Meeting

Introduction

Several years ago, the CSNI issued a mandate to verify, and if necessary provide the rational for adjustment of, current fuel safety limits in view of 'new' design elements. These are changes in fuel design and operation, such as new cladding materials and optimized core loading strategies, which have been made in order to support better fuel utilization including, in particular, high discharge exposures. Fuel safety limits have not always been updated in concord with these changes, and there is a general feeling that safety limits, especially those related to DBA's, are no longer adequate and that significant safety margin has been lost. CNRA raised a number of questions to CSNI (December 2000) basically addressing the same concerns.

The CSNI Task Force on Fuel safety Criteria (TFFSC) responded to the above mandate; in its report (NEA/CSNI/R(99)25 of July 2000) the TFFSC issued a number of conclusions and recommendations, that constitute the basis for the work of the CSNI Special Expert Group (SEG) on Fuel Safety Margins. In essence, each safety criterion was reviewed against today's fuel characteristics; it was attempted to evaluate any (experimental & analytical) research needed to verify - or possibly adjust - the criterion, and to subsequently identify such research that is still missing. Naturally, not each and every detail was investigated by the TFFSC; the SEG therefore has the mandate to go into the necessary detail on the important issues raised by the TFFSC.

Accordingly, this topical meeting aimed at providing a forum for detailed discussions on the issues related to LOCA limits. The LOCA - issues identified in the said TFFSC report are the following:

a. Effect of pre-transient oxidation
b. Quenching & post quenching effects, changes in oxidation rate and embrittlement
c. Ballooning and fuel relocation
d. Beta-stabilizing effects during ballooning & burst
e. Subchannel blockage
f. Radiological consequences (extent of burst, release of fission products)
g. Modeling adequacy & accuracy.

These issues provided the perspective for the presentations and discussions, which will be summarized in the text below.

Post Quench Ductility

Presentations were made on the history of the ECR and PCT criteria (Hache, Chung) and on results of quench tests and post-quench ductility tests on hydried zircaloy (Uetsuka) and E110 (Maroti, Andreeva-Andrievskaya, Smirnov), M5 (Lebourhis) and Zirlo (Leech) alloys.

At the end of the ECCS Rule-Making Hearing (Docket RM-50-1, 1973), the ECR and PCT criteria were based on retention of ductility at 275°F (135°C, the saturation temperature during reflood) according to slow ring-compression tests of non-ballooned samples (Hobson, Salt Lake City, 1973); the selection of the 17% ECR value was specific to the use of the Baker-Just correlation. Some non-OECD countries are using another methodology (Andreeva-Andrievskaya).

During the late 70's – early 80's, the performance of slow ring-compression tests of ballooned and bursted samples (Chung, NUREG/CR-1344; Uetsuka, J. Nucl. Sci. Tech. 18(1981) and 19(1982)) showed that the 1973 criteria failed to ensure retention of ductility at 135°C in narrow local regions near burst opening, where H content exceeds about 600 ppm. This phenomenon was not known in 1973. However, the 1973
criteria still ensure resistance to 0.3 J impact tests (Chung, NUREG/CR-1344) and survival after fully constrained quench tests (Uetsuka, J. Nucl. Sci. Tech. 20(1983)) for low-burnup Zircaloy. Following these results, Japan modified the basis of its ECR criterion to ensure survival after fully constrained quench tests.

For high-burnup zircaloy, if irradiation-induced H uptake exceeds about 600 ppm, the 17% ECR criterion may fail to ensure retention of ductility at 135°C, nor survival after fully constrained quench tests, if it is applied to transient oxidation alone, while it is sufficient to ensure survival after unconstrained quench tests (Uetsuka). The participants think that there is a need of better understanding of combined effects of H and O uptakes on post-quench ductility of high burnup zircaloy.


First results presented by FRAMATOME (at 1100°C oxidation temperature) and WESTINGHOUSE (preliminary results) show similar behaviour to zircaloy 4 rather than to previously published results on E110 alloy (the differences with these last results are not well understood). The participants think that there is a need to better understand the combined effects of Nb addition and H and O uptakes on post-quench ductility, in particular to provide knowledge able to support the verification of the PCT criterion for these alloys.

The type of post-quench ductility test should be defined.

Presentations on E110 alloy showed no big difference between fresh and irradiated results up to 50 GWD/t.

Effects of axial restraints

The presentation of work at JAERI (Uetsuka) also covered quench tests with and without axial restraints. Failure levels were around 60% ECR without restraint, but were reduced to about 20% ECR with restraint. Pre-charging with hydrogen further reduced the ECR at failure. Axial forces were quite strong, however, at about 1500 N, and the presentation suggested that half this force might be more appropriate. Analysis by EPRI and EDF (Waackel) showed that axial forces should only be in the range of 200-300 N, far below the level in the JAERI tests. During the discussion, a question was raised as to whether internal forces from pellet-cladding bonding might be more important than the external forces modeled by EPRI. This question was not resolved, but questions of this type were the reason that retention of ductility was originally used as the basis for embrittlement criteria rather than quench test results.

Fuel Relocation

Presentations by IPSN and EDF/Framatome dealt with the issue of axial fuel relocation (slumping) into the ballooning area. The phenomenon has been observed in various experiments, but a possible impact on the LOCA behaviour is not directly considered in present safety regulations.

The fuel relocation increases the local power and reduces the pellet-clad gap, thus changing the heat transfer. Modified versions of the CATHARE code were used to analyse the consequences for the LOCA
transient. Assumptions made for the analysis included filling ration (40 – 60% based on actual experimental observations), particle geometry, and modelling of the conductivity using a debris bed model.

The IPSN results indicated the possibility of a significant effect on cladding temperature during the late phase of the transient, although considerable margins to both fuel and cladding melting remain. The consequences were believed to be more severe for low burnup UO2 and high burnup MOX fuel. IPSN is preparing a testing programme to address the issue.

In contrast, the EDF/Framatome evaluation concluded that the effect of relocation on cladding temperature is weak. Since the consequences are covered by conservative assumptions for more limiting effects, it is believed that it is not necessary to take the phenomenon into account in safety regulation.
1. The Rationale of the LOCA 10CFR50.46b Criteria for Zircaloy and Comparison With E110 Alloy

G. Hache, IPSN, France

Paper summary

One of primary factors that exacerbate the susceptibility of oxidized cladding to post-quench embrittlement is an hydrogen uptake which may occur during irradiation (e.g., in high-burnup Zircaloy-4) or during transient oxidation in steam (e.g., from cladding inner surface in contact with stagnant steam near a ballooned and burst region). For cooling rates typical of bottom flooding of core most hydrogen atoms remain in solution in the beta phase at Leidenfrost temperature, and in such state, hydrogen has little effect on the fracture resistance of an oxidized Zircaloy. However, when load is imposed at temperatures below the Leidenfrost temperature, precipitated hydrides strongly influence the fracture resistance of cladding.

To correctly interpret the results of ongoing investigations on the performance of the old and new types of fuel cladding, especially at high burnup, it is necessary to accurately understand the history and relevant databases of current LOCA embrittlement criteria. For this purpose, documented records of the 1973 U.S Atomic Energy Commission (AEC) Emergency Core Cooling System (ECCS) Rule-Making Hearing were carefully examined to clarify the rationale and data bases used to establish the 1204°C peak cladding temperature and 17% maximum oxidation limits.

In the 1973 Rule-Making Hearing, the U.S. Atomic Energy Commission (AEC) staff and commissioners were clearly reluctant to neglect the effect of mechanical constraints on the susceptibility of oxidized fuel cladding to thermal-shock fragmentation. Subsequent constraint-quench test by JAERI and Phoebus LOCA 219 bundle test justified this rationale.

The AEC staff and commissioners were of the opinion that retention of ductility was the best guarantee against potential fragmentation of fuel cladding under various types of not-so-well-quantified loading, such as thermal shock, hydraulic, and seismic forces, and the forces related with handling and transportation.

Primary rationale of the 17% oxidation criterion was retention of cladding ductility at temperatures higher than 275°F (135°C), i.e., the saturation temperature during reflood. The threshold equivalent cladding reacted (ECR) of 17% is tied with the use of Baker-just correlation. If a best-estimate correlation other than Baker-Just equation (e.g., Cathcart-Pawel correlation) had been used, the threshold ECR would have been <17%.

Investigations conducted after the 1973 Rule-Making Hearing show that for oxidation temperatures ≤1204°C, the 17% oxidation limit (calculated with Baker-Just correlation) is adequate to ensure survival of unconstrained or fully constrained cladding under quenching thermal shock. It was also shown that the 17% limit (ECR determined on the basis of measured phase layer thickness) is adequate to ensure retention of ductility and resistance to 0.3-J impact failure in non-irradiated non-ruptured two-side-oxidized Zircaloy cladding in which hydrogen uptake during a LOCA-like transient is small.

However, the 17% ECR limit appears to be inadequate to ensure post-quench ductility at hydrogen concentrations > 700 wppm. A major finding from tests performed after the 1973 Rule-Making Hearing shows that post-quench ductility is strongly influenced by not only oxidation but also hydrogen uptake. It seems that this effect of large hydrogen uptake was not known at the time of 1973 Hearing.

The 1204°C peak cladding temperature (PCT) limit was selected on the basis of slow-ring-compression tests that were performed at 25-150°C. Samples oxidized at 1315°C were far more brittle than samples
oxidized at 1204°C in spite of comparable level of total oxidation. This is because oxygen solid-solution hardening of the prior-beta phase is excessive at oxygen concentrations >0.7wt%. Consideration of potential for runaway oxidation was a secondary factor in selecting the 1204°C limit. The 1204°C limit was subsequently justified by the observations from impact tests and handling failure of fuel rods exposed to high temperatures in the Power Burst Facility. The 1204°C PCT and the 17% ECR limits are inseparable, and as such, constitute an integral criterion.

Post-quench ductility and toughness are determined primarily by the thickness and the mechanical properties of transformed-beta layer. The mechanical properties are strongly influenced by several factors such as: oxygen solubility in beta, concentrations of alpha-(tin and oxygen) and beta-stabilizing elements (niobium and hydrogen), the nature of beta-to-alpha-prime transformation, redistribution of oxygen, niobium, and hydrogen during the transformation, and precipitation of hydrides. Significantly large hydrogen uptake can occur in some types of fuel cladding, during normal operation to high burnup, during breakaway oxidation at <1120°C, and, for localized regions near a rupture opening, during LOCA transients. Hydrogen uptake and its effect on the properties of transformed beta could differ significantly in Zircaloy and in niobium-containing alloys, such as Russian E110 or M5 (Framatome) or ZIRLO (Westinghouse).

Steam oxidation results for E110 published in early nineties by Böhmert (Germany) and Vrtilkova et al. (Czech republic) showed much higher hydrogen content, especially below 1100 °C. This higher hydrogen content is likely the cause why the residual plasticity of E 110 alloy decreases much more rapidly (at 6% oxide thickness) than that of Zircaloy (17%). Thus the application of the 1973 ECCS Rule-making hearing methodology for E 110 alloy would lead to a limit of 6% ECR associated with the use of the conservative Bochvar's Institute correlation. The question how is it with other Nb containing alloys (M5, ZIRLO) remains opened. It is therefore recommended to obtain a better understanding of the effects of more realistic hydrogen uptake and niobium addition on the properties of transformed-beta layer and post-quench ductility.
Rationale of the LOCA 10 CFR 50.46b Criteria for Zircaloy and Comparison with E 110 Alloy

G. Hache, IPSN, France

CONTENTS

Embrittlement phenomena
Opinion of the regulatory staff and Commissioners
Selection of the 17% ECR criterion
H uptake
Impact tests - constrained quench tests
Selection of the 2200° F PCT criterion
Recapitulation
High burnup Zircaloy
Transposition to E 110 alloy
Other Nb containing alloys
EMBRITTLEMENT PHENOMENA (2)

- Hydrogen Pick-up in the β-Zr Layer:
  - either during irradiation (high-burnup Zircaloy-4...)
  - or during high-temperature steam oxidation (stagnant steam in the ballooned and burst region...).

  ➔ at the Leidenfrost temperature, direct effect on fracture resistance is insignificant (H in solution in the β phase during thermal shock).
  ➔ during post-quench phase, precipitation of hydrides (δ-ZrH₂) strongly influences the mechanical properties.

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OPINION OF THE REGULATORY STAFF AND COMMISSIONERS (1)
(ECCS Rule - Making Hearing, 1973)

- Reluctance to neglect the effect of mechanical constraints on thermal-shock fragmentation
  - rod-rod interaction due to ballooning or bowing
  - rod-grid interaction due to differential shrinkage between fuel rods and guide tubes

- Justified later by JAERI constraint-quench tests and Phebus LOCA-219 bundle test

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Failure map for Zircaloy-4 cladding by thermal shock under no constraint condition relative to duration and temperature of isothermal oxidation after rupture in steam.
Retention of ductility is the best guarantee against potential fragmentation under various types of loadings (thermal shock, hydraulic, seismic forces).

Results from unconfined quench tests (simple thermal-shock test) were considered only corroborative and reassuring.

Later studies showed a large margin compared to 17% - ECR and 2200°F - PCT criteria.

- 17% - ECR and 2200°F - PCT criteria are based on results from post-quench ductility tests (Hobson's slow-ringing compression tests).

Rod 18: Temperature higher than 1204 °C, fragmented despite 16% ECR

- bundle effect (mechanical constraints)
- necessity of a PCT criterion
SELECTION OF THE 17%-ECR CRITERION  
(Multistep Process)

- Primary criterion: zero ductility temperature (ZDT) \( \leq 275°F \) 
  (135°C, the saturation temperature during reflood)

- According to Hobson's slow-ring-compression tests, 
  ZDT \( \leq 275°F \) is equivalent to \( \xi/w \leq 0.44 \):
  \[ \xi = \text{combined thickness of oxide and } \alpha-\text{Zr(O) layers} \]
  \[ w = \text{thickness before oxidation.} \]

- 17% ECR was derived in association with the use of the 
  Baker-Just correlation.

- For peak-cladding temperature (PCT) \( > 1204°C \), 17% ECR 
  criterion coincides with simple thermal-shock failure 
  boundary.
Specimen ductility as a function of deformation temperature and fraction of as-oxidized wall thickness ($F_{wa}$) consisting of transformed $\beta$ phase for the low-strain-rate tests.

Oxygen Distribution across Cladding Wall

- Resistance to thermal-shock during reflood and post-quench ductility and fracture toughness are largely determined by the thickness of and the oxygen concentration in the prior-beta layer.
Correlation between distributions of inner surface oxide layer thickness, ring compression deflection and absorbed hydrogen content.

- Effect of internal oxidation by stagnant steam in ballooned and burst region:
  - Large H uptake in the necks (not known in 1973)

- 17% ECR even with Baker-Just correlation doesn't ensure ZDT ≤ 135 °C at high H concentrations such as in the necks.

Critical hydrogen concentration at 100 °C ~ 550 ppm.

Major finding after 1973: post quench ductility strongly influenced by not only oxidation but also by H uptake.
After 1973 it was shown that 17\% ECR with Baker/Just correlation is consistent with resistance to 0.3 J impact tests and survival after fully constrained thermal shock tests, despite high H uptake in the neck of the ballooned and burst region.

"Ambient impact of 0.3 J (was) thought to be a reasonable approximation to post - LOCA quench ambient impact loads" (CSNI 129, December 1986)

Failure Characteristics of Zircaloy-4 Cladding under 0.3-J Impact Load at 300 K Relative to Equivalent-cladding-reacted Parameter and Oxidation Temperature. The ECR parameter was calculated based on observed phase layer thicknesses.
SELECTION OF THE 2200°F (1204°C) PEAK CLAD TEMPERATURE (PCT) CRITERION

- Based on Hobson's slow-ring-compression tests at 77-300°F (25-149°C)
- Samples oxidised at 2400°F were far more brittle than samples oxidised below 2200°F, in spite of comparable level of total oxidation.
- Proposed mechanism: O solid-solution hardening of previous-β phase at O concentration > 0.7 wt.%
- Final selection by regulatory staff and commissioners: PCT ≤ 2200°F (1204°C)
Later justified by ANL 0.3-J pendulum-impact tests and PBF handling-failure data; these results take into account of H pick-up during stagnant steam oxidation in the burst region (all ANL but only 2 PBF rods data).

Threshold O concentration of 0.7 wt.% was a key parameter used later at ORNL, AECL and ANL to propose several embrittlement criteria.

Consideration of potential for runaway oxidation would have lead to a PCT value > 2200°F.
RECAPITULATION

17% ECR + Baker - Just + 2200 °F PCT were chosen in 1973 to ensure ZDT ≤ 135 °C where wall thinning, power, temperature and oxidation are high (β thickness is low) - however H uptake is low.

Accordingly to post 1973 knowledge, 17% ECR + Baker - Just + 2200 °F PCT ensure global resistance to 0.3 J impact tests and global survival to fully constrained thermal shock tests (higher β thickness in the necks, residual fracture toughness and elastic properties of the fully brittle material).

Despite the fact that 17% ECR + Baker - Just + 2200 °F PCT don't ensure ZDT ≤ 135 °C in the neck (high H uptake not known in 1973) - However wall thinning, power, temperature and oxidation are lower - This was not known in 1973.

HIGH BURUP ZIRCALOY

Effect of H uptake (during irradiation) at the location where β thickness will become low: transient 17% ECR + Baker - Just + 2200 °F PCT inadequate to ensure ZDT ≤ 135 °C nor global survival to fully constrained thermal shock tests (see H. Uetsuka's presentation) nor likely global resistance to 0.3 J impact tests.

Need of better understanding of combined effect of H uptake (initial + transient) and O uptake (transient) on post - quench ductility.
TRANSPOSITION TO E 110 ALLOY

STEAM OXIDATION - E 110
CRITERION 1 (PCT) - E 110
CRITERION 2 (ECR) - E 110
QUENCH TESTS
LOW TEMPERATURE DUCTILITY TESTS
CONCLUSIONS

E 110 STEAM OXIDATION

Microstructure of steam oxidized specimen

Weight gain - 24,06 mg/cm².

Layered and cracked oxide structure below 1100 °C, even for low exposure times - more compact between 1100 and 1200 °C
STEAM OXIDATION (3)

Coefficient of growth of ZrO₂ and oxygen stabilized α- Zr layers.

Temperature, °C

1600 1500 1400 1300 1200 1100 1000

10⁻²

K, cm²/s⁻¹/²

Reverse temperature 10⁶/T, 1/K

Urbanic (Zr2.5%Nb)

this work (Zr1%Nb)

STEAM OXIDATION (2)

Bohmer (Nuclear Engineering and Design)

Zr-Nb1

Zr-Nb1

Zr-Nb1

Zr-Nb1

X Experimental results Zr-Nb1

X Experimental results Zr-Nb1

Zr-Y-1

Zr-Y-1

Zr-Y-1

Zr-Y-1

X Experimental results Zr-Y-1

X Experimental results Zr-Y-1

Fig. 4. Temperature dependence of the growth of the α-layer thickness δα for comparison of the experimental results for Zr-Nb1 and Zr-Y-1.

d(z)/Zr) phase thicker above 950 °C confirmed by V. Vrblík et al., NRI Rez 1993

29
Above 900 °C, oxygen content in the β phase higher (Böhmer - Nuclear Engineering and Design) confirmed by V. Vrtlíková, M. Valach et al., NRI Rez, 1993

Fig. 5. Radial profile of microhardness for Zr:Nb1 and Zry-4 after high-temperature steam oxidation.
STEAM OXIDATION (7)

STEAM OXIDATION (6)

Böhmer (NED)

Hydrogen content and uptake during steam oxidation for ZrNb1 and Zry-4.

<table>
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<th>$T$ (°C)</th>
<th>$T$ (min)</th>
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<td>Relative hydrogen uptake(%)</td>
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1000 - 2000 ppm confirmed by V. Vrtílková, M. Valach et al., NRI Rez, 1993

Fig 4 Zr1%Nb hydrogen uptake vs mass gain during steam oxidation

Higher hydrogen content, especially below 1100 °C, but even between 1100 and 1200 °C
Fig. 11. Oxide scale structure for ZrNb1 and Zry-4.

Despite macroscopically unlimited steam supply, microscopically stagnant steam conditions between oxide sublayers.

E 110 - QUENCH TESTS

Only unconstrained quench tests published in English

- large margin below 1200 °C

however, less margin to O solid-solution hardening

- lack of tin
- Nb addition
- large H uptake are shifting the O solubility in the β phase to higher value

Figure 8 Thermal shock tests. Fuel rod simulators of the WWER-type (Zr1%Nb alloy). Filler - UO₂ pellets
The residual plasticity of the Zr1%Nb decreases much more rapidly than that of Zircaloy, the sudden drop down is for Zr1%Nb at 6% oxide thickness and at 17% for Zircaloy, so for now the question of applicability of the Zircaloy oxidation limit remains for Zr1%Nb open.
"The degree of oxidation where complete embrittlement can be expected (shows) a 6% oxidation rate value in comparison with the 17% of Zry-4" (TOPSAFE 95)

DUCTILITY TESTS (2)

Ductility of steam-reacted Zr1% Nb claddings. Ring compression results.
Bochvar Institute, Varna 1994

Very good linear correlation for ZDT. The line reaches 275 °F (135°C) for a weight gain of 4.7 mg/cm² that is to say 6% ECR.

The application of the hearing methodology leads to ZDT ≤ 275 °F or ECR ≤ 6%.
E 110 ALLOY - ECR CONCLUSIONS

The application of the 1973 ECCS Rule - making hearing Methodology leads for E 110 alloy to a limit of 6% ECR associated with the use of the conservative Bochvar Institute's correlation.

PCT - CONCLUSIONS

Total lack of published ductility results above 1200 °C - quench tests show greater susceptibility to O solid - solution hardening

OTHER Nb - CONTAINING ALLOYS

DO OTHER Nb - CONTAINING ALLOYS SUCH AS M5 BEHAVE LIKE E110?

Need of better understanding of combined effects of Nb addition, H and O uptake on post-quench ductility
Discussion:

Question from the audience: Could you comment on effects of fuel relocation?

Answer by G. Hache: There will be a special paper presented during this meeting devoted to the possible impact of fuel relocation (see paper No. 11)
Supporting paper presented during the Water Reactor Safety Meeting - Washington 2000

THE HISTORY OF LOCA EMBRITTLEMENT CRITERIA

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Abstract

Performance of high-burnup fuel and fuel cladding fabricated from new types of alloys (such as Zirlo, M5, MDA, and duplex alloys) under loss-of-coolant-accident (LOCA) situations is not well understood at this time. To correctly interpret the results of investigations on the performance of the old and new types of fuel cladding, especially at high burnup, it is necessary to accurately understand the history and relevant databases of current LOCA embrittlement criteria. In this paper, documented records of the 1973 Emergency Core Cooling System (ECCS) Rule-Making Hearing were carefully examined to clarify the rationale and data bases used to establish the 1204°C peak cladding temperature and 17% maximum oxidation limits. A large amount of data, obtained for zero- or low-burnup Zircaloy cladding and reported in literature only after the 1973 Rule-Making Hearing, were also evaluated and compared with the current criteria to better quantify the margin of safety under LOCA conditions.

1. Introduction

Because of major advantages in fuel-cycle costs, reactor operation, and waste management, the current trend in the nuclear industry is to increase fuel discharge burnup. At high burnup, fuel rods fabricated from conventional Zircaloy often exhibit significant degradation in microstructure. This is especially pronounced in pressurized-water reactor (PWR) rods fabricated from standard Zircaloy-4 in which significant oxidation, hydriding, and oxide spallation can occur. Thus, many fuel vendors have developed and proposed the use of new cladding alloys, such as low-tin Zircaloy-4, Zirlo, M5, MDA, duplex cladding, and Zr-lined Zircaloy-2. Performance of these alloys under loss-of-coolant-accident (LOCA) situations, especially at high burnup, is not well understood at this time. Therefore, it is important to verify the safety margins for high-burnup fuel and fuels clad with new alloys. In recognition of this, LOCA-related behavior of various types of high-burnup fuel cladding is being actively investigated in several countries [1-6]. However, to correctly interpret the results of such investigations, and if necessary, to establish new embrittlement thresholds that maintain an adequate safety margin for high-burnup operation, it appears necessary to accurately understand the rationale, history, and data bases used to establish the current LOCA criteria, i.e., maximum cladding temperature limit of 1204°C (2200°F) and maximum oxidation limit of 17%. For this purpose, documented records of the 1973 Atomic Energy Commission (AEC) Emergency Core Cooling System (ECCS) Rule-Making Hearing were carefully examined and the relevant databases were reevaluated in this paper. Since the establishment of the current criteria, large amounts of data were obtained in many countries for zero- or low-burnup fuel cladding. The results of these investigations were also critically evaluated to determine the validity of the current criteria and safety margins for a wider range of conditions.
2. Primary Objectives of Current Criteria

In 1967, an Advisory Task Force on Power Reactor Emergency Cooling [7], appointed to provide "additional assurance that substantial meltdown is prevented" by core cooling systems, concluded that:

"The analysis of a LOCA requires that the core be maintained in place and essentially intact to preserve the heat-transfer area and coolant-flow geometry. Without preservation of heat-transfer area and coolant-flow geometry, fuel-element melting and core disassembly would be expected... Continuity of emergency core cooling must be maintained after termination of the temperature transient for an indefinite period until the heat generation decays to an insignificant level, or until disposition of the core is made."

This rationale makes it plainly clear that it is most important to preserve the heat transfer area and the coolant flow geometry not only during the short-term portion of the core temperature transient but also for long term.

Consistent with the conclusions of the Ergen Task Force, the U.S. Atomic Energy Commission (AEC) promulgated Criterion 35 of the General Design Criteria [8] which states that: "... fuel and clad damage that could interfere with continued effective core cooling is prevented." It also promulgated Criterion 3 of the Interim Acceptance Criteria for ECCS for LWR [9] which states that: "The clad temperature transient is terminated at a time when the core geometry is still amenable to cooling, and before the cladding is so embrittled as to fail during or after quenching."

These criteria were subjected to a Rule-Making Hearing in 1973, which was extensively documented in the Journal of Nuclear Safety in 1974 [10,11]. During the hearing process, the last part of the Criterion 3 was replaced by the modified Criterion 1 and the new Criterion 2 of the Code of Federal Regulations, Title 10, Part 50.46, Article (b), commonly referred to as 10 CFR 50.46 [12]. Thus, the AEC Commissioners wrote:

"In view of the fundamental and historical importance of maintaining core coolability, we retain this criterion as a basic objective, in a more general form than it appeared in the Interim Acceptance Criteria. It is not controversial as a criterion... Although most of the attention of the ECCS hearings has been focused on the events of the first few minutes after a postulated major cooling line break, up to the time that the clad would be cooled to a temperature of 300°F or less, the long-term maintenance of cooling would be equally important [13]."

There are two key factors to consider to evaluate the change in coolable geometry of core, a brittle mode and a ductile mode of deformation in fuel cladding. The ductile mode is related to cladding ballooning, burst, and coolant channel blockage. This mode will not be treated in this paper. Our focus in this paper is on the change in coolable geometry due to cladding embrittlement and failure.

3. Metallurgy of Cladding Embrittlement

In 1960s, Wilson and Barnes performed laboratory tests simulating steam reactions with Zircaloy-clad fuel rods at high temperatures. They observed embrittlement of oxidized cladding well below the melting temperature of Zircaloy, either during the test itself or during removal of the specimen from the oxidizing furnace. The results were reported in Argonne National Laboratory (ANL) progress reports and synthesized later in Ref. 14. At the same period, investigators in Oak Ridge National Laboratory (ORNL) conducted TREAT Test No. 6 with Zircaloy cladding in steam and observed that the specimen was severely embrittled by oxidation [15]. Also at about the same period, many tests were conducted that simulated reactivity-initiated accident (RIA) in SPERT-CDC and TREAT reactors. Results of metallurgical examination in these tests showed that embrittlement was caused by severe microstructural
modification of the cladding. Brittle cladding cross sections exhibited oxide layer, oxygen-stabilized alpha-phase layer and a region of acicular prior beta-phase. The results were later reported by Fujishiro et al. [16].

As a result of these observations, the scientific community was alerted to the fact that oxidation of Zircaloy above the alpha-to-beta transformation temperature results in the formation of inherently brittle phases, i.e., Zr oxide, oxygen stabilized alpha-Zr (fcc structure), and diffusion of oxygen into the underlying beta phase (bcc structure). This is shown schematically in Fig. 1. Ductility of cladding could be severely degraded if the degree of oxidation is high. It was also realized that, if the embrittled cladding fragments into small pieces, the coolability of the core could be seriously impaired.
Fig. 1. Schematic illustration of microstructure (top) and oxygen distribution (bottom) in oxide, stabilized alpha, and prior-beta (transformed-beta) layers in Zircaloy cladding after oxidation near 1200°C.

Significantly embrittled cladding can fragment during the quenching phase of a LOCA. The action of rewetting by ECCS water involves the collapse of the vapor film that covers the cladding outer-diameter (OD) surface prior to subsequent transition to nucleate boiling. This event takes place at a more or less constant temperature, i.e., the Leidenfrost temperature. For oxidized Zircaloy-4 cladding rewetted by bottom-flooding water, ANL investigators reported that rewetting occurs in the range of 475-600°C [17]. The abrupt change in the heat transfer conditions induces large thermal-shock stress, which can fracture the cladding, if it is sufficiently embrittled by oxidation.

Below the Leidenfrost temperature, there is continued risk of fragmentation after quenching. In accordance with the opinions of the Ergen Task Force and the AEC staff and commissioners mentioned earlier, other experts also wrote a similar opinion for OECD Committee on Safety of Nuclear Installations (CSNI) [18]. "The ability of the cladding to withstand the thermal-shock stresses of quenching during rewetting or post-LOCA forces is related to the extent and detailed nature of oxidation during the transient. The post-LOCA forces, which need to be taken into account, are the hydraulic, seismic, handling, and transport forces."

There are two primary factors that exacerbate the susceptibility of oxidized cladding to post-quench embrittlement in comparison with susceptibility to fragmentation during quenching: i.e., (1) more pronounced effect of oxygen dissolved in beta phase at lower temperature of loading (i.e., more pronounced after quench than during quench) and (2) more pronounced effect of hydrogen uptake which may occur during irradiation (e.g., in high-burnup Zircaloy-4) or during transient oxidation in steam (e.g., from cladding inner surface in contact with stagnant steam near a ballooned and burst region). For cooling
rates typical of bottom flooding of core (i.e., 1-5°C/s), most hydrogen atoms remain in solution in the beta phase at Leidenfrost temperature, and in such state, hydrogen has little effect on the fracture resistance of an oxidized Zircaloy. However, when load is imposed at temperatures below the Leidenfrost temperature, precipitated hydrides strongly influence the fracture resistance of cladding. Eutectoid decomposition of hydrogen-stabilized beta phase at temperatures below ≈550°C [19] is the major factor that causes this deleterious effect (see Fig. 2).

4. Opinion of Regulatory Staff and Commissioners during 1973 Rule-Making Hearing

4.1 Reluctance to Neglect Effects of Mechanical Constraints

Some factors during a LOCA, such as ballooning of the rod near the spacer grid, rod-grid spring chemical interaction, and the friction between the fuel rod and spacer grids, can restrict the axial movement of the cladding. Also, guide tubes in a PWR fuel assembly are mechanically fixed to the spacer grids. Because of these factors, fuel rods during reflooding will be subject to tensile load that is produced due to the differential axial shrinkage between a cladding and the guide tube. Rods may interact each other due to ballooning or bowing. For high-burnup fuels in which tight pellet-cladding bonding is common, axial shrinkage can be restricted if the tight bonding remains unchanged after ballooning and burst. These constraints will remain after quench, when deleterious effects of oxygen and hydrogen are far more pronounced.

In recognition of this, the AEC Staff wrote during the 1973 Rule-Making Hearing that "the loads due to assembly restraint and rod-to-rod interaction may not be small compared to the thermal shock load and cannot be neglected [20]." Subsequently, it was concluded that: "The staff believes that quench loads are likely the major loads, but the staff does not believe that the evidence is as yet conclusive enough to ignore all other loads [21]."
Then, the Commissioners added: "There is some lack of certainty as to just what nature of stresses would be encountered during the LOCA.... (We want) to draw attention to the fact that it may not be possible to anticipate and calculate all of the stresses to which fuel rods would be subjected in a LOCA. Although we believe the calculations of thermal shock stresses are worthwhile and informative, we agree with the regulatory staff that they are not sufficiently well defined to depend on for regulatory purposes [13]."

Before 1973, no thermal-shock quench test was performed on mechanically constrained cladding specimens. Then in early 1980s, Uetsuka et al. performed quenching tests on cladding sections under severely constrained condition [22]. In their experiment, cladding tube was fixed at the bottom but was allowed to freely elongate in axial direction during oxidation at high temperature. As a result, cladding length increased freely because of thermal expansion and oxide-induced creep. At the end of the isothermal oxidation, the specimen top was fixed to the crosshead of an Instron tensile facility. Then, the load-time curve was continuously monitored during quenching. Thus, at Leidenfrost temperature, the cladding tube was subjected to combined axial-tensile and thermal-shock stresses. The results of the tests are summarized in the Fig. 3. Similar tests were also performed on unconstrained tubes (Fig. 4).

Figure 3.
Failure-nonfailure boundary for fully constrained Zircaloy-4 after oxidation in steam and quenching as function of oxidation time and temperature; total oxidation calculated with Baker-Just equation is also indicated (from Uetsuska et al., J. Nucl. Sci. Tech. 20, 1983, pp. 941-950).

A comparison of the results from the two contrasting types of test shows a large effect of the mechanical constraint. However, it is difficult to conclude whether the degree of constraint in the experiments of Uetsuka et al. is prototypic of a LOCA or unrealistically too severe. The 17% oxidation limit, calculated with Baker-Just correlation, appears to be adequate for protection of constrained rods against thermal-shock failure (Fig. 3), whereas a large margin is evident for unconstrained rods (Fig. 4).
Unlike other bundle tests such as NRU, REBEKA, JAERI and ORNL multitrod tests that were entirely devoted to the study of ballooning, burst, and flow-channel blockage, some of the tests in Phebus LOCA program were devoted to the study of embrittlement [23]. The fragmented Rod 18 of the Test 219, exposed to \( \approx 1330^\circ \text{C} \), is especially interesting (see Fig. 5). For this oxidation temperature, results of calculation with PRECIP-II Code [24] indicates that the O content in the beta phase was higher than 0.9 wt.\%, a threshold O concentration found to be associated with thermal-shock failure or survival [17]. Rod 18 fragmented despite it was oxidized to an equivalent-cladding reacted (ECR) value of only \( \approx 16\% \). This observation indicates a deleterious bundle effect, i.e., an additional mechanical constraint.

As a conclusion, results of the JAERI constraint quench test and the PHEBUS-LOCA Test appear to justify the reluctance of the AEC staff and commissioners to neglect the effect of mechanical constraints on the susceptibility to thermal-shock failure.

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure4.png}
\caption{Failure-nonfailure boundary for unconstrained Zircaloy-4 after oxidation in steam and quenching as function of oxidation time and temperature; total oxidation calculated with Baker-Just equation is also shown (from Uetsuska et al., J. Nucl. Sci. Tech. 20, 1983, pp. 941-950).}
\end{figure}
4.2 Preservation of Ductility and Consideration of Results from Unconstrained Quench Test

At the end of the 1973 Hearing, the AEC Commissioners wrote: "...Nevertheless we find the quench results encouraging in that they provide assurance that the 2200°F limit is conservative. Our selection of the 2200°F limit results primarily from our belief that retention of ductility in the Zircaloy is the best guarantee of its remaining intact during the hypothetical LOCA... The thermal shock tests are reassuring, but their use for licensing purposes would involve an assumption of knowledge of the detailed process taking place in the core during a LOCA that we do not believe is justified [13]."

Without much ambiguity, this conclusion clearly expressed the belief that retention of ductility was considered the best guarantee against potential fragmentation under various types of loading (thermal-shock, bundle constraints, hydraulic, handling, and seismic forces). During the 1973 Hearing, results from unconstrained quench tests (simple thermal-shock test) were considered only corroborative and reassuring. However, their use for regulatory purposes was not accepted.

Results of later investigations on unconstrained or partially constrained cladding [17,18] showed a large margin of survival under thermal shock relative to 17%-ECR and 2200°F (1204°C) peak temperature limits. Such results are summarized in Fig. 6. No fragmentation occurred for ECR < 17% for all oxidation temperatures, whereas significant margin of survival was observed for oxidation temperatures <1204°C. The results in Fig. 6 were limited for thermal-shock tests in which cladding tube or ring was directly quenched from the maximum oxidation temperature without slow cooling through the range of beta-to-alpha-prime transformation. For slow-cooling conditions, more pronounced margin of survival was observed [17].
5. **17%-Oxidation Criterion**

5.1 **Establishment of 17% Criterion During 1973 Rule-Making Hearing**

The rationale for establishment of the two criteria in 10 CFR 50.46(b) is described in this section. As indicated in a few reports [17,18] that reviewed the results of the LOCA-related tests performed before and after the 1973 Hearing, the 17%-ECR and 1204°C criteria were primarily based on the results of post-quench ductility tests conducted by Hobson [25,26].

Figure 7 summarizes the results of Hobson's ring compression tests performed at 23-150°C. Zircaloy-4 cladding tubes were oxidized in steam on two sides, followed by direct quenching into water. Then, short ring specimens cut from the oxidized tube were either compressed slowly to a total deflection of 3.8 mm or squashed by impact loading. After the test, the broken pieces of the ring were assembled back to determine the degree of brittleness. Zero ductility was defined on the basis of the macroscopic geometry of the broken pieces and the morphology of the fracture surface on microscopic scale. Each data point in Fig. 7 indicates failure type, test identification number, oxidation time in min., oxidation temperature in °F, and first maximum load in pound.
Figure 7.
Ductility of two-side-oxidized Zircaloy rings as function of slow- or fast-compression temperature and fraction of transformed-beta-layer (from Hobson, Ref. 25 and 26).

The dashed line on the left side of Fig. 7 denotes the zero ductility domain for slow-compression rate. This domain is valid only for oxidation temperatures of $<$2200°F or $<$1204°C. During the 1973 Hearing, ORNL investigators suggested to consider a zero-ductility temperature (ZDT) no higher than the saturation temperature during reflood, i.e., $\approx$135°C. Zero-ductility threshold at this temperature is equivalent to a beta-layer fraction of $\approx$0.58, or a fraction of combined oxide layer plus alpha layer thickness (defined as $X_T$) of $\approx$0.42 (based on as-oxidized cladding wall). The latter fraction corresponds to $\approx$0.44 if it is calculated based on fresh nonoxidized cladding wall (defined as $W_0$).

The threshold fractional thickness of the combined oxide and alpha layer ($X_T/W_0$, defined as $X_{oa}$ in Fig. 8) of 0.44, which corresponds to zero ductility threshold for slow compression at 135°C, was the key
number in the establishment of 17% oxidation criterion in the 1973 Hearing. During the hearing, the AEC Regulatory Staff wrote:

"Giving due credit to the numerous quench experiments and the ORNL zero ductility experimental data points for both impact and slow compression, the staff suggests that an embrittlement criterion be based on a calculated $X_T/W_o$ that shall not exceed 0.44. This is equivalent to a zero ductility temperature of about ... 275°F based on the slow compression tests [20]."

Then, it was concluded:

"To preclude clad fragmentation and to account for effects noted in the tests described above, a limit of $X_T/W_o \cdot 0.44$ was earlier suggested by the Regulatory staff as an embrittlement criterion (Exhibit 1113, page 18-18). This limit was inferred from quench tests and mechanical tests. Criterion (b)(2) is now proposed as a better method of specifying a similar limit on the extent of cladding oxidation. The bases for proposing this method are described below: (The) use of the 17 percent reaction limit with the Baker-Just equation is conservative when compared to the previously suggested limits of $X_T/W_o \cdot 0.44$. This is shown in Figure 8 (of this paper) for isothermal conditions. Four lines of constant calculated $X_T/W_o$ (two for 0.44 and two for 0.35) are constructed on the plot of percent reaction versus a parameter proportional to the square root of exposure time. The solid $X_T/W_o$ lines are based on Pawel's equation (Exhibit 1133) (Ref. 27 of this paper), and the dashed lines are based on Exhibit 09, page 9, Figure 5 (Ref. 25 of this paper). As can be seen, the $X_T/W_o = 0.44$ lines are both above the 17 percent reaction line..."

Results of a total of five key tests and calculations are summarized in Fig. 8, a complex but the most important step used to reach the 17% oxidation limit. They are: (1) equivalent cladding reacted (ECR) calculated as function of oxidation temperature and square root of time based on Baker-Just correlation, (2) two broken curves which define the time and temperature to reach the threshold fractional thickness of the combined oxide and alpha layer (denoted as Xoa) of 0.44 and 0.35, as determined based on the data given in Ref. 25, Page 9, Fig.5, (3) two solid curves that define the time and temperature to reach the threshold fractional thickness of the combined oxide and alpha layer of 0.44 and 0.35, as determined based on the method of Ref. 27, (4) six ECR-(time)$^{0.5}$ curves from the thermal-shock tests of Hesson et al., Ref. 14, and (5) results from Combustion Engineering (CE) ring compression tests after one-sided oxidation.
Figure 8.

Summary of multitstep procedure used to establish 17% oxidation criterion during 1973 Rule-Making Hearing (from Docket RM-50-1, April 16, 1973). Note equivalent cladding oxidized was calculated per Baker-Just correlation. For comparison, time to reach threshold fraction of combined oxide and alpha layers of 0.44 is shown as determined per Hobson and Rittenhouse (ORNL-4758, January 1972) and Pawel (J. Nucl. Mater. 50, 1973, pp. 247-258).

By definition, ECR parameter varies depending on cladding wall thickness, either due to differences in fuel design or due to ballooning and burst during the heatup phase in a LOCA. Figure 8 shows how to take account of the effects of variations in wall thickness and one- vs. two-sided oxidation.

Two of Hesson’s thermal-shock experiments resulted in cladding fragmentation at calculated ECR values of ≈19 and ≈30%, as indicated in the figure. The other four did not fail at ECR values of ≈21, ≈16.5, ≈10, and ≈9.5%. The time-temperature transients in Hesson’s tests were integrated also by using the Baker-Just equation.

The CE data, discussed in the Hearing, are represented by squares on the oxidation isotherms of 2500, 2400, 2300, and 2100°F. If the sample fractured on compression by CE’s load standard, it was
considered to have failed and is denoted with a filled square. Open squares denote CE's non-failed specimens. By the CE's load standard, only those samples with calculated ECR values >17% failed.

Based on the results given in Fig. 7 and the five sets of information shown in Fig. 8, one can conclude that no samples tested by slow compression at >135°C failed with zero ductility if equivalent cladding reacted (ECR), calculated on the basis of Baker-Just correlation, was less than 17%. Furthermore, all samples oxidized to <17% ECR (again calculated with Baker-Just correlation) survived direct quenching.

In summary, the ABC Commissioners concluded that the very good consistency between the 17% limit, if calculated with the Baker-Just equation, and a wide variety of experiments supports adoption of this procedure [21], and it was further stated:

"There is relatively good agreement among the industrial participants as to what the limit on total oxidation should be.... The regulatory staff in their concluding statement compared various measures of oxidation and concluded that a 17% total oxidation limit is satisfactory, if calculated by the Baker-Just equation... As argued by the regulatory staff, it appears that the 17% oxidation limit is within the Rittenhouse criteria. Thus a remarkable uniformity of opinion seems to exist with regard to the 17% oxidation limit [13]."

It is clear that the primary rationale of the 17% criterion is retention of cladding ductility at temperatures higher than 275°F (135°C, i.e., the saturation temperature during reflood). Of major importance in this proceeding is that the threshold ECR value of 17% is tied with the use of Baker-Just correlation. That is, the 17% ECR criterion is specific to Baker-Just correlation that must be used to determine the degree of total oxidation. If an oxidation correlation other than the Baker-Just equation (e.g., Cathcart-Pawel correlation) were used, the threshold ECR would have been less than 17%. This means that use of a best-estimate correlation may not necessarily be conservative in evaluating post-quench cladding ductility.

5.2 Other Embrittlement Criteria Proposed after the 1973 Hearing

Few months after the 1973 Hearing, Pawel proposed a new criterion based on <95% saturation of the average oxygen concentration in the beta phase [27]. However, such a criterion fails to recognize that in addition to a sufficiently low O concentration, a minimum thickness of beta layer is required to ensure adequate resistance to failure. Such criterion is less facilitated to handle, especially during non-isothermal LOCA transients, and it requires a computer code that can accurately calculate O diffusion under moving-phase-boundary conditions, a task more difficult than the calculation of a simple parabolic oxidation correlation. Nevertheless, many of such computer codes have been developed after the 1973 Hearing, e.g., those reported in Refs. 17 and 24.

Sawatzky performed room-temperature tensile tests on specimens exposed to high-temperature spikes in steam [28]. Based on results of microhardness measurement, the distribution of O in the transformed beta (or prior beta) layer was found to be nonuniform, an observation confirmed subsequently by ANL investigators by Auger electron spectroscopy (Fig. 32-43, Ref. 17). In spite of total oxidation of only 16%, a specimen with average O concentration >0.8 wt% in the prior beta exhibited very low strength and negligible elongation, whereas a specimen with O content <0.6 wt% in the prior beta retained some ductility. Based on this observation, Sawatzky proposed to replace the 1204°C PCT and the 17% ECR criteria by a unified criterion, that is, oxygen concentration in beta layer shall be <0.7 wt% over at least half of the cladding thickness. At temperatures >1280°C, Sawatzky's criterion is virtually identical to Pawel's criterion (see Fig. 9).
Validity of the three criteria illustrated in Fig. 9 is, however, subject to variations in cladding wall thickness, because the time to reach the specified threshold state of material is strongly influenced by the clad wall thickness which may vary with fuel design and the degree of ballooning and burst. Thus, it was deemed desirable to develop a unified embrittlement criterion that would be valid independent of variations in wall thickness and oxidation temperature [17].

5.3 One- vs. Two-Side Oxidation and Thermal-Shock Failure

Grandjean et al. have reported results of extensive thermal-shock tests which were performed in TAGCIS facility [29,30]. Hydrogen uptake in their short ring specimens was not excessive. In their investigation, ECR was calculated with PECLOX oxidation code [31], and failure-survival behavior was determined based on the result of gas-leakage check. The results of the tests were included in Fig. 6. The effect of one- vs. two-side oxidation on thermal-shock failure was the focus of investigation. As indicated in Fig. 8, such effect was considered negligible in establishing the 17% ECR limit in the 1973 Rule-Making Hearing. Interestingly, Grandjean et al.'s failure threshold for two-side oxidation appears to be slightly higher than the threshold for one-sided oxidation, i.e., \( \approx 21 \) vs. \( \approx 20 \)% ECR. Nonetheless, this study provides an independent confirmation of the validity of the 17% ECR criterion relative to susceptibility to thermal-shock failure.

![Figure 9.](image)

Comparison of current embrittlement criteria with those proposed by Pawel (Ref. 27) and Sawatzky (Ref. 28).
5.4 17% Oxidation Limit and Impact Failure at Small Hydrogen Uptake

After the 1973 Hearing, ANL investigators conducted impact tests to provide an independent verification of the validity of 17% ECR threshold with respect to cladding resistance to impact failure [17]. Impact tests were performed at room temperature on non-pressurized open-ended Zircaloy-4 tubes that were oxidized on two sides in steam at 1100-1400°C and cooled through the beta-to-alpha-prime transformation range at 5 or ≈100°C/s. Because the sample was oxidized on both OD and ID sides, hydrogen uptake was limited to <130 wppm. Therefore, microstructure and oxygen and hydrogen distributions in the specimens were similar to those of the ring-compression specimens of Hobson [25,26] that were cooled fast through the beta-to-alpha-prime transformation range.

It was found that slow-cooled specimens were more resistant to impact failure than fast-cooled specimens (Fig. 65, Ref. 17). Results obtained for slow-cooled specimens are summarized in Fig. 10. The ECR values in Fig. 10 were directly determined based on measured phase layer thickness, therefore, are considered more accurate than values calculated based on Baker-Just correlation. The results in Fig. 10 show that for cladding oxidized at <1315°C to <17% ECR, a sufficient level of resistance to impact failure is retained at 23°C, i.e., failure impact energy of >0.8 J.

![Figure 10.](image)

Failure impact energy vs. equivalent cladding reacted, from tests at 23°C on undeformed Zircaloy-4 tube oxidized in steam at 1100°C and cooled at 5°C/s (Ref. 17).

5.5 17% Limit and Ring-Compression Ductility at Small Hydrogen Uptake

As shown in Fig. 8, the 17% threshold ECR was derived by indirect multistep procedure. Of particular importance in this procedure is the accuracy of two key factors, i.e., (1) temperature measurement in the experiments of Baker-Just and Hobson-Rittenhouse [25,26] and (2) definition of nil-ductility as given in Fig. 7. In consideration of this, ANL investigators performed independent compression tests at room temperature on short Zircaloy-4 ring specimens. Rings were sectioned from long tubes that were oxidized in steam at 1100-1400°C and cooled through the beta-to-alpha-prime transformation range at 5 or ≈100°C/s. Hydrogen uptake in the ring specimens was <130 wppm. This procedure reproduced the conditions of the ring-compression tests of Hobson. In the ANL compression tests, however, load-deflection curves were obtained to better quantify the degree of remaining ductility and the magnitude of load that a ring can sustain.
It was found that slow-cooled specimens retained more ductility than fast-cooled specimens under otherwise identical conditions (Fig. 67, Ref. 17). Figure 11 summarizes results obtained for a slow-cooling rate of \( \approx 5^\circ\text{C}/\text{s} \), a rate probably more prototypic of a LOCA than fast cooling. The ECR values in the figure were determined based on measured phase layer thickness and time-temperature history. This result shows that for cladding oxidized at \( <1315^\circ\text{C} \) to \( <17\% \) ECR, ductility is retained at \( 23^\circ\text{C} \) (i.e., relative diametral deflection \( >16\% \)); no brittle failure was observed. This experiment provides an independent confirmation of the validity of the 17\% oxidation limit for undeformed Zircaloy specimens that contain hydrogen \( <130 \) wppm.

![Figure 11.](image)

**Figure 11.**

**Total deflection at 23°C vs. equivalent cladding reacted, from ring-compression tests on Zircaloy-4 oxidized on two-sides and cooled at 5°C/s (Ref. 17).**

5.6 **Resistance to Impact Failure at Large Hydrogen Uptake**

In addition to the impact tests on non-ruptured empty tubes, ANL investigators performed 0.15- and 0.3-J pendulum impact tests at \( 23^\circ\text{C} \) on pressurized Zircaloy-4 tubes that were burst, oxidized, cooled at \( \approx 5^\circ\text{C}/\text{s} \), and survived quenching thermal shock [17]. The CSNI experts [18] considered that: "Ambient impact of 0.3 J were thought to be a reasonable approximation to post LOCA quench ambient impact loads." The results of the 0.3-J impact tests, summarized in Fig. 12, indicate that the 17\%-ECR limit is adequate to prevent a burst-and-oxidized cladding from failure under 0.3-J impact at \( 23^\circ\text{C} \), as long as peak cladding temperature remained \( \leq 1204^\circ\text{C} \). The ECR values in the figure were determined based on measured thickness of oxide, alpha, and beta phase layers, rather than calculated based on Baker-Just correlation, and hence, are considered more accurate.
In contrast to two-side-oxidized non-pressurized non-ruptured tubes in which hydrogen uptake was small (<130 wppm), burst Zircaloy-4 tubes exhibited peculiar oxidation behavior near the burst opening. The inner-diameter (ID) surfaces of the top and bottom "necks," ~30-mm away from the burst center, were exposed to hydrogen-rich stagnant steam-hydrogen mixture which is produced because of poor mixing of steam and hydrogen at the narrow gap between the alumina pellets and the ID surface of the necks. As a consequence, thick breakaway oxides formed at 900-1120°C [17], and hydrogen uptake as high as ~2200 wppm was observed at the "necked" regions. Subsequently, JAERI investigators confirmed occurrence of the same phenomenon [32,33].

The results from the same tests shown in Fig. 12 were converted to failure-survival map based on average hydrogen content of the impact-loaded local region and the thickness of transformed-beta layer containing <0.7 wt.% oxygen. This failure-survival map is shown in Fig. 13. On the basis of the figure, ANL investigators proposed to replace the 1204°C PCT and 17% BCR criteria by a unified criterion which specifies that the thickness of transformed-beta layer containing <0.7 wt.% oxygen shall be >0.3 mm [17]. The criterion implicitly incorporates a limit in peak cladding temperature. This limiting temperature corresponds to the temperature at which oxygen solubility is 0.7 wt.% in Zircaloy that contains 700-1200 wppm hydrogen. This temperature is believed to be between 1200 and 1250°C, although the exact data from applicable Zircaloy-O-H ternary diagrams are not. This criterion is not subject to variations in cladding wall thickness and oxidation temperature.
The results in Fig. 13 show that for a given thickness and a given oxygen content in transformed-beta layer, resistance of cladding to impact failure is significantly reduced if hydrogen uptake exceeds ≈700 wppm. Such situation does not occur in non-pressurized, non-ruptured, two-side-oxidized Zircaloy cladding, such as those tested by Hobson [26] or discussed in Figs. 10 or 11.

5.7 Ring-Compression Ductility at Large Hydrogen Uptake

Investigators in ANL [17] and JAERI [32,33] conducted extensive tests on tube or ring specimens of Zircaloy-4 that contained high concentrations of hydrogen. In the former investigation, Zircaloy-4 tubes filled with alumina "pellets" were pressurized, heated, burst, oxidized, slow-cooled, and quenched with bottom-flooding water. Then, the tubes that survived the quenching thermal shock were compressed diametrically at 23°C [17]. Such specimens contained H up to ≈2200 wppm. In the latter investigation, short rings, sectioned from tubes that were exposed to similar conditions, were compressed at 100°C. The ring specimens contained H up to ≈1800 wppm. Typical distributions of oxide layer thickness, hydrogen concentration, and ring deflection to failure are shown in Fig. 14. The top and bottom "necks" that contained the highest concentration of hydrogen and the thinnest transformed-beta layer exhibited the lowest ductility.

However, ANL investigators observed that the rate of hydrogen generation, amount of hydrogen uptake, and hence, the degree of embrittlement of the necked regions are strongly influenced by the method of heating cladding tubes during LOCA-like transients, i.e., more uniform (indirect heating in JAERI) vs. less uniform (direct heating in ANL) heating [17]. This is schematically illustrated in Fig. 15.
The effect of hydrogen uptake on post-quench ductility, determined either from diametral-compression test of burst-and-oxidized tubes at 23°C [17] or compression at 100°C of ring specimens sectioned from burst-and-oxidized tubes [32,33], is summarized in Fig. 16. At hydrogen uptake >700 wppm, significant embrittlement of cladding is evident, even if total oxidation is <17% (see Fig. 14). Similar dependencies of plastic deflection on beta-layer oxygen content and total hydrogen content have been also reported in Fig. 88, Ref. 17 and Fig. 89, Ref. 17, respectively. These results show that post-quench ductility of Zircaloy is strongly influenced by not only oxidation but also hydrogen uptake. This is shown in Fig. 17. Apparently, the important effect of hydrogen uptake on post-quench ductility was not well realized at the time of 1973 Hearing.
Essentially similar observation has also been reported by Komatsu et al. [34,35]. They reported that the load to initial ring cracking is strongly influenced by total oxidation and hydrogen uptake. For oxidation temperatures >1260°C in which the oxygen content in the beta layer exceeds ≈0.7 wt.% in short period of time, the embrittling effect of oxygen appears to be predominant (see Fig. 18). The "zero-ductility" region denoted in Fig. 18 appears to have been determined based on a threshold load to initial cracking rather than based on ductility consideration. As such, this "zero-ductility" threshold differs significantly from that defined by Hobson [25,26].
5.8 17% Oxidation Criterion - Summary

It is clear that the primary rationale of the 17% ECR criterion is retention of cladding ductility at temperatures higher than 275°F (135°C), i.e., the saturation temperature during reflood. The threshold ECR value of 17% is tied with the use of Baker-Just correlation. If a best-estimate correlation other than Baker-Just equation (e.g., Cathcart-Pawel correlation) were used, the threshold ECR would have been <17%.
NEA/CSNI/R(2001)18

Investigations conducted after the 1973 Rule-Making Hearing showed that for oxidation temperatures \( \leq 1204°C \), the 17% oxidation limit (as calculated with Baker-Just correlation) is adequate to ensure survival of fully constrained or unconstrained cladding under quenching thermal shock. It was also shown that the 17% limit (ECR determined on the basis of measured phase layer thickness) is adequate to ensure retention of ductility and resistance to 0.3-J impact failure in non-irradiated, non-ruptured, two-side-oxidized Zircaloy cladding in which hydrogen uptake during a LOCA-like transient is small.

However, the 17% limit appears to be inadequate to ensure post-quench ductility for hydrogen uptake >700 ppm. Such level of large hydrogen uptake could occur in some types of fuel rods during normal operation, especially at high burnup, or during a LOCA-like transient in localized regions in a ballooned and ruptured node.

6. 1204°C (2200°F) Peak Cladding Temperature Criterion

6.1 Selection of 1204°C Criterion in 1973 Hearing

From the results of posttest metallographic analysis of the slow-ring-compression specimens, Hobson [26] observed a good correlation between zero ductility temperature (ZDT) and fractional thickness of transformed-beta layer (or the sum of oxide plus alpha layer thickness) as long as the specimen was oxidized at \( \leq 2200°F \) (1204°C) (see Fig. 7). However, in spite of comparable thickness of transformed beta layer, specimens oxidized at 2400°F (1315°C) were far more brittle. This observation was explained on the basis of excessive solid-solution hardening of transformed-beta phase at high oxygen concentrations. For mechanical properties near room temperature the critical concentration of oxygen in the transformed-beta was estimated to be \( \approx 0.7 \text{ wt%} \). Above this concentration, transformed beta phase becomes brittle near room temperature. Because of the solubility limit of oxygen in the beta phase, this high O concentration cannot be reached at 2200°F (1204°C) but can be reached at 2400°F (1315°C). Hobson concluded that: "embrittlement is not simply a function of the extent of oxidation alone, but is related in yet another way to the exposure temperature."

During the 1973 Rule-Making Hearing, AEC Staff endorsed Hobson's conclusion and wrote: "The staff recognizes the importance of oxygen concentration in the beta phase in determining the load bearing ability of Zircaloy cladding, and the implication from the recent compression tests that this may not be satisfactorily characterized above 2200°F by a ZDT as a function of remaining beta fraction only. We therefore believe that peak cladding temperatures should be limited to 2200°F [20]."

Subsequently, it was also concluded that:

"Additional metallurgical and slow compression mechanical tests on other quenched samples from the ORNL experiments indicated that an important consideration was the amount and distribution of oxygen in the nominally ductile prior-beta phase. However, these factors could not be correlated as functions of time and temperature in the same manner as the (combined oxide and alpha layer) penetration. In particular, the slow compression tests indicated a greater degradation in cladding ductility at higher temperatures than would be expected from considerations of (combined oxide and alpha layer) penetration alone. It was on this basis that the staff previously suggested a 2200°F maximum cladding temperature... What was observed in the slow compression tests was that 6 samples exposed at 2400°F for only two minutes and with relatively high values of Fw (Fw being fractional thickness of prior beta; all greater than 0.65) all fractured with nil ductility... Only when brittle failure was detected at high Fw in the slow compression tests did the suspicion arise that ductility was a function of both Fw and the exposure temperature... As the temperature rises above 2200-2300°F, solid solution hardening in the beta phase appears to contribute significantly to formation of a brittle structure. That is, brittle failure occurs even though alpha incursions are not observed, and the fraction of remaining beta is greater than that observed

58
in lower temperature tests. This is confirmed by examination of the six samples from the ORNL exposed at 2400°F for two minutes (Exhibit 1126)... From the foregoing, there is ample evidence that load bearing ability and ductility decrease with increasing exposure temperatures, even for transients with comparable Fw. Increased solubility of oxygen in the prior-beta phase has been discussed as a contributing factor. The staff believes that because of high temperature degradation phenomena (... strongly suggested by the experimental evidence cited), the suggested 2200°F limit should be imposed [21]."

Then it was added:

"The situation is complicated by the fact that not all of the prior beta phase is equally strong or ductile, since these properties depend on the amount of dissolved oxygen. This fact has been suspected for some time. From the phase diagram, given by both Scatena and Westinghouse, it is obvious that it is possible for the beta phase zirconium to take on a higher oxygen content at 2600°F than at 2000°F. Furthermore, since the diffusion rate depends exponentially upon temperature, one might expect a greater incursion of oxygen into the beta phase for a given thickness of oxide and stabilized alpha phase at higher temperatures. Others (than Hobson) have also observed that the resistance to rupture depends upon the temperature at which oxidation occurs as well as the extent of oxidation. To recapitulate, measures of Zircaloy oxidation, whether by percent, XT, or Fw, are largely or wholly determined from the brittle layers of zirconium oxide or stabilized alpha phase, while the ductility and strength of oxidized zirconium depend upon the condition and the thickness of the prior beta phase. Thus a criterion based solely on the extent of total oxidation is not enough, and some additional criterion is needed to assure that the prior beta phase is not too brittle. The specification of a maximum temperature of 2200°F will accomplish this adequately. The data cited in exhibit 1113 would not support a choice of a less conservative limit [13]."

Few months after the Hearing, Pawel [27] explained Hobson's observation based on data that indicate oxygen solubility in the beta Zr at 2200-2400°F (1204-1316°C) is ≈0.7 wt.%. The O solubility in beta Zircaloy is significantly influenced by not only temperature but also the concentration of hydrogen, a strong beta stabilizer. Nevertheless, Pawel endorsed that: "...the above reasoning easily explains why the mechanical or load bearing properties of the oxidized specimens should not be a unique function of the extent of (total) oxidation." Consequently, Pawel proposed to replace the peak cladding temperature (PCT) criterion by a new criterion that specifies the average oxygen concentration in the beta phase shall be less than 0.7 wt% [see Fig. 9].

6.2 **1204°C Limit vs. In-Pile Test Results**

In 1970s, high-temperature oxidation and embrittlement behaviors were investigated extensively in TREAT and PBF test reactors. During the TREAT-FRF2 test, a seven-rod cluster was oxidized at 2400°F (=1315°C) [36]. According to hardness measurements, all rods contained portions that possessed no ductility at room temperature. Three rods were broken accidentally during handling in ORNL hot cell, which indicates the degree of brittleness of a badly embrittled rod and the magnitude of a typical load during handling in hot cell (see Fig. 19).

Some fuel rods tested in the Power Burst Facility (PBF) were also known to have failed during handling or posttest examination in hot cell. This information is summarized in Fig. 20 [37]. Total oxidation of several failed rods was <17%. Of particular interest is Rod IE-019 of Test IE 5, because ballooning and burst occurred in the rod before exposure to temperatures >1100°C. In spite of the fact that ECR was only ≈12%, the rod broke into pieces after exposure to an "equivalent" oxidation temperature of ≈1262°C. Most likely, actual peak temperature was higher than this equivalent temperature. Rod A-0021 also ruptured before entering high temperature transient; this caused ingress of steam to the rod interior.
The rod failed after exposure to $\approx 1307^\circ \text{C}$, although ECR was only $\approx 6\%$. Hydrogen uptake in the two rods was excessive because of exposure to stagnant steam near the rupture opening.

![TREAT-FRF 2 Test. Rod 11 Fragmented during Posttest Examination](image)

Figure 19.
Fuel pellet released through fragmented cladding section of Rod 11, TREAT-FRF 2 Test (from Ref. 37).

It is not clear if the failure behavior of Rods IE-019 and A-0021 is predicted based on Pawel's criterion (Fig. 9). However, because the exposure temperatures of the rods exceeded $\approx 1262^\circ \text{C}$, the thickness of beta layer that contained $\leq 0.7$ wt.% oxygen should have been zero or close to zero. However, because oxygen solubility in beta is influenced by hydrogen and because accurate peak temperatures reached in the rods are not well known, it is difficult to calculate accurately the thickness of beta layer that contains $O \leq 0.7$ wt.%. Therefore, it is not clear if the failure behavior of the two rods is consistent with the criterion shown in Fig. 13.

As long as clad oxidation temperature was limited to $\leq 1204^\circ \text{C}$, a handling failure at measured ECR $< 17\%$ was not observed from the TREAT and PBF tests or the ANL 0.3-J impact tests (see Fig. 20). This observation clearly demonstrates the importance of the $1204^\circ \text{C}$ PCT limit. That is, the $1204^\circ \text{C}$ PCT and the $17\%$ ECR limits are inseparable, and as such, constitute an integral criterion.

6.3 Summary of $1204^\circ \text{C}$ Criterion

The $2200^\circ \text{F}$ ($1204^\circ \text{C}$) peak cladding temperature (PCT) criterion was selected on the basis of Hobson's slow-ring-compression tests that were performed at 25-150°C. Samples oxidized at $2400^\circ \text{F}$ ($1315^\circ \text{C}$) were far more brittle than samples oxidized at $< 2200^\circ \text{F}$ ($< 1204^\circ \text{C}$) in spite of comparable level of total oxidation. This is because oxygen solid-solution hardening of the prior-beta phase is excessive at oxygen concentrations $> 0.7$ wt.%.

The selection of the $1204^\circ \text{C}$ criterion was subsequently justified by the observations from the ANL 0.3-J impact tests and the handling failure of rods tested in the Power Burst Facility. These results also take into account of the effect of large hydrogen uptake that occurred near the burst opening. Consideration of potential for runaway oxidation alone would have lead to a PCT limit somewhat higher than $2200^\circ \text{F}$ ($1204^\circ \text{C}$). In conjunction with the $17\%$ oxidation criterion, the primary objective of the PCT criterion is to ensure adequate margin of protection against post-quench failure that may occur under hydraulic, impact, handling, and seismic loading.
7. Conclusions

1. In the 1973 Rule-Making Hearing, the U. S. Atomic Energy Commission (AEC) staff and commissioners were clearly reluctant to neglect the effect of mechanical constraints on the susceptibility of oxidized fuel cladding to thermal-shock fragmentation. Subsequent test results appear to justify this rationale. Results from unconstrained or partially constrained quench tests were considered only corroborative; their use for regulatory purposes was not accepted.

2. The AEC staff and commissioners and OECD-CSNI specialists were of the opinion that retention of ductility was the best guarantee against potential fragmentation of fuel cladding under various types of not-so-well-quantified loading, such as thermal shock, hydraulic, and seismic forces, and the forces related with handling and transportation.

3. Primary rationale of the 17% oxidation criterion was retention of cladding ductility at temperatures higher than 275°F (135°C), i.e., the saturation temperature during reflood. The threshold equivalent cladding reacted (ECR) of 17% is tied with the use of Baker-Just correlation. If a best-estimate correlation other than Baker-Just equation (e.g., Cathcart-Pawel correlation) had been used, the threshold ECR would have been <17%.

4. Investigations conducted after the 1973 Rule-Making Hearing show that for oxidation temperatures ≤1204°C, the 17% oxidation limit (calculated with Baker-Just correlation) is adequate to ensure survival of unconstrained or fully constrained cladding under quenching thermal shock. It was also shown that the 17% limit (ECR determined on the basis of measured phase layer thickness) is adequate to ensure retention of ductility and resistance to 0.3-J impact failure in non-irradiated non-ruptured two-side-oxidized Zircaloy cladding in which hydrogen uptake during a LOCA-like transient is small.
5. However, the 17% ECR limit appears to be inadequate to ensure post-quench ductility at hydrogen concentrations >700 wppm. A major finding from tests performed after the 1973 Rule-Making Hearing shows that post-quench ductility is strongly influenced by not only oxidation but also hydrogen uptake. It seems that this effect of large hydrogen uptake was not known at the time of 1973 Hearing.

6. By definition, an embrittlement criterion expressed in terms of ECR is subject to uncertainties because calculated ECR varies with variations in cladding wall thickness and the degree of ballooning.

7. The 1204°C peak cladding temperature (PCT) limit was selected on the basis of slow-ring-compression tests that were performed at 25-150°C. Samples oxidized at 1315°C were far more brittle than samples oxidized at 1204°C in spite of comparable level of total oxidation. This is because oxygen solid-solution hardening of the prior-beta phase is excessive at oxygen concentrations >0.7 wt%. Consideration of potential for runaway oxidation was a secondary factor in selecting the 1204°C limit. The 1204°C limit was subsequently justified by the observations from impact tests and handling failure of fuel rods exposed to high temperatures in the Power Burst Facility. The 1204°C PCT and the 17% ECR limits are inseparable, and as such, constitute an integral criterion.

8. The degree of oxygen saturation and the thickness of beta layer that contains oxygen concentrations ≤0.7 wt.% were important parameters used by investigators to develop new embrittlement criteria based on beta phase thickness rather than total oxidation. Such a criterion is not subject to inherent uncertainties associated with variations in cladding wall thickness and pre-LOCA oxidation.

9. Post-quench ductility and toughness are determined primarily by the thickness and the mechanical properties of transformed-beta layer. The mechanical properties are strongly influenced by several factors such as: oxygen solubility in beta, concentrations of alpha- (tin and oxygen) and beta-stabilizing elements (niobium and hydrogen), the nature of beta-to-alpha-prime transformation, redistribution of oxygen, niobium, and hydrogen during the transformation, and precipitation of hydrides. Significantly large hydrogen uptake can occur in some types of fuel cladding, during normal operation to high burnup, during breakaway oxidation at <1120°C, and, for localized regions near a rupture opening, during LOCA transients. Hydrogen uptake and its effect on the properties of transformed beta could differ significantly in Zircalloys and in niobium-containing alloys. Considering these factors, it is recommended to obtain a better understanding of the effects of more realistic hydrogen uptake and niobium addition on the properties of transformed-beta layer and post-quench ductility.

Acknowledgments

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References


37. Haggag, F. M., Zircaloy-Cladding-Embrittlement Criteria: Comparison of In-Pile and Out-of-Pile Results, NUREG/CR-2757
2. NRC Program for Addressing Effects of High Burnup and Cladding Alloy on LOCA Safety Assessment
Ralph Meyer, US NRC

Paper summary

In 1993, NRC initiated several small research efforts to investigate the effects of high-burnup fuel operation on regulatory methods and criteria. In 1994, results from Cabri in France and the Nuclear Safety Research Reactor in Japan demonstrated some clear effects for reactivity accidents, and the possibility of other effects was soon recognized. After several years of planning, NRC issued an "Agency Program Plan for High-Burnup Fuel" on July 6, 1998, in which it was observed that it is likely that the criteria and models for analysis of loss-of-coolant accidents (LOCAs) will be affected at high burnup. A program was put in place at Argonne National Laboratory to investigate LOCA effects, and this program is described below in a presentation by Chung. This work is being performed with active co-operation from the Electric Power Research Institute.

A short time later, NRC issued Information Notice 98-29 (August 3, 1998) to describe a specific instance of predicted increase in fuel rod cladding oxidation. In that information notice, the NRC stated that total oxidation, as mentioned in 10 CFR 50.46 (acceptance criteria for LOCA analysis), includes both pre-accident oxidation and oxidation occurring during a LOCA. This statement clarified the meaning of total oxidation and has become a de facto NRC position, subject to future modification that might come as the result of further research. The requirement to add together the pre-accident oxidation (corrosion) and the oxidation occurring during a LOCA was seen by some as overly conservative and led to additional emphasis on research efforts to address LOCA effects.

To help improve NRC's efforts to address high-burnup effects, a series of meetings was held with fuel experts to develop Phenomenon Identification and Ranking Tables (PIRTs) for three types of accident scenarios, one of which was the LOCA. Detailed information on the PIRT meetings and resulting reports can be found at www.nrc.gov/RES/pirt. During one of these meetings in 2000, Georges Hache presented a review of early LOCA regulatory decisions (see the presentation above). This presentation highlighted the importance of post-LOCA ductility, a fact that had been largely forgotten during the last 25 years. An immediate result of this presentation was the proposed modification of NRC's research program at Argonne National Laboratory to include such tests in the ongoing work.

Shortly after the presentation by Hache, some earlier work in Eastern Europe on post-LOCA ductility of Zr-1%Nb cladding came to the attention of IPSN in France and NRC in the U.S. This work appeared to show a substantial reduction of post-LOCA ductility compared with the Zircaloy alloys used in the West. However, a Zr-1%Nb alloy had been recently introduced by Framatome in the U.S. (M5 alloy), and the Westinghouse ZIRLO alloy that was introduced in the 1980s also contains about 1%Nb. Consequently, there was a strong interest in this subject in the East and the West, and post-LOCA ductility became the central focus of the present meeting that was then being organized.

In February, 2001, the NRC held meetings with Framatome and Westinghouse to discuss this subject. Both showed data for their new alloys that were in agreement with earlier results on Zircaloy, although the conflict with the Eastern European data was not explained (see presentations below by Lebourhis and Leech). At these meetings, NRC also proposed new co-operative work with Framatome and Westinghouse to address the high-burnup issues for the new cladding types. This proposed work would include post-LOCA ductility measurements on unirradiated M5 and ZIRLO tubing, respectively, as well as broader testing of high-burnup fuel rods with these cladding alloys at a later time. The testing that was outlined was similar to that underway with Zircaloy cladding alloys at Argonne National Laboratory. It was
proposed that this work be done by NRC with the co-operation of EPRI, Framatome, and Westinghouse, with due respect for proprietary considerations.

In addition to the research being conducted at Argonne National Laboratory, the NRC has other LOCA-related research on which it is depending. Part of it is conducted by other organizations and results are made available to NRC through agreements.
NRC PROGRAM FOR ADDRESSING EFFECTS OF HIGH BURNUP AND CLADDING ALLOY ON LOCA SAFETY ASSESSMENT

Ralph Meyer
Office of Nuclear Regulatory Research

OECD Topical Meeting on LOCA Fuel Safety Criteria
Aix-en-Provence
March 22-23, 2001
DEFINING EVENTS

- Agency Program Plan for High-Burnup Fuel (July 6, 1998)
- NRC Information Notice IN-98-29 (August 3, 1998)
- Phenomenon Identification and Ranking Tables (PIRTs 2000)
- NRC Meetings with Framatome and Westinghouse (February 2001)
SCAPE OF WORK ON HIGH-BURNUP ISSUES AT ARGONNE
(PIRT Adjusted, EPRI Cooperation)

- Testing in Current ANL Program for Zircaloy-2 and Zircaloy-4 (Target 2003)
  - Integral Test (Ballooning, Rupture, Oxidation, Quench — with Fuel)
  - Oxidation
  - Thermal Shock (to be determined)
  - Phase Relations
  - Mechanical Properties (including Post-Quench Ductility)
  - Post-LOCA Seismic Loading
  - Fuel Relocation (limited to Observation during Integral Test)

- NRC is Interested in Conducting Confirmatory Tests on ZIRLO and M5

- May only need Subset of Tests for Other Cladding Types like ZIRLO and M5
  - Oxidation
  - Thermal Shock (to be determined)
  - Phase Relations
  - Mechanical Properties (including Post-Quench Ductility)
PROPOSED WORK ON UNIRRADIATED ZIRLO AND M5
(Target 2001)

- Review All Test Methods to Determine Test Conditions (Zircaloy Specimens first)
- Agreement on Test Conditions will involve EPRI, Westinghouse, and Framatome
- Post-Quench Standard Test (perhaps Axial Tensile Test) on Unirradiated Cladding
- Post-Quench Ring-Compression Tests (probably also) on Unirradiated Cladding
- Oxidation Rate and Phase Relations as needed to interpret Ductility Results
- No Mechanical Properties or Other Testing at this Time (later in High Burnup Program)
- Proprietary Treatment of Data may be arranged if Requested
PROPOSED COOPERATION

- Pattern after Current ANL Program with EPRI Cooperation
- Westinghouse and Framatome would be Included in all Test Planning
- EPRI is also Interested in further Cooperation (Subject to Approval of RFP)
- Once Agreement is Reached, Start Unirradiated Testing in 2001 and Irradiated Testing in 2003
OTHER "NRC" WORK

- Halden’s LOCA Test and General Thermal Properties Work
- JAERI’s LOCA-Related Research on Thermal Shock and Hydrogen Effects
- RRC-Kurchatov Institute’s Mechanical Properties under LOCA Conditions
- PNNL’s FRAPTRAN Code Development includes LOCA Models
- Penn. State University’s Consulting and Related Work
Discussion:

Question by G. Hache: Results of testing of high-burnup fuel rods with new cladding alloys depend on the type of test. What type of test you consider the most representative for real reactor conditions?

Answer by R. Meyer: I don’t really know what would be the most representative test for real reactor conditions, but we will discuss the preferred type of tests with our consultants before performing any tests on new cladding alloys in our program at Argonne National Laboratory. We will probably perform post-quench ring-compression tests to have a basis for comparing with the existing body of data, and we may also perform some other post-quench test like an axial-tensile test or a four-point bend test, but this will be decided later.

Question by N. Waeckel: Looking back to 1973 conclusions from hearings: We are trying to analyze the initial writing with today’s knowledge including the burnup effect, the in-reactor corrosion impact, etc. By doing so we may distort the initial intent of the staff. In addition, relying on the 1973 statements literally maybe wise from a legal point of view but doesn’t reflect the progress made since then. My question is: is there room to address “the unknown process that may take place during a LOCA” (expressed by the staff in 1973) in a different way than the one proposed in 1973? Can we use the improvements in knowledge, experience feedback and modeling we have gained during the last 30 years to address, for instance, the unknown process the staff referred to in 1973? For example the direction and the magnitude of the axial forces during quench or the intensity of the load resulting from a post quench seismic event or the type of post-quench mechanical tests that should be used to assess the residual ductility, etc.

Answer by R. Meyer: I cannot say what would or would not be accepted if submitted to NRC for a decision, but I do have an opinion about the commission’s conclusions in 1973 and the applicability of that conclusion today. The commission stated that “it may not be possible to anticipate and calculate all of the stresses to which fuel rods would be subjected in a LOCA” and I believe that is still true today. While it may be possible to calculate some of those stresses better today than it was then, I don’t think there has been a breakthrough that would give us confidence in knowing the nature of all the stresses that would be encountered. In this meeting we have mentioned the possibility of stresses arising from bonding between pellets and cladding at high burnup, and this is one more type of stress that would have to be considered. It seems to me that retention of ductility is still the best way of ensuring that fuel rods will remain intact during a LOCA. However, I do think there is room to determine the best measure of ductility, and that might not be a ring-compression test.
3. Ring-Compression Test Results and Experiments Supporting LOCA PCT, Oxidation and Channel Blockage Criteria.
Laszló Maróti, AEKI, Budapest, Hungary

Paper summary

The four VVER-440/213 type units of the Hungarian NPP apply Russian fuel where the cladding is made of Zr1%Nb. The operational experiences revealed the good quality of the fuel. The number of inhermetic elements was low during long period of operation.

At the same time, data on the accidental behaviour of the fuel were scarce. For the investigation of the high temperature behaviour of the Zr1%Nb cladding material an experimental program was launched in 1991.

In the framework of the project different kind of experimental investigations were carried out. The most important tests related to fuel LOCA criteria were the following:

- ring compression tests,
- quench tests,
- bundle ballooning tests.

Comparison of Zr1%Nb and Zry-4 ring compression tests showed stronger embrittlement of Zr1%Nb after high temperature steam oxidation. This behaviour of the Zr1%Nb is likely due to the higher H₂ uptake during the oxidation.

Quench tests on 5 cm long fuel pieces confirmed the conservatism of the Russian 18% ECR criterion.

It cannot be expected that the flow channel blockage in a VVER-440 type fuel assembly exceeds 80% during the LOCA process. So the coolability is not endangered.

Introduction

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In the framework of the project different kind of experimental investigations were carried out. The most important tests related to fuel LOCA criteria were the following:

- ring compression tests,
- quench tests,
- bundle ballooning tests.
Subsequently the results of the series of experiments are summarised in short.

**Ring compression tests**

In Ref. [1] German experts published ring compression test results showing much stronger embrittlement of the Zr1%Nb than Zry-4 as a result of high temperature steam oxidation.

It has been decided in the Atomic Energy Research Institute (AEKI) to perform reproducibility tests with available Russian cladding material.

The selected steam oxidation temperatures were 900, 1000, 1100 and 1200°C. The oxidation was made in a tube furnace and the steam supply was unlimited. The compression test was executed by means of a tensile machine and both the deformation and the compression forces were recorded. Generally two or three cracks were detected but always the first crack was considered as tube failure.

The Hungarian results were in good agreement with that obtained in Germany and revealed complete embrittlement of the Zr1%Nb at about 7-8% ECR while the same appeared at 17-20% ECR in case of Zry-4.

To clarify the role of H₂ and O₂ in the embrittlement process further experiments were conducted where the oxidation and hydrogen filling were separated. The oxidation was made in Ar+O₂ mixture at 900°C then samples with different degree of oxidation were filled with H₂. The temperature of the H₂ absorption was 900°C.

The ring compression tests performed on the above samples showed the following results. The H₂ uptake without oxidation (ECR=0) resulted immediately the embrittlement of the material depending on the H₂ content. The oxidation up to about 5% ECR did not cause any significant embrittlement. In general the two materials (Zr1%Nb, Zry-4) behaved similarly especially at higher values of the H₂ content.

The conclusion that could be drawn after the test was that the H₂ uptake during steam oxidation must be higher at the Zr1%Nb. Measurement of the H₂ content in samples oxidised in steam was subsequently executed and the results proved this conclusion of the ring compression experiments.

**Quench tests**

Based on the ring compression test results there was a doubt whether the 17% ECR oxidation LOCA criterion could be used for the Zr1%Nb. There was an objection saying that the ring compression test is not representative for the LOCA conditions therefore the decision was made to carry out quench tests for the clarification of the applicability.

During the tests samples were oxidised at constant temperature then reaching the required value of ECR they were dropped into cold water and after cooldown it was examined whether the tube pieces are intact or failed.
The main data of the tests are listed below.

Oxidation temperatures: 1000, 1100, 1200, 1250°C
Oxidation time range: 0 - 240 min.
Length of the samples: 50 mm
Filling of the tubes: 10 samples empty
  11 samples filled with single Al₂O₃ pellet
  3 samples filled with 3 Al₂O₃ pellets

At the evaluation of the test results the ECR value was calculated by the equation validated for the Zr1%Nb:

\[ \Delta m = 920 \cdot \exp \left( -\frac{10410}{T} \right) \cdot \sqrt{t} \]

\[ \Delta m = \text{weigh gain, mg/cm}^2 \]
\[ T = \text{temperature, K} \]
\[ t = \text{oxidation time, s} \]

From the results of the quench tests one could conclude that for the Zr1%Nb both the Hungarian 17% or the Russian 18% criterion was conservative until there was no direct force acting on the samples beyond the stress caused by the quench process.

Ballooning and blockage tests

It is known that the relatively small fuel assembly of the VVER-440 type reactor is surrounded by a closed shroud. This shroud tube does not allow the cooling above a blockage by crossflow from the surrounding bundles. Therefore the degree of blockage developing in a LOCA is especially important. For the investigation of the problem ballooning experiments were performed in AEKI.

The test conditions can be summarised as follows:

Geometry: 7-rod bundle
Bundle length: 15 cm
Cladding material: Zr1%Nb
Initial filling pressure: 3 - 30 bar
Temperature increase: linear

During the tests the temperature and pressure histories were recorded. Each test terminated when the last rod failed. After the test cuts of the bundle were prepared at the place of the maximum coolant channel blockage and the area of the remaining cross section was measured.

As a final conclusion it was possible to state that in most of the cases the flow-cross-section reduction was in the range of 40-50% and its value never exceeded 80%.
Summary

Comparison of Zr1%Nb and Zry-4 ring compression tests showed stronger embrittlement of Zr1%Nb after high temperature steam oxidation. This behaviour of the Zr1%Nb is likely due to the higher H₂ uptake during the oxidation.

Quench tests on 5 cm long fuel pieces confirmed the conservatism of the Russian 18% ECR criterion.

It cannot be expected that the flow channel blockage in a VVER-440 type fuel assembly exceeds 80% during the LOCA process. So the coolability is not endangered.

Ring-Compression Test Results and Experiments Supporting

LOCA PCT, Oxidation and Channel Blockage Criteria

presented by

L. Maróti

prepared for SEG FSM LOCA Topical Meeting
2001. March
INTRODUCTION

- The four units of the Paks NPP were put into operation in the following years: 1983, 1984, 1986, 1987.
- That time limited fuel data were available. Not enough to perform detailed fuel behaviour analysis.
- At the same time the Soviet vendor guarantied the operational safety of the fuel in VVER-440 nominal conditions.
- Operational experiences proved the good quality of the fuel elements and assemblies. The number of hermetic fuel was very low during long period of operation.
- However, the question arose how this fuel behaves under accidents.
- In 1991 an experimental program was launched to investigate the Zr1% Nb cladding material at high temperatures.
Formulation of the problem

The licencing criteria concerning the fuel behaviour during the postulated accident large break LOCA contain the following limits:

- temperature of the cladding less than 1200°C
- local oxidation of the cladding surface in the core less than 17%
- core coolability should be preserved
Ring compression tests with preoxidised Zr1%Nb and Zircaloy-4 claddings
For clarification of the effect of H₂ content samples were made where oxidation and H₂ absorption was separated.

**Oxidation:**

\[ \text{Ar} + \text{O}_2 \quad \text{mixture} \]

**temperature:** 900 °C

**H₂ filling:**

pure H₂ gas

**temperature:** 900 °C
Zircaloy cladding

Relative deformation [%]

Equivalents oxidation [%]

H=0
H=30 ppm
H=100 ppm
H=300 ppm
H=700 ppm

newcrushing.xls
Zr1%Nb
H-content of metal phase after steam oxidation
Zr1%Nb (10μm oxid, 300ppm H₂)
Zircaloy-4 (10μm oxid, 300 H₂)
Problem 1

Ring compression test data showed stronger embrittlement of the Zr1%Nb cladding and
the question arose: is 17% oxidation allowable for Zr1%Nb?

Problem 2

Experimental data were not available for the ballooning behaviour of Zr1%Nb and
the problem of coolability could not be answered.
Experimental procedure
and results of the quench test
for checking the 17% criterion
Quench test facility
Main data of the tests

- Oxidation in steam
- Oxidation temperature before quenching — 1000 - 1100 - 1200 - 1250°C
- Oxidation time range — 0 - 240 min.
- Type and number of samples — length 50 mm
  - empty tubes 10 samples
  - filled with single Al₂O₃ pellet 11 samples
  - filled with four Al₂O₃ pellets 3 samples

Correlation used for the calculation of ECR value from temperature and oxidation time (Solyany)

\[ \Delta m = 920 \times \exp\left(-\frac{10410}{T}\right) \times \sqrt{t} \]

\[ \Delta m \] - weight gain, mg/cm²
\[ T \] - temperature, K
\[ t \] - oxidation time, s
Temperature, °C

Time of oxidation, s

18% ECR

1200°C

- intact
- failed

10^(-3) / T, 1/K

Thermal shock test results with Zr1%Nb cladding
Conclusion

- 17 or 18% ECR limit is conservative until no force acting on the cladding is supposed

- in Japanese experiments an additional tensile load is applied

the result is 15% limiting value of ECR
Ballooning and blockage
formation test
The clarification of the blockage problem is especially important in the VVER-440 because the closed shroud does not allow the cooling above the blockage by crossflow which is possible in an open core.

Conclusion of the test

- Mainly the flow-cross-section reduction is in the range of 40-50 %
- The maximum value of blockage ratio is less than 80 %
Test procedure

- The tests were performed with 7-rod bundles of hexagonal VVER geometry
- The cladding material was Zr1%Nb
- The bundle length was 15 cm
- The experiments applied linear temperature increase until failure of all rods
- Two test series were made: one in Ar and another in steam, to clarify the effect of steam oxidation
- The initial fill pressure was between 3 and 30 bars
Experimental arrangement
7-rod ballooning test in Ar,
inital pressure 20 bar
Ballooning Test № 8
Initial Pressure: 10 bar
Environment: Steam

Pressure and temperature history

Increase of rod cross sections and blockage rate of bundle.
Cross section of 7-rod bundle after test in steam, initial pressure 20 bar
Discussion:

Question by H.M. Chung:

Total oxidation in your investigation was calculated based on Solyany correlation. Were the results of calculation in good agreement with actual measured values?

Answer by L. Maroti:

The agreement between measurement results and data calculated by the Solyany correlation was satisfactory. Nonetheless, the uncertainty was never determined and the statement is valid only until the break-away effect accelerates the oxidation as one could see from experimental results that the scattering of the values was small except at the higher oxidations at 900 and 1000 °C where the break-away was evident...
4. Justification of the M5™ behavior in LOCA
A. Le Bourhis, Framatome, France

Paper summary

The work performed by Framatome ANP in the past 5 years on the M5 behaviour in LOCA (mechanical, oxidation, quench, embrittlement) shows that M5 alloy behaves at least in an identical way compared to Zircaloy-4 during the different phases of the accident.
This conclusion is obtained from experimental tests conducted in co-operation with EDF and CEA.
This paper is particularly focused on high temperature oxidation kinetics, quench and post-quench embrittlement.
In order to show that Zircaloy-4 criteria (PCT, ECR) and Baker-Just correlation can still be used for M5 alloy in the LOCA range (temperature and time), the approach adopted has been to provide a continuous behaviour comparison between both alloys and to be compatible with previous French programs (choice of the facility, operating conditions and test matrix).
The main results obtained so far can be summarised as follows:

oxidation tests
- oxidation tests performed between 700°C and 1400°C show that the approved "Baker-Just" model is conservatively applicable for M5,
- hydrogen content has a slight effect on the oxidation kinetics for hydrogen contents bounding the end of life values,
- at 1000°C and for an oxidation time of 3270s., both alloys offer equivalent β-Zr thickness.

quench tests
- quench tests performed between 1000°C and 1300°C on as-fabricated and pre-hydrided M5 tubes show that ECCS criteria (1204°C and 17% ECR) are comfortably fulfilled by the M5 alloy and can be applied for this alloy,
- For temperatures lower than 1100, M5 survives up to 2 times longer than Zircaloy-4

post-quench tests
- post-quench tests (three point bend, impact and ring compression) performed at room temperature on tubes oxidated at 1100°C with experimental ECR's in the range 3 to 17% show that both alloys have a similar behaviour,
- Consequently, M5 does not exhibit embrittlement within the limits of the ECCS criteria and results obtained by H. Bohmert on ZrNb1% are not applicable to M5.

conclusion
- Framatome ANP testing has sufficiently validated the current 50. 46 criteria as applicable to alloy M5.
- Peak cladding temperature: 1204°C, maximum cladding oxidation:17% and use of Baker-Just oxidation correlation can be applied for M5 justification in LOCA.
JUSTIFICATIONS OF THE M5™ BEHAVIOR IN LOCA

1 - ACCEPTANCE CRITERIA FOR ECCS EFFECTIVENESS

2 - FRAMATOME ANP LICENSING APPROACH FOR M5™ CLADDING IN LOCA CONDITIONS

3 - OXIDATION KINETICS

4 - QUENCH EMBRITTLEMENT

5 - POST QUENCH MECHANICAL TESTS

6 - CONCLUSION
1. ACCEPTANCE CRITERIA FOR ECCS EFFECTIVENESS

☐ ACCEPTANCE CRITERIA DEFINED IN 1973 AND BASED ON ZIRCALOY ALLOY

☐ CRITERIA
- PEAK CLADDING TEMPERATURE: 1204 °C
- MAXIMUM CLADDING OXIDATION: 17 %
- MAXIMUM HYDROGEN GENERATION: 1 %
- COOLABLE GEOMETRY
- LONG TERM COOLING

☐ USE OF THE BAKER - JUST OXIDATION CORRELATION FOR COMPLIANCE OF CRITERIA
2. FRAMATOME ANP LICENSING APPROACH FOR M5™ CLADDING IN LOCA CONDITION

- THE M5™ ALLOY BELONGS TO THE ZIRCONIUM BASED ALLOY FAMILY
- CONTINUOUS BEHAVIOR COMPARISON WITH ZIRCALOY 4
- DOMAINS OF JUSTIFICATION OF THE BEHAVIOR OF THE M5™ ALLOY
  - SPECIFIC THERMOMECHANICAL MODEL
  - HIGH TEMPERATURE OXIDATION AND EMBRITTLEMENT
- FACILITY AND TEST MATRIX DEVELOPED TO BE COMPATIBLE WITH PREVIOUS FRENCH PROGRAMS
- VERIFICATION OF RESIDUAL DUCTILITY FOLLOWING THE QUENCH
- SHOW THAT ZIRCALOY-4 CRITERIA (PCT, ECR) AND BAKER JUST CORRELATION BE USED FOR M5™ ALLOY IN THE LOCA RANGE
2. FRAMATOME ANP LICENSING APPROACH FOR 
M5™ CLADDING IN LOCA CONDITION

M5™ LOCA BEHAVIOR: EXPERIMENTAL PROGRAMS

- M5™ TESTS PERFORMED IN THE FRAMEWORK OF A 
  COOPERATION AMONG: FRAMATOME, EDF AND CEA

- THERMAL MECHANICAL BEHAVIOUR: EDGAR MODEL 
  PHASE TRANSFORMATION TEMPERATURE AND KINETICS 
  CREEP AND THERMAL RAMP TESTS

- OXIDATION AND QUENCHING EMBRITTLEMENT: CINOG 
  TESTS OXIDATION KINETICS 
  TIME TO FAILURE DURING QUENCHING TESTS

- MECHANICAL BEHAVIOR AT ROOM TEMPERATURE AFTER 
  OXIDATION AND QUENCHING

- EFFECT OF IRRADIATION: ANALYTICAL TESTS 
  AS MANUFACTURED, PRE-HYDRIDED AND IRRADIATED MATERIALS
3. OXIDATION KINETICS

OXIDATION 700 TO 1400°C Zr-4 and M5™

- M5™ Oxidation Less Than Zr-4 From 900 to 1100°C
Zy4 METALLOGRAPHIC OBSERVATIONS AFTER OXIDATION 1000°C - 3270 SECONDS

Trace of spalling on inner and outer zirconia layers
M5™ METALLOGRAPHIC OBSERVATIONS AFTER OXIDATION 1000°C - 3270 SECONDS

Outer layer

Inner layer

The inner and outer zirconia layers are homogeneous
No trace of spalling
OXIDATION KINETICS - CINO
Comparison with the results in the literature

- BAKER-JUST MODEL IS BOUNDING IN ALL ENCOUNTERED CONFIGURATION
- LESTIKOW MODEL ACCURATELY PREDICTS ZY4 RESULTS AND IS BOUNDING FOR M5™
3. OXIDATION KINETICS

M5™ LOCA BEHAVIOR: HIGH TEMPERATURE OXIDATION - ZY4/M5™ COMPARISON

- Zy4 VALUES ARE CONSISTENT WITH LITERATURE
- M5™ OXIDATION LOWER COMPARED TO Zy4 IN THE TEMPERATURE RANGE 900°C TO 1100°C
- SLIGHT EFFECT OF HYDROGEN CONTENT (UP TO 450 ppm) ~ 10 %
- BAKER JUST CORRELATION IS BOUNDING ENCOUNTERED CONFIGURATIONS AND CAN BE USED FOR LOCA LICENSING PURPOSE FOR M5™
- 1000°C - 3270s: BOTH ALLOYS OFFER EQUIVALENT β-Zr THICKNESS
<table>
<thead>
<tr>
<th>Layer Description</th>
<th>ZIRCALOY-4</th>
<th>M5&lt;sup&gt;TM&lt;/sup&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>External Zirconia Layer (μm)</td>
<td>55.2 to 61.0</td>
<td>18.9 to 20.3</td>
</tr>
<tr>
<td>External α Zr-O Layer (μm)</td>
<td>53.9 to 71.6</td>
<td>53.9 to 61.9</td>
</tr>
<tr>
<td>β&lt;sub&gt;ZR&lt;/sub&gt; Layer (μm)</td>
<td>351 to 379</td>
<td>394 to 409</td>
</tr>
<tr>
<td>Internal α Zr-O Layer (μm)</td>
<td>53.5 to 70.4</td>
<td>47.4 to 57.3</td>
</tr>
<tr>
<td>Internal Zirconia Layer (μm)</td>
<td>48.7 to 55.8</td>
<td>19.0 to 21.7</td>
</tr>
</tbody>
</table>
4. QUENCH EMBRITTLEMENT : MAIN RESULTS (1/2)

<table>
<thead>
<tr>
<th>ALLOY</th>
<th>OXIDATION TEMPERATURE(°C)</th>
<th>TIME TO FAILURE (S)</th>
<th>ECR FAILURE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zy4</td>
<td>1000</td>
<td>6500</td>
<td>22*</td>
</tr>
<tr>
<td></td>
<td>1100</td>
<td>2970</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td>1200</td>
<td>950</td>
<td>29</td>
</tr>
<tr>
<td></td>
<td>1300</td>
<td>390</td>
<td>29</td>
</tr>
<tr>
<td>M5™</td>
<td>1000</td>
<td>13500</td>
<td>16 *</td>
</tr>
<tr>
<td></td>
<td>1100</td>
<td>2959</td>
<td>28</td>
</tr>
<tr>
<td></td>
<td>1200</td>
<td>1200</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td>1300</td>
<td>495</td>
<td>31</td>
</tr>
</tbody>
</table>

* conservative value
4. QUENCH EMBRITTLEMENT : MAIN RESULTS (2/2)

- M5\textsuperscript{TM} and Zy-4 HYDROGEN UPTAKE AFTER QUENCH TESTS IS LOW IN THE LOCA RANGE

- M5\textsuperscript{TM} ACCIDENT SURVIVAL IS SUPERIOR TO Zy-4

  T > 1100\degree C M5\textsuperscript{TM} AND Zy-4 HAVE SIMILAR SURVIVAL ABILITY

  T < 1100\degree C M5\textsuperscript{TM} SURVIVES UP TO 2 TIMES LONGER THAN Zy-4

- AT MODERATE TEMPERATURES (900\degree C < T < 1100\degree C) M5\textsuperscript{TM} REQUIRES EXCESSIVE OXIDATION TIMES TO ACHIEVE ECR NEAR 17 \%
5. POST-QUENCH MECHANICAL TESTS

TEST MATRIX

OXIDATION
- T = 1100°C
- t → ECR = 3, 6, 10 AND 17% (OXIDA MODEL WITH LESTIKOW LAW)
- SINGLE FACE OXIDATION
- AS-FABRICATED M5™ AND ZY-4 CLADDING TUBES

WATER QUENCH

MECHANICAL TESTS
- THREE POINT BEND
- IMPACT
- RING COMPRESSION
OXIDATION - DEVICE

- Set Pin
- Support Ring
- Safety Valve
- Steam Boiler
- Internal Alumina Tube
- Furnaces
- SAMPLE
- External Alumina Tube
- Cooling Annular Device
- High Temperature Oxidation + Quenching Device
  1700°C - max.

White Tissue
Cooling Bath
Slag Wool
PERCENT OF SPALLED OXIDE AFTER OXIDATION AT 1100 °C AND QUENCH FOR THE LONGEST EXPOSURE TIME (3098 - 3800 S)

<table>
<thead>
<tr>
<th>Alloy</th>
<th>Oxide Spalled (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zr-4</td>
<td>66.9</td>
</tr>
<tr>
<td></td>
<td>64.6</td>
</tr>
<tr>
<td></td>
<td>83.2</td>
</tr>
<tr>
<td>M5</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>2.2</td>
</tr>
<tr>
<td></td>
<td>3.9</td>
</tr>
</tbody>
</table>
METALLOGRAPHIC OBSERVATIONS OF LOW-TIN Zr-4 AFTER OXIDATION AT 1100°C

\[ t = 1349 \text{ s and quenched} \]

- \( \alpha\)-Zr(0) layer: large \( \alpha \)-grains
- \( \alpha\)-Zr(0) layer: cracks
METALLOGRAPHIC OBSERVATIONS OF M5™
AFTER OXIDATION AT 1100°C
\[ t = 3600 \text{ S AND QUENCHED} \]

- \( \alpha \text{ Zr (O) layer} \): Linear distribution of niobium particles in \( \alpha \) platelets
- \( \alpha \text{ Zr (O) layer} \): no cracks
POST-QUENCH MECHANICAL TEST
3 POINT BEND TEST APPARATUS

Starting position

7.5 mm displacement
POST-QUENCH MECHANICAL TEST
3 POINT BEND TEST RESULTS

➢ M5™ and Zy4 behave similarly
POST-QUENCH MECHANICAL TEST
RING COMPRESSION TEST

Starting position
POST-QUENCH MECHANICAL TEST RING COMPRESSION TEST RESULTS

- M$^{\text{TM}}$ behaves slightly better than Zr-4
POST-QUENCH MECHANICAL TEST
IMPACT TEST RESULTS

- $\text{M5}^{\text{TM}}$ behaves slightly better than Zr-4

![Graph showing resilience vs. weight gain for Zr-4 and M5](image)
CONCLUSION
POST-QUENCH MECHANICAL TESTS

☐ M5™ PERFORMED BETTER THAN OR SIMILAR TO ZY-4

- NO DELAMINATION
- SIMILAR BEND TEST RESULTS
- SLIGHTLY BETTER IMPACT TEST RESULTS
- SLIGHTLY BETTER THAN ZY-4 IN RING COMPRESSION TESTS
CONCLUSIONS
M5™ TEST PROGRAM

☐ HIGH TEMPERATURE OXIDATION PERFORMANCE OF M5™ IS EQUIVALENT OR SUPERIOR TO ZY-4

☐ AT TEMPERATURES BETWEEN 900 AND 1100 °C, M5™ REQUIRES EXCESSIVE OXIDATION TIMES TO ACHIEVE ECR’S NEAR 17%

☐ M5™ ACCIDENT SURVIVAL IS SUPERIOR TO ZR-4
  - T>1100 °C M5™ AND ZR-4 HAVE SIMILAR SURVIVAL ABILITY
  - T<1100 °C M5™ SURVIVES UP TO 2 TIMES LONGER THAN ZY-4
CONCLUSIONS
M5™ TEST PROGRAM

☐ FRAMATOME ANP TESTING HAS SUFFICIENTLY VALIDATED THE CURRENT 50.46 CRITERIA AS APPLICABLE TO ALLOY M5™

☐ PEAK CLADDING TEMPERATURE: 1204°C, MAXIMUM CLADDING OXIDATION: 17%, USE OF BAKER-JUST OXIDATION CORRELATION, CAN BE RETAINED FOR M5™ JUSTIFICATION IN LOCA
Discussion:

Question by H.M. Chung: It appears that in addition to the effect of oxidation, hydrogen uptake larger than a certain threshold level plays an important role in the degradation of post-quench ductility of E110 alloy. What were the hydrogen contents of the M5 specimens you tested?

Answer by A. Le Bourhis: Hydrogen content determinations were conducted in support of the mechanical tests; the hydrogen uptake was ~ 20ppm.

Question by L. Belovský: What was the colour of the oxide grown on the M5 alloy in the high-temperature oxidation tests, namely after 3270 s at 1000 °C?

Answer by A. Le Bourhis: The colour of the oxide on the M5 alloy in the high temperature oxidation tests at 1000°C and 3270 s was black.

Question by M. Valach: Could you kindly comment on differences between Zry and M5™ at T=1000 deg C in your table 4 "Quench Embrittlement: Main Results"? It’s really interesting, that for the higher temperatures both alloys exhibit nearly similar results.

Answer by A. Le Bourhis: 1000°C is the zone of the breakaway for long oxidation time and zirconium base alloys. It appears that at this temperature, the time to failure for the M5 is much longer than for the Zircaloy 4. However it is worth to note that at this temperature of oxidation, for both alloys the time to rupture is greater than the duration of a LOCA.

Question by R. Meyer: In your opinion, why are your results different from Boehmert’s results?

Answer by A. Le Bourhis: Results look different. We know the technical specifications of the tested M5 alloy and the followed test procedure which is similar to the IPSN one. This is not the case for the alloy which was tested 10 years ago in Germany as also the test procedure. In consequence for the time being we have no comments on the interpretation of these tests.

Question by R. Yang: Were the tests performed with irradiated claddings or just with as-fabricated?

Answer by A. Le Bourhis: Tests presented were performed with as-fabricated claddings.

Question by N. Sokolov: What are the α→β transient temperatures, I mean the temperature of beginning α to α+β transformation and the temperature of completing α+β to β transformation, for M5?

Answer by A. Le Bourhis: The equilibrium (α,α+β) phase transformation temperature (for a few % of beta phase) is 760°C and (α+β,β) temperature is 950°C. These temperatures are varying with dynamic effects (heating, cooling rates).

Question by G. Hache: Have you performed quench ductility tests above 1200 °C in order to better understand solid-solution hardening at high oxygen concentrations?

Answer by A. Le Bourhis: We did not perform quench ductility above 1200°C.
5. Ductility Testing of Zircaloy-4 and ZIRLO™ Cladding After High Temperature Oxidation in Steam
W.J. Leech, Westinghouse Electric Co., USA

Paper summary

The purpose of this presentation was to give an overview of current Westinghouse efforts on ductility testing of Zircaloy-4 and ZIRLO cladding, after high temperature oxidation in steam. This is an ongoing program, which was started in late January of this year and will be completed in May. The overall program is described and some preliminary data are presented.

Licensing of ZIRLO cladding, a proprietary Westinghouse alloy, was initiated in 1991. An extensive testing program was performed to support the licensing application. Data were obtained for material mechanical properties, density, thermal expansion, thermal conductivity, specific heat, phase changes, high temperature creep, high temperature oxidation, and rod burst characteristics. The data showed that except for phase change temperatures, the properties of ZIRLO and Zircaloy-4 were essentially equivalent. It was argued that because of the close similarity of ZIRLO and Zircaloy-4 properties, the 10 CRF 50.46 criteria also applied to ZIRLO. The NRC agreed, and 10 CFR 50.46 was amended to state that the criteria also applied to ZIRLO.

During January of this year we became aware of NRC concerns relative to the applicability of the 17% ECR criterion for Zircconium - 1% niobium alloys. These concerns were based on tests conducted by Böhmert, and others, on the Russian E110 alloy, which is a Zirconium-1% niobium alloy. Böhmert conducted high temperature steam oxidation tests on both alloy E110 and Zircaloy-4. He found that the limiting ECR for completely brittle behavior was about 1/3 the value for Zircaloy-4. Although both ZIRLO and alloy E110 contain 1% niobium, they are not equivalent. ZIRLO also contains 1% Tin, 0.1% Iron, and 1200 ppm Oxygen versus about 700 ppm in E110. Both tin and oxygen are alpha phase stabilizers and raise the alpha to beta phase transition temperature relative to Zr-Nb binary alloys. There are significant differences of the oxide layer structure reported for the E110 alloy and those observed for either ZIRLO or Zircaloy4.

However, the information referred to by the NRC convinced Westinghouse to conduct further tests to verify that the 17% ECR criterion applied to ZIRLO. A test program was initiated in late January and is still underway.

A test apparatus with a test section, consisting of an inconel tube containing the specimen is heated in a clam shell resistance furnace was used. The test section contains both ZIRLO and Zircaloy test samples of open ended tubing about 1.25 inches long and are placed in the constant temperature zone, one above the other. Locations are alternated between tests. Steam flows through the test section. Final temperatures are in the range of 1800°F to 2200°F and are held at times ranging from 5 to 30 minutes. The goal is to produce specimens with a range of ECRs for subsequent evaluations. A set of specimens has been prepared and is being evaluated. Test conditions were chosen to produce a range of ECR values.

The condition of the oxide layer itself is of major interest. Significantly different oxide layer structures have been reported for Alloy E110 and Zircaloy-4.

Ring compression tests are being conducted on all samples to assess the cladding ductility. Tests are being conducted at room temperature and at a temperature of 275°F. The tests are similar to those performed by Hobson and Rittenhouse (ORNL 4758) and Böhmert. Ring sections are cut from the tubing specimens and compressed in the radial direction. Load-displacement curves are obtained and used to
Ductility Testing of Zircaloy-4 and ZIRLO™ Cladding After High Temperature Oxidation in Steam

W. J. Leech
Westinghouse Electric Company
Columbia, SC USA

Topical Meeting on LOCA Fuel Safety Criteria

Aix-en-Provence, France
March 22, 2001
Specimen Preparations

- Isothermal steam oxidation
  - ZIRLO and Zircaloy-4 cladding samples prepared simultaneously
  - Heat up in a resistance furnace
  - Flowing steam
  - Temperatures: 1800 - 2200°F
  - Times: 5 - 30 minutes
  - Radiation and Convection Cooling
  - No specimen quenching
  - Goal is to produce specimens with a range of ECR for subsequent evaluation.
Specimen Evaluations (In Progress)

- Specimen Evaluations
  - Oxide Layer Characteristics
  - Ring compression tests
    - Assess cladding ductility.
    - Room temperature and 275°F.
    - Test performed similar to Hobson & Rittenhouse (ORNL Report 4758) and Böhmert.
  - Optical metallography
    - Oxide thickness, $\alpha$-stabilized layer, transformed-β layer.
    - Microhardness to assess oxygen penetration.
Comparison of ZIRLO™ and Zircaloy-4 Sample Weight Gains

![Graph showing comparison of ZIRLO™ and Zircaloy-4 sample weight gains.](image)
Measured Oxide Thickness vs. Oxide Thickness Based on Weight Gain

Oxide Thickness Based on Weight Gain, microns

Measured Oxide Thickness, microns
Relative Displacement at Fracture vs Measured ECR at a Temperature of 275F (PRELIMINARY)
Comparisons of ZIRLO™ and Zircaloy-4

- Both oxide layers were black, shiny, adherent, and with no laminations
- Both have similar fractions of oxide in the oxide layer and in the metal
- Ring compression tests show similar values of displacement at fracture versus the measured Equivalent Cladding Reacted
- ZIRLO™ and Zircaloy-4 exhibit similar behavior and the 17% oxidation criteria is conservative for both alloys
Discussion:

Question by G. Hache: Did you measure hydrogen uptake?

Question by H.M. Chung: In addition to total oxidation, did you measure alpha- and prior-beta-layer thicknesses? Were the phase layer thicknesses significantly different from those measured on Zircaloy-4 for similar exposure?

Question by R. Meyer: Do you plan to do tests with oxidation done in the range of 900-1200 C?

Answer by W. J. Leech: 1800-2200 F is the range we're covering, but we will take the comment under advisement and will probably go down to 900 C.

* The answers will be provided in the Proceedings of the Topical Meeting
6. Progress in ANL/USNRC/EPRI Program on LOCA
Hee Chung, R.V. Strain, T. Bray, M.C. Billone, Argonne Nat. Lab., USA

Paper summary

A hot-cell testing program is being conducted in Argonne National Laboratory to investigate the performance of high-burnup fuel under loss-of-coolant-accident (LOCA) situations. The primary objectives of the integral program are to determine the LOCA-related behavior at high burnup, such as high-temperature oxidation kinetics, ballooning and burst, fuel relocation and release in the burst section, quenching thermal-shock failure, post-quench ductility, and bending response to seismic forces. In-cell integral LOCA facilities are being constructed and initial tests are being performed on low-Sn Zircaloy-4 PWR fuel (burnup %<50 GWD/t), Zr-lined Zircaloy-2 BWR fuel (burnup %<57 GWD/t), standard Zircaloy-4 PWR fuel (burnup %<70 GWD/t), and their unirradiated counterparts. Microstructural characteristics and initial results on the oxidation behavior of the BWR fuel have been determined. Hydroxide structure of the BWR fuel is characterized by clusters of small hydrides in the clad midwall, which appears to be related to irradiation-induced amorphization of the second phase precipitates. The results of the tests on oxidation kinetics are consistent with Cahn-Car-Pawel correlation rather than Baker-Just correlation. However, noticeable susceptibility of the BWR cladding to locally enhanced oxidation at the outer surface has been observed. Further investigations are being conducted to determine if this observation is related to test procedure(s) or inherent material property of the highly irradiated cladding. The current LOCA criteria were established in 1973 primarily on the basis of post-quench ductility rather than resistance to quenching thermal shock. Therefore, a significant effort is being given to analyze the post-quench ductility and impact properties to identify the most meaningful test procedure(s), and at the same time, to provide a better understanding of the effects of high burnup and the composition of Nb-containing alloys. The focus of the analyses is the effect of hydrogen uptake and hydriding. Effects of large hydrogen uptake on impact-failure and ring-compression behavior of unirradiated Zircaloy-4 and Zr 1Nb (E110) have been analyzed. Three routes that lead to large hydrogen have been identified, i.e., (1) hydrogen uptake during normal (burnup-dependent) operation, (2) through unprotected (oxide-free) inner surface in the burst section, which is exposed to stagnant mixture of steam and hydrogen gas during a LOCA-like transient, and (3) through breakaway oxide that forms on the outer surface during a LOCA-like transient. The first route, strongly influenced by alloy type and burnup, is most difficult to avoid at high burnup. This route is not important for zero or low burnup. Post-quench ductility of all types of high-burnup fuel cannot be understood well as a function of oxidation only; it is important to evaluate and understand post-quench ductility as a function of total hydrogen uptake as well as oxidation. Large hydrogen uptake occurs in the ballooned and burst section in a pressurized Zircaloy-4. Large hydrogen uptake and significantly degraded tensile and impact properties of such sections were investigated extensively in the early 1980s. Post-quench ductility from ring-compression tests was found to be strongly influenced by not only oxidation but also hydrogen concentration in the ring. The threshold hydrogen concentration for severe hydrogen-induced embrittlement in Zircaloy-4 appears to be %<600 wppm. However, according to published data, similar threshold hydrogen concentration for E110 appears to be lower. This appears to be related with the beta-stabilizing effect of Nb. It is, therefore, suggested to gain a better understanding of the phenomenon of eutectoid decomposition (i.e., the rate of decomposition of beta phase into hydride and alpha phase, eutectoid temperature, and hydrogen concentration at the eutectoid composition) in Zircaloy-4 and Nb-containing alloys that contain high concentrations of hydrogen.
Primary Effects - Issues
High Burnup Fuel Under LOCA

- Thick pre-transient (in-reactor) oxide layers (<100 μm)
- Hydrogen uptake during normal operation (<700 wppm)
- Increased cladding mechanical constraint due to tight fuel-cladding bonding
- Fuel rim zone, possibility of fuel relocation to ballooned and burst region of cladding and release of fuel particles
ANL Integral LOCA-Criteria Tests

- Test Rod Sections with Intact Fuel ($\approx 300$-mm long)
  - Unrestrained
  - Internally pressurized
  - Temperature ramp ($300^\circ C$ to $1204^\circ C$), burst expected.
  - Hold in steam until desired level of oxidation is reached.
  - Controlled cool to $\approx 800^\circ C$
  - Water quench by bottom flooding
  - Post-quench ductility test
  - Behavior under post-LOCA seismic loading

- Issues Addressed
  - Quench resistance vs. 17%-ECR limit
  - Effects of pre-oxidation, hydrogen uptake, fuel bonding
  - Fuel relocation
  - Post-quench ductility
  - Validity of 17% criterion at high burnup
Oxidation Kinetics Studies - Objectives

- Provide data for LOCA-Criteria pre-test planning.
- Determine high-burnup effects on total oxidation and phase layer growth rates.
- Provide data and understanding necessary to develop oxidation model(s) applicable to LOCA at high burnup.
Test Fuel

• Zr-liner Zircaloy-2 Fuel:
  – BWR-L
  – burnup 57 MWd/kgU and lower
  – several grid spans
  – OD oxide 3-30 μm

• standard Zircaloy-4 Fuel:
  – PWR-H
  – burnup ≈70 MWd/kgU and lower
  – several grid spans
  – OD oxide up to ≈100 μm

• M5 and Zirlo cladding:
  – uncertain, not in current program
Oxidation Kinetics at High Burnup

- Zr-lined Zircaloy-2
- Test on archive material
- Test on 57 MWd/kgU BWR-L fuel
Hydride Structure, As-Spent Fuel
BWR-L Zr-lined Zircaloy-2
≈56 MWd/kgU
BWR-L Zr-lined Zircaloy-2, unirradiated. Weight gain rate (at 1204°C) consistent with Cathcart-Pawel correlation.
BWR-L, Zr-lined Zircaloy-2, Fuel Burnup 56 MWd/kgU (1204°C, 10 min.)

- No clear boundary between “pre-transient” oxide and high-temperature oxide, “alpha-incursion” as expected.

- Noncracked pre-transient oxide in this cladding appears to provide “barrier” effect to high-T oxidation (in contrast to “non-barrier” effect observed for cracked oxide in high-burnup standard Zircaloy-4)
BWR-L, Zr-lined Zircaloy-2
Fuel Burnup 56 MWd/kgU
(1197°C 20 min.)

- From several initial tests at ≈1200°C in flowing steam in hot-cell environment, uneven oxidation was observed in the irradiated cladding.
- It is not clear whether this is due to some unknown test factor(s) or reflects true material property.
- Tests in modified facility are under progress for clarification.
Post-Quench Ductility

- background
- additional considerations
- test fuel
- test plan
Background
Post-Quench Ductility

• Key rationale for LOCA embrittlement criteria--1204°C (2200°F)
PCT and 17% ECR limits:
  - avoid zero-ductility in cladding
  - ensure coolable core geometry

• Primarily based on Hobson's test 1972-73:
  - Zircaloy-4 tube oxidized at 1100-1315°C on two sides
  - short ring cut, compressed 3.8 mm slowly
  - crack-free adherent oxide, H uptake low, <150 wppm
  - reflects O-induced embrittlement only
Background (Continued)
Post-Quench Ductility

- H-induced embrittlement at H contents higher than about 600-700 wppm:
  - observed in 1980-1983, ANL & JAERI
  - local regions near burst opening, Zircaloy-4 tube
  - H alone (low O in beta layer) not much deleterious

- Significant effect of H uptake in E110 Zr-1Nb:
  - Boehmert 1992, Griger et al. 1999
  - at H contents higher than about 150-200 wppm

- Effects of 4 factors appear inseparable:
  - oxidation (before and during LOCA transient)
  - H uptake (larger than a threshold amount)
  - high burnup
  - Nb addition (E110, M5, Zirlo, Alloy A)
Additional Considerations
Post-Quench Ductility

- Effect of hydrogen uptake
- High-burnup effect
- Advanced cladding alloys
Effect of Hydrogen on Ring-Compression Ductility

Data indicate E110 could be brittle at H uptake > 200 wppm.
Large H uptake in E110 due to breakaway oxidation.
No breakaway oxide on Zry-4, H low <130 wppm.
Effect of Hydrogen on Ring-Compression Ductility

- Ductility of Zircaloy-4 and Zr-1Nb (E110) is strongly influenced by total hydrogen content.
- Post-quench ductility cannot be predicted based on the degree of oxidation only (e.g., ECR).
Plastic Deflection to Maximum Load as Function of Oxidation and H Uptake

- Post-quench ductility is primarily determined by O and H concentrations in the beta-phase layer.

- An embrittlement threshold expressed in terms of oxidation alone may not be adequate for certain situations, even for peak cladding $T < 1204^\circ C$.

Chung & Kassner 1980

for fresh Zircaloy-4
burst and oxidized in steam near 1 atm. pressure
Eutectoid Decomposition of Beta Zr-Alloys Predicted at High H Content

E. Zuzek et al.
Bulletin of Alloy Phase Diagrams
Vol. 11, No.4 (1990) 385-395

- Nb (a beta stabilizer) is likely to lower the threshold H concentration for eutectoid decomposition.
3 Routes for Large Hydrogen Uptake

#1 During normal operation to high burnup (≈62 MWd/kgU)
  - standard Zircaloy-4 up to ≈700-800 wppm
  - low-Sn Zircaloy-4, Zirlo
  - M5

#2 Through "unprotected" ID surface near burst opening

#3 Through "high-temperature breakaway" oxides on the OD surface
  - H uptake through normal high-temperature oxide (crack-free, tetragonal, protective) is limited to <150 wppm.
Route #2, Important Questions

#2 Through "unprotected" ID surface of localized "necks" exposed to stagnant H-rich steam-hydrogen mixture near burst opening:

- probable for all types of cladding
- a localized phenomenon

Q2.1 Will this phenomenon occur even at high system pressure during LOCA (e.g., 50-500 MPa steam pressure)?

Q2.2 Will this phenomenon occur in high-burnup cladding of which ID surface is covered with (Zr,U)O₂ layer?

Q2.3 What would be the effect of fuel relocation and slump into the ballooned and burst region which may occur at high burnup?

*** Current ANL integral LOCA test facility is designed to provide database to answer Q2.2 and Q2.3.***
Route #3, Important Questions

#3 H permeation through "breakaway" oxide that forms on the OD surface in steam at high temperatures:

- Zircaloy-4 is not susceptible to this phenomenon under LOCA-like conditions.
- (On the ID surface, Zircaloy-4 is susceptible to breakaway oxidation in H-rich stagnant steam-H mixture at 900-1120°C).
- Large H uptake through this route has been observed for E110 Zr-1Nb (Boehmert 1992, Vrtílova et al., 1996, Griger et al., 1999)

Q3.1 Do E110 and M5 exhibit similar behavior (Route #3)?

Q3.2 Is exposure to certain length of time at certain range of temperature (e.g., 900-1120°C) required for this phenomenon to occur?

Q3.3 What is the time-temperature envelope under which this phenomenon occurs? Is this envelope relevant to a LOCA?

Q3.4 What is the effect of pretransient (pre-LOCA) oxide and H uptake?
Test Plan
Post-Quench Ductility

• Method of ductility test being evaluated (by ANL, NRC, and EPRI), but not determined yet.

• Types of Test being considered:
  - diametral compression of tube that survived quenching during integral LOCA-criteria test (tube burst, oxidized, slow-cooled, and quenched at ≈800°C)
  - slow compression of rings cut from integral-test tube above, fuel at ring center removed.
  - compression of undeformed unirradiated rings after oxidation at <1204°C with and without H charge at low Ts.
  - axial tensile test of tube from integral test above
  - ring stretch test
  - tube impact test (dynamic fracture toughness)
Summary

- Integral LOCA-criteria test on high-burnup BWR and PWR rod sections with intact fuel
- Successful construction of and tests in mockup facility, base experience for in-cell facility
- Microstructural characterization and initial oxidation tests on 57 MWd/kgU BWR fuel
- Inseparable integral effects of oxidation, hydrogen uptake, high burnup, and Nb addition on post-quench ductility
Discussion:

Comment from the audience: One of key parameters seems to be the critical concentration of hydrogen within the β layer, or thickness of beta phase layer. Can you comment on that?

Comment by H. Chung: Yes, both are key factors. Also important is the oxygen concentration in the beta layer. To be more quantitative, for a given thickness of prior beta layer, the distributions of oxygen solutes and hydrides in the transformed beta layer are the key factors. For Zircalloys, O and H solutes redistribute themselves during beta-to-alpha-prime transformation, that is, O atoms diffuse into the growing alpha-prime grains and H atoms diffuse toward the remaining beta at the periphery of alpha-prime grains. Hydride precipitation is via nucleation and growth process and appears to be insignificant because of fast quench. However, beta-to-alpha transformation in Zr-1Nb may occur via a different mechanism, as suggested by Boehmert 1992. What I suggest on the basis of information from binary phase diagrams is occurrence of eutectoid decomposition of beta in Zr-1Nb into alpha and hydrides when hydrogen content in the beta exceeds the eutectoid composition. In such situation, hydrides will precipitate fast.

Comment from the audience: Phase transformation temperature (diagram) of M5 and Zircaloy-4 is, up to 1000 ppm of hydrogen, very similar.

Comment by H. Chung: Before I accept the statement, I would like to understand in detail how the phase diagrams were determined and see the data. The exact location of alpha-to-beta transformation temperature in Zr-based alloys is quite tricky. This is because when very small beta islands are present (at temperatures slightly higher than the transformation temperature) and the material is cooled, prior beta is invisible at room temperature unless the material is quenched very fast. Because of these characteristics, erroneously higher alpha-to-beta transformation temperatures have been reported in several investigations in 1970s and 1980s. When O content is low (e.g., %0.1 wt%) and H content is high (e.g., >700 wppm), the beta-stabilizing effect of %1-wt% Nb could be secondary because the beta-stabilizing effect of the high-concentration H is predominant. However, the first step would be to understand exactly how the test materials were prepared and how the transformation temperatures were measured. I believe that really applicable phase diagrams are "pseudo-ternary" phase diagrams of M5 and Zircaloy-4 that contain 0.3-0.7 wt% O and 150-800 wppm H.
7. Thermomechanical Properties of Oxydized Zirconium based Alloy Claddings in LOCA Conditions
Yu. K. Bibilashvili, N.B. Sokolov, L.N. Andreeva-Andrievskaya, at al. VNIINM, Russian Federation

Paper summary

The limits, which characterize the Emergency Core Cooling System (ECCS) acceptance criteria in the part of allowable degree of the cladding oxidation, are stipulated in the Russian normative document. The embrittlement criterion limits the local depth of the cladding oxidation and the maximal temperature:
PCT / maximal cladding temperature: \(< 1200\, ^\circ\text{C}\)
ECR / maximal local depth of the cladding oxidation: \(< 18\%\) of its initial thickness.
The experimental research to justify the Zr1%Nb fuel rod cladding of VVER-type embrittlement criterion have been carried out in Russia since the 70s. The technique for Zr1%Nb alloy was coordinated by the Chief Designer of the nuclear power plants with VVER-type reactor (OKB "Hydrometal") and the Regulatory Body of Russia (GAN). The experimental research comprises two stages:

First - the research of Zr1%Nb alloy oxidation in steam with the purpose of defining the dependences conservatively describing the oxidation kinetics of the Zr1%Nb alloy in a wide temperature range \((550 - 1700)^\circ\text{C}\). The oxidation lasts from several minutes to several days. Thus the influence of a range of factors on "Zr1%Nb - steam" reaction rate was investigated. They are: cladding deforming under the action of excess internal or external pressure; presence of hydrogen, air, nitrogen additives in steam; the excess steam pressure, irradiation.

Second - the experiments with the purpose of estimation of the oxidized Zr1%Nb claddings heat resistance, i.e. the ability of the cladding to withstand thermal-force loading under LOCA conditions and to keep the mechanical strength after LOCA sufficient for further manipulations with fuel assemblies (unloading, transportation).

The purpose of the heat resistance experiments was ascertainment of minimum time of claddings oxidation and corresponding ECR degree, resulting in the claddings failure: on quenching, during pulling from the experimental installation, during disassembly, during handling (for example preparation for metallographical research, etc.).
The experiments on heat resistance included:

1. Research of the ability of the oxidized fuel rod claddings to withstand the thermal shock (thermal shock tests);
2. Estimation of the physico-mechanical state of the oxidized claddings after the thermal shock (cold water bottom flooding or quickly (1-2sec) moving in a vessel, filled by distilled water of room temperature): estimation of impact elasticity (impact tests), estimation of residual ductility (compression tests), estimation of hydrogen content, the metallographical research of the claddings after the thermal shock (measurement of thickness and microhardness of the "alloy - steam" interaction layers).

The main result of the thermal shock experiments which were carried out is the experimental confirmation of the embrittlement criterion "1200\, ^\circ\text{C}\, \text{PCT} - 18\%\, \text{ECR}" for the Zr1%Nb fuel rod claddings of VVER type. The rupture (upon quenching, during dismantling, during handling) of the unirradiated Zr1%Nb fuel rod claddings oxidized at \((900 - 1200)^\circ\text{C}\) temperatures took place outside of the cladding's allowable state range. The rupture of the irradiated up to burnup of 46.2, 48.3, 49.5 49.8 MWd/kgU Zr1%Nb claddings (during disassembly, during handling, during transportation) oxidized up to \(1100\, ^\circ\text{C}\) temperatures took place outside of the cladding's allowable state range. No claddings were ruptured upon quenching.

The comparison of the compression tests results revealed an absence of distinction in residual ductility values of the irradiated and unirradiated Zr1%Nb claddings.
The experimental data base obtained by investigating the behaviour and properties of VVER type fuel rod claddings from Zr1%Nb alloy under LOCA simulating loading conditions is sufficient for the conclusion about the character and numerical value of criterial parameters of the embrittlement criterion.
conclusion about the character and numerical value of criterial parameters of the embrittlement criterion from the point of view of claddings ability to withstand thermal-force loading upon quenching and to keep the mechanical strength after LOCA sufficient for further manipulation with the FA (unloading, transportation).

The representative embrittlement criterion comprises all together the maximum temperature of the cladding heating and the local depth of the cladding oxidation. The numerical values "1200 °C - 18 % ECR" are justified by the experimental data obtained in oxidation kinetics and in special heat resistance experiments.
THERMOMECHANICAL PROPERTIES OF OXIDIZED ZIRCONIUM-BASED ALLOY CLADDINGS IN LOSS OF THE COOLANT ACCIDENT CONDITIONS

Yu.K. Bibilashvili¹, N.B. Sokolov¹, L.N. Andreeva-Andrievskaya¹, V.Yu. Tonkov¹, A.V. Salatov¹, A.M. Morosov², V.P. Smirnov³

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Presented by
L. Andreeva-Andrievskaya
Abstract

The research of Zr1%Nb fuel rod claddings of the VVER type reactors behavior and characteristics in loading conditions simulating accidents with loss of the coolant and active zone quenching experimental data are presented.

The experimental data allow to estimate type and numerical value of the embrittlement criteria parameters (ECR).

The thermal shock experiments under loading conditions simulating accidents with loss of the coolant (temperature, environment, deforming, limitation of the claddings axial deforming, quenching rate, irradiation) data are presented.

It is shown, that the mechanical characteristics of the oxidized claddings material after the thermal shock (impact elasticity, residucal ductility) are sufficient to withstand quenching and for further removing and transportation.

Introduction

Under LOCA conditions (Design Basis Accident - DBA conditions), there may be a short period of time before the fuel rods are recovered with cooling water. During this period, the decay heat causes the fuel rods to undergo a temperature excursion, and Zr1%Nb fuel rod claddings may reach temperatures of about 1200°C or possibly higher. Under such conditions they are intensively oxidized in steam.

The degree of the claddings oxidation is determined by the level of temperature, pressure, time of oxidation, deformation and other factors.

Owing to the cladding material embrittlement the initial thermophisical and mechanical properties of Zr1%Nb alloy, characteristics of ductility change. The thickness of the claddings unoxidized metal decreases. Therefore it is obvious, that the stage of failure, connected with quenching of the AZ (Active Zone) by cold water from the emergency core cooling system, is the most dangerous because of the thermal stresses in the fuel rod claddings.

The main constructive requirement to the fuel rods consists in absence of the fuel rod fragmentation under LOCA conditions. The fuel rod claddings and fuel rod assemblies (FA) should keep their geometry. The cladding should sustain dynamic loadings, arising with unloading the FA or fuel rod from the AZ, accommodation them in a storage and further transportation.

The limits, which characterize the Emergency Core Cooling System (ECCS) acceptance criteria in the part of allowable degree of the cladding oxidation, are stipulated in the Russian normative document [1].
The embrittlement criterion limits the local depth of the cladding oxidation and the maximal temperature. This criterion directly determines state of the fuel rod as a geometrical construction:

<table>
<thead>
<tr>
<th>PCT</th>
<th>Maximal cladding temperature:</th>
<th>Not above 1200°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>ECR</td>
<td>Maximal local depth of the cladding oxidation:</td>
<td>Not more than 18% of its initial thickness</td>
</tr>
</tbody>
</table>

The conservative dependence [2] is recommended to be used for estimation of the claddings degree of oxidation (oxygen weight gain).

**Experimental research of the Zr1%Nb claddings heat resistance**

The experimental research to justify the Zr1%Nb fuel rod cladding of VVER-type embrittlement criterion have been carried out in Russia since the 70th. The technique for Zr1%Nb alloy was coordinated by the Chief Designer of the nuclear power plants with VVER-type reactor (OKB "Hydrometall") and the Regulatory Body of Russia (GAN).

The experimental research comprises two stages.

First - the research of Zr1%Nb alloy oxidation in steam with the purpose of defining the dependence conservatively describing the oxidation kinetics of the Zr1%Nb alloy in a wide temperature range (550 - 1700)°C. The oxidation lasts from several minutes to several days. Thus the influence of a range of factors on "Zr1%Nb - steam" reaction rate was investigated. They are: cladding deforming under the action of excessed internal or external pressure; presence of hydrogen, air, nitrogen additives in steam; the excessed steam pressure, irradiation.

Second - the experiments with the purpose of estimation of the oxidized Zr1%Nb claddings heat resistance.

The ability of the cladding to withstand thermal-force loading under LOCA conditions and to keep the mechanical strength after LOCA sufficient for further manipulations with FA (unloading, transportation) is understood as heat resistance.

The purpose of the heat resistance experiments was ascertainment of minimum time of claddings oxidation and corresponding ECR degree, resulting in the claddings failure: on quenching, during pulling from the experimental installation, during disassembly, during handling (for example preparation for metallographical research, etc.).
High-temperature oxidation

The experimental research of the Zr1%Nb fuel rod claddings oxidation have been carried out in Russia since the 70th. The main results and dependences are represented in the report [2].

The requirements to the oxidation experimental procedure are the following:
- indirect method of the specimens heating in well temperature-controlled working zone;
- well controllable, uniform on height and radius of the specimens’ heating zone temperature field;
- high measurement accuracy of the specimens temperature, despite of oxide layer formation of significant thickness and possible deformation of the simulator;
- quality and structure of water steam (or other oxygen medium) should not be changed during the experiment;
- possibility of ensuring of isothermal regimes with the given pressure and steam flow rate;
- careful degreasing of the simulators surfaces that will be oxidized;
- use of experimentally defined simulators weight gain as oxidation reaction measure.

Estimation of the degree of oxidation

The degree of Zr%Nb claddings oxidation is estimated by the weight gain (Δm), or the local depth of oxidation (ECR) value.

The local depth of the cladding oxidation (ECR) is understood as the total equivalent Zr-layer thickness (that would react with steam assuming the whole locally absorbed oxygen goes on formation of stoichiometrical zirconium dioxide ZrO₂) related to the initial cladding thickness.

If the fuel rod depressurizes, oxidation of both outer and inner surfaces of the cladding are taken into account.

The weight gain can be defined simultaneously with oxidation (the experimental weight gain), or it can be calculated under the dependence, received on the basis of the experimental data (calculated weight gain). The experimental weight gain is solely correct method of definition of the oxidation degree.

The weight gain Δm and the local depth of oxidation ECR are connected by the relation [2]
ECR = \( N \times (\frac{\delta_e}{\delta_o}) \times 100, \% \)

where 
- \( N \) - the coefficient taking into account two-sided oxidation of the cladding, \( N = 2 \);
- \( \delta_o \) - initial thickness of the specimen, cm;
- \( \delta_e \) - thickness of the equivalent layer (calculated Zr thickness, which go on \( \text{ZrO}_2 \) formation), cm
  \( \delta_e = \mu_{\text{Zr}} / \mu_{\text{O}_2} \times 1/ \rho_{\text{Zr}} \times \Delta m; \)
- \( \mu_{\text{Zr}}, \mu_{\text{O}_2} \) - molecular weights of Zr and oxygen accordingly;
- \( \rho_{\text{Zr}} \) - Zr density, mg/cm\(^3\);
- \( \Delta m \) - specific weight gain, mg/cm\(^2\);
- \( \Delta m = \Delta M/S_o \)
- \( \Delta M \) - oxygen weight gain, mg
- \( S_o \) - cladding initial surface square, cm\(^2\)

It is to be pointed out that the specific weight gain of a cladding is found by dividing the weight gain by the square of the initial cladding surface. This gives a conservative estimation of the specific weight gain. Real oxidation area can be greater due to possible cladding deforming.

The relation for the ECR definition has a kind

\[
ECR = N \times 4,355 \times 10^{-2} \times \Delta m / \delta_o, \% 
\]

In the temperature range (900-1200)°C and the oxidation time up to 900 sec the parabolic rate equation is recommended as a conservative one for assessment of the degree of Zr1%Nb claddings oxidation [2]

\[
\Delta m = 920 \times \exp (-10410/T) \times \sqrt{\tau}, \quad (1)
\]

where
- \( \Delta m \) - specific weight gain, mg/cm\(^2\);
- \( T \) - temperature, K;
- \( \tau \) - time, s.

The experimental results have shown, that in the indicated temperature-time range the dependence (1) defines the Zr1%Nb claddings weight gain with the safety margin for the cases: availability of hydrogen addition in steam; exceeded steam pressure; the fuel rod claddings deformation and irradiation [2].

Fig. 1 presents experimentally obtained values of the oxidation reaction rate constant, dependence (1) for Zr1%Nb alloy, as well as Zry-4 oxidation rate and Baker - Just dependence, obtained for zirconium.
Fig. 1  Zr1%Nb and Zry-4 oxidation rate constant vs temperature

Heat resistance of oxidized Zr1%Nb claddings

The experiments on heat resistance included:
1. research of oxidized fuel rod claddings ability to withstand cold water quenching (thermal shock tests);
2. estimation of the physico-mechanical state of the oxidized claddings after thermal shock (cold water bottom flooding or quickly (1-2 sec) moving in a vessel, filled with distilled water of room temperature): estimation of impact elasticity (impact tests), estimation of residual ductility (compression tests), estimation of hydrogen content, the metallographical research of the claddings after the thermal shock (measurement of thickness and microhardness of the "alloy-steam" interaction layers).

Thermal shock experiments

Requirements for the experimental procedure used to test for thermal shock comprise the following:
- indirect method of the simulators heating in well temperature-controlled working zone;
- isothermal exposure, time and temperature are fixed;
- quick (1-2 sec) moving of the simulator to the vessel, filled by distilled water of room
temperature or cold water bottom flooding;
> analysis of the simulator state after thermal shock tests;
> formation of the simulators failure map.

The experiments were carried out with the short-length fuel rod simulators. Main loading factors present in LOCA were taken into account in the experiments. The use of a short-length simulator ensured a uniform in height and time controlled temperature field. The presence of $\text{UO}_2$ pellets (or sintering $\text{Al}_2\text{O}_3$) in the simulator gave practically real values of temperature stresses, arising in the cladding upon quenching. Both tight (internally pressurized) and having unsealed ends fuel rod simulators were tested.

The thermal shock experiments with unirradiated claddings are carried out on two installations: with heating of the simulator cladding by located in the central hole of $\text{UO}_2$ pellets heater (TEFSAI) and with continuous registration of the specimen weight gain during oxidation (UNOPRO) (fig2). The experiments with irradiated claddings are carried out on the universal installation (UVS). The main element of the installation is the heating modulus (fig.3).

The thermal shock tests are carried out in a wide temperature range (900 - 1300)°C. The claddings oxidation degree was various (up to 60% ECR).

The schemes of the simulators with unirradiated and irradiated undeformed claddings used for thermal shock tests are given in fig.4. The undeformed (fig.4 - type 1, 2, 4), and deformed and ruptured under the action of excessed internal pressure of inert gas (Ar) claddings (fig.4 - type 3) were tested.

The heat resistance data for the VVER type fuel rod claddings, irradiated up to burnup of 46,2, 48,3; 49,5 49,8 MWd/kgU were obtained.
Fig. 2 Scheme of the UNOPRO installation

Fig. 3 Universal UVS installation. The scheme of the heating modulus

1, 2 - thermocouple
3 - withdrawal mechanism
4 - suspension
5 - heating modulus
6 - specimen
7 - channel
8 - water tank
9 - specimen position after withdrawal
10 - channel for the thermocouple
Type 1

Type 4

1 - simulator head
2 - \( \text{UO}_2 \) pellets
3 - cladding
4 - adapter
5 - pellets holder

Type 2

1 - cladding
2 - pellet (\( \text{Al}_2\text{O}_3 \) - types 2, 3, 4, \( \text{UO}_2 \) - type 2)
3 - tungsten heater

Type 3

Fig. 4 Schemes of simulators
The parameters of the thermal shock tests were as follows:

<table>
<thead>
<tr>
<th>Parameter</th>
<th>UNOPRO</th>
<th>TEFSAI</th>
<th>UVS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Simulator cladding temperature, °C</td>
<td>(900 - 1200)</td>
<td>(900 - 1300)</td>
<td>(1000 - 1200)</td>
</tr>
<tr>
<td>Simulators heating rate, grad/s</td>
<td>var</td>
<td>10 - 20</td>
<td>1 - 3</td>
</tr>
<tr>
<td>Steam pressure, MPA</td>
<td>0.1</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>Specific steam flow rate, mg/cm²/s</td>
<td>7</td>
<td>50</td>
<td>2 g/min</td>
</tr>
<tr>
<td>Gas flow rate (argon), cm³/min</td>
<td>-</td>
<td>-</td>
<td>140 ± 6</td>
</tr>
<tr>
<td>Flooding water temperature (immersion), °C</td>
<td>20</td>
<td>20</td>
<td>25±35</td>
</tr>
<tr>
<td>flooding rate (lowering in), m/s</td>
<td>0.2</td>
<td>0.2</td>
<td>0.5 - 0.8</td>
</tr>
<tr>
<td>Cooling rate, grad/s</td>
<td>-100</td>
<td>-100</td>
<td>-100</td>
</tr>
</tbody>
</table>

The flooding rate values (or moving into the water) are chosen in view of the maximal conservatism of the received results (respect to cladding ability to withstand thermal shock). It was shown in works [3, 4], that fast cooling causes larger stresses in the cladding during the thermal shock (in case of large oxidation degree especially).
The simulators of types 1 - 4 consist of the following elements:

<table>
<thead>
<tr>
<th>Element</th>
<th>Installation</th>
<th>UNOPRO</th>
<th>TEFSAI</th>
<th>UVS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cladding Material</td>
<td>Zr1%Nb</td>
<td>Zr1%Nb</td>
<td>Zr1%Nb</td>
<td></td>
</tr>
<tr>
<td>Geometry Ø9,13x7,72x80mm</td>
<td>Ø9,13x7,72x120 mm</td>
<td>Ø9,13x7,72x60 mm</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pellet Material</td>
<td>Al₂O₃</td>
<td>UO₂</td>
<td>UO₂</td>
<td></td>
</tr>
<tr>
<td>Geometry</td>
<td>VVER type</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Plug Material</td>
<td>Zr1%Nb</td>
<td>-</td>
<td>Zr1%Nb</td>
<td></td>
</tr>
</tbody>
</table>

The ability of the Zr1%Nb cladding to withstand thermal shock (by bottom flooding with water or fast removing into the cold water) is shown in a map - the failure map. The thermal shock tests results are shown in the map in coordinates «time of oxidation» – «temperature of oxidation».

The cladding allowable state range is shown in the map. Borders of the allowable state range are:

<table>
<thead>
<tr>
<th>PCT</th>
<th>ECR</th>
</tr>
</thead>
<tbody>
<tr>
<td>&quot;Maximal allowable temperature&quot;</td>
<td>&quot;The maximal allowable degree of oxidation&quot;</td>
</tr>
<tr>
<td>vertical direct</td>
<td>inclined direct</td>
</tr>
</tbody>
</table>

The results of the thermal shock tests (post-test appearance and failure maps) are shown in fig. 5 - 8.

LOCA Topical Meeting, Cadarache, March 22-23, 2001
Fig.5 Post-test appearance and failure map. Simulator of type 1

Fig.6 Post-test appearance and failure map. Simulator of type 2
1st stage - 900°C
2nd stage - 1100°C

<table>
<thead>
<tr>
<th></th>
<th>Simulator</th>
<th>t = 40 min</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td></td>
<td></td>
</tr>
<tr>
<td>b</td>
<td></td>
<td></td>
</tr>
<tr>
<td>c</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Burnup 49.5 MWd/kgU

**Fig. 8** Post-test appearance and failure map.
Simulator of type 3

**Fig. 7** Post-test appearance and failure map.
Simulator of type 4
The simulator claddings state was various after thermal shock. The number and graphic symbol was given to each of the cladding state types.

<table>
<thead>
<tr>
<th>N</th>
<th>Cladding type of state</th>
<th>Graphic symbol</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>Survived quenching</td>
<td>Not filled cycle</td>
</tr>
<tr>
<td>1</td>
<td>Failed during handling (during disassembly with application of small loading)</td>
<td>Filled triangle</td>
</tr>
<tr>
<td>2</td>
<td>Failed during disassembly</td>
<td>Filled rhombic</td>
</tr>
<tr>
<td>3</td>
<td>Failed on quenching</td>
<td>Filled cycle</td>
</tr>
</tbody>
</table>

The generalized data of the thermal shock tests analysis - the critical parameters of the Zr1% Nb fuel rod claddings fracture are represented in fig. 9. The rupture (on quenching, during disassembly, during handling) of the unirradiated and irradiated Zr1% Nb fuel rod claddings of the 1-4 type simulators oxidized at (900 - 1200)°C took place outside of the cladding allowable state range.

![Fig.9 The fracture critical parameters. Simulators of type 1 - 4](image-url)
Estimation of the oxidized claddings physico-mechanical state after the thermal shock impact tests

In our view, the estimation of the oxidized Zr-based alloys cladding mechanical strength from the results of impact tests is the most objective for the embrittlement criterion substantiation. The specimens, made of unirradiated claddings of type 1 fuel rod simulators (fig.4), which have kept the integrity, were used for the tests. The scheme of the specimen for impact tests is represented in fig.10. In the simulator claddings of the 100 mm length was made a cut by a diamond disk. Its width was 0,5 mm and depth - 1,5 mm. The prepared specimens were tested on impact elasticity on pendulum impact installation PSV-1,5.

<table>
<thead>
<tr>
<th>temperature of tests</th>
<th>20°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>the maximal energy of impact</td>
<td>15 J</td>
</tr>
<tr>
<td>rate of impact</td>
<td>3-4 m/s</td>
</tr>
</tbody>
</table>

The account of the impact elasticity value was made using the relation \( a_v = \frac{W}{F} \), where \( W \) - work of rupture, J; \( F \) - the specimen cross section square in the place of the impact load application, cm².

The impact elasticity of Zr1%Nb claddings in initial, unirradiated state equals 64,2 - 89,3 J/cm². This value is two times higher than the value for the Zry-4F claddings.

Fig.10 Impact tests specimen scheme

The tough fracture mode shown by Zr1%Nb claddings oxidized at (900 - 1300)°C takes place upon oxidation to 5% ECR in impact tests. At a higher oxidation degree, claddings fracture in a mixed mode while at the oxidation degree more than 7% ECR, the fracture of claddings is brittle. As far as Zry-4F claddings, the critical oxidation degree at ductile to brittle fracture transition is not less than 7% ECR. The same procedure was used to test the specimens of both alloys.
The analysis of the impact tests results has shown, that values $a_x$ for Zr1%Nb and Zry-4F alloys practically do not differ (fig.11).

At the degrees of Zr1%Nb cladding oxidation up to 18 % ECR, the impact elasticity value remains not lower than 1 J/cm².

Fig.11 The impact elasticity of oxidized claddings vs oxidation degree

According to [5], the critical impact elasticity of Zry-4 claddings was $1.25$ J/cm². If this value is normalized to the transformed $\beta$-Zr cross section square, it will be $\sim 1$ J/cm². The impact elasticity values range, within which Zr1%Nb claddings do not fracture when cold quenched (subjected to thermal shock), is marked in fig.11.

Compression tests

The previously oxidized pieces of 30 mm length without fuel were used as specimens. The specimens were cut out from the thermal shock tested unirradiated and irradiated claddings which have kept the integrity. The fuel was removed from the irradiated claddings with the minimal mechanical load on the cladding (shaking out).

The mechanical tests with compression of the specimens in the direction, perpendicular to the axis of symmetry of the specimen, were carried out on the universal test machine ($T_{exp}=20^\circ$C, fig.12) and high-temperature vacuum installation. The rate of the grip displacement was 1 mm/min. Record of the diagram "force" - "grip displacement" was made during each test. The rupture deformation $\varepsilon_0$ was accepted as the ratio of the grip displacement, corresponding to the first load peak, to the initial diameter of the specimen. (in percentage). The rupture deformation $\varepsilon_0$ included the elastic and plastic components of the rupture deformation. The typical deformation diagram is shown in fig.12.
Fig. 12. The compression test scheme and typical deformation diagram.

The results of the compression tests with oxidized Zr1%Nb claddings after thermal shock are shown in fig. 13, 14. The results of the compression tests are conditionally divided into three groups:

<table>
<thead>
<tr>
<th>State of the oxidized Zr1%Nb claddings</th>
<th>Deformation value $\varepsilon_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td><em>Brittle</em></td>
<td>deformation value $\varepsilon_0$ does not exceed 0.1%</td>
</tr>
<tr>
<td><em>Low ductility</em></td>
<td>deformation value $\varepsilon_0$ does not exceed 4.0%</td>
</tr>
<tr>
<td><em>Presence of ductility</em></td>
<td>ductility $\varepsilon_0$ is present (more than 4.0%)</td>
</tr>
</tbody>
</table>

The results of the compression tests on the oxidized fuel rod claddings revealed a clear-cut distinction between oxidized claddings which had margin of ductility (complete ductility, low ductility), and claddings, which had a brittle character of rupture (fig. 13). The dependence of the "ductility boundary" on temperature of oxidation and a specimen's weight gain [5] can be seen. The ductility of the oxidized Zr1%Nb claddings essentially increase with the rise of the compression testing temperature.

However the critical values bounding the $\varepsilon_0$ margins for the "complete ductility" and "partial ductility" are assigned neither in Russia, nor abroad.

The results of compression tests on the oxidized unirradiated (type 2) and irradiated (type 4) fuel rod claddings are represented in fig. 14. The technique of the $\varepsilon_0$ estimation in cases of unirradiated and irradiated claddings was the same. The comparison
of the results revealed absence of distinction in residual ductility values of the irradiated and unirradiated Zr1%Nb claddings.

Fig. 13 Compression tests at $T=20^\circ C$

Fig. 14 Deformation $\varepsilon_0$ of the oxidized fuel rod claddings. Compression tests at $T=20^\circ C$
Metallographical research

The research of the claddings microstructure allowed to determine the thickness of the Zr1%Nb-steam interaction layers (ZrO₂, α-Zr(O), β-Zr), formed as a result of oxidation. The evaluation of the degree of the claddings oxidation based on the results of metallographical measurements of interaction layer thickness.

The results of the interaction layers thickness measurements are illustrated in fig.15.

The knowledge of the oxygen distribution within the interaction layers thickness gives an idea of the what influence produced by absorbed oxygen on the ductile (plastic) properties of the claddings. The indirect estimation of the oxygen content and distribution within interaction layers was made by the results of measuring the microhardness of the interaction layers.

The microhardness of the claddings was measured with PMT-3 microhardness tester at the load of 50g. The error of the measurements made ± 8 kg/mm².

Fig.15 Oxidation degree vs (metal + α-Zr(O)) layer thickness
The microhardness of the Zr1%Nb claddings metal part oxidized at <1050°C increases with increase of the oxidation degree. The microhardness of the oxidized at (1100-1200)°C claddings is weakly dependent upon the oxidation degree (fig.16).

Fig.16 Variation in (metal + α-Zr (O)) layer thickness averaged microhardness of Zr1%Nb claddings vs oxidation temperature

The microhardness distribution on the unirradiated Zr1%Nb cladding thickness is represented in fig.17.

Fig.17 Microhardness distribution on the cladding thickness

The Zr1%Nb α-phase microhardness increase rate is higher, than for Zry-4 alloy (particularly, at the early stages of oxidation). This difference may result from different ultimate solubilities of oxygen in β-phase of the two alloys at identical temperatures of their oxidation.
Definition of the absorbed hydrogen content

The analysis on the absorbed hydrogen content in oxidized Zr1%Nb claddings after thermal shock tests were carried out on the RH-2 device of the Leco firm by high-temperature vacuum extraction from melt in flow of gas-carrier.

Calibration of the RH-2 device was carried out on pure hydrogen, and also on the steel specimens with known hydrogen content. Relative mean square deviation (the error of the analysis) did not exceed 0.10.

The Zr1%Nb alloy possesses the greatest ability for absorption of hydrogen at 1000°C. This fact is explained by formation of the flaking oxide films on the cladding surface. Probably the dense oxide film is a good barrier for hydrogen penetration into the cladding.

The comparison of Zr1%Nb and Zry-4 data has shown, that for ECR up to 10% the hydrogen content in the two alloys differs weakly. For the large degrees of oxidation (15, 18% ECR) the hydrogen content in oxidized at 1000, 1100°C Zr1%Nb specimens is little higher, and after oxidation at the temperature 800°C is lower.

The dependence of the hydrogen content in zircaloy based claddings from the oxidation time is submitted in fig. 18.

![Graph](image)

Fig.18 Hydrogen content vs time of oxidation
Conclusions

1. The behaviour and properties of the VVER type Zr1%Nb fuel rod claddings in loading conditions, imitating the LOCA flooding stage were investigated.
2. The experimental data base obtained by investigating the behaviour and properties of VVER type fuel rod claddings from Zr1%Nb alloy under LOCA simulating loading conditions is sufficient for the conclusion about the character and numerical value of criterial parameters of the embrittlement criterion from the point of view of claddings ability to withstand thermal-force loading upon quenching and to keep the mechanical strength after LOCA sufficient for further manipulation with the FA (unloading, transportation).
3. The representative embrittlement criterion comprises all together the maximum temperature of the cladding heating and the local depth of the cladding oxidation.
4. The numerical values "1200°C - 18 % ECR" are justified by the experimental data obtained in oxidation kinetics and in special heat resistance experiments.

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10. Vrtilikova V., Valach M., Molin L. Oxidizing and hydriding properties of Zr1%Nb cladding material in comparison with zircalloys. Technical committee meeting on influence of water chemistry on fuel cladding behaviour. Rez, Czech Republic, 4-8 October 1993.
Comment by G. Hache:

Your metallographical results show that in 1200 °C there are much less margins for Zr1%Nb than for Zircaloy-4.

Answer by L. Andreeva-Andievskaya:

Yes, we have margins, but different for various kinds of simulators.

Question by R. Meyer:

Can you comment on Boehmert’s results?

Answer by L. Andreeva-Andievskaya:

No, I am not acquainted with his test conditions; ECR measure method and detailed information concerning test materials.

Question by G. Hache:

What is the reason of the distinction in results for the test with Al₂O₃ and UO₂ pellets?

Answer by L. Andreeva-Andievskaya:

The difference in the results explained by various heating method: external in case of Al₂O₃ pellets and internal (by located in the central hole of UO₂ pellets heater) in case of UO₂ pellets, leading to different oxidation of the claddings internal surface.

Question by H.M. Chung:

What were the hydrogen contents of the specimens of Fig. 14?

Answer by L. Andreeva-Andievskaya:

We did not measure the hydrogen content in these tests with irradiated claddings.
8. Progress in JAERI Program on High Burnup Fuel Behavior under a LOCA Transient
Hiroshi UETSUKA, F.Nagase, JAERI, Japan

Paper summary

For the better fuel cycle economy, the maximum fuel burnup has been extended to high levels in many countries. The maximum discharge burnup of fuel assembly is currently limited to 55GWd/t for BWRs and 48GWd/t for PWRs, respectively, in Japan and the research and development has been extensively conducted by industries to aim at a higher level of burnup. With the burnup extension, various changes in a fuel rod occur which might reduce the margin of fuel reliability and safety. Waterside corrosion and accompanied hydrogen absorption as well as irradiation damage become significant in the Zircaloy cladding tube, besides FP gas accumulation in fuel pellets and chemical bonding between pellet and cladding tube progresses at high burnup. These phenomena could influence on fuel behavior under a LOCA condition. In particular, degradation of cladding mechanical property due to corrosion and hydrogen pickup is considered the key factor influencing the integrity of fuel rod under a LOCA.

Current safety criteria for LOCA were established during the late 60's and early 70's in most countries. These criteria were mainly based on experiments conducted with fresh fuel cladding in the USA. In Japan, the criteria were once revised in 1981 based on newly accumulated knowledge on LOCA including a finding of so-called inner surface oxidation phenomena that could cause severe embrittlement of cladding. Although burnup effect was generally taken into account for the revised criteria, the level of fuel burnup was rather low at that time.

High burnup fuel behavior under a LOCA condition is a subject of great concern in many countries, consequently, extensive research programs have been performed in France, the United States and Japan etc. A systematic research program is being conducted at JAERI, which aims at a wide range data base for evaluating the influence of burnup extension on fuel behavior under LOCA conditions. The results obtained so far are summarized as follows:

- The parabolic rate law constants of Zircaloy/steam oxidation were determined for the wide temperature range from 773K to 1573K for the limited short time range up to 900 s.
- The influence of absorbed hydrogen on the Zircaloy oxidation rate varied depending on the temperature and hydrogen concentration. The influence was not significant within the conditions of postulated LOCA and hydrogen concentration in high burnup fuel cladding.
- The mechanical property of pre-hydrided or irradiated Zircaloy-4 cladding tube was remarkably degraded after short time annealing at temperatures above α to α+β phase transformation temperature of Zircaloy.
- The integral thermal shock test with pre-hydrided cladding tube has indicated almost no influence of absorbed hydrogen on the threshold value defined by oxidation condition, ECR, for cladding failure by quenching under no restraint condition. On the other hand the threshold ECR is obviously reduced by absorbed hydrogen for the restraint condition.
Progress in JAERI Program on High Burnup Fuel Behavior under a LOCA Transient

H. UETSUKA and F. NAGASE
Department of Reactor Safety Research
Japan Atomic Energy Research Institute

Abstract

With a view to obtaining basic data to evaluate high burnup fuel rod behavior under a LOCA condition, a systematic research program is being conducted at the Japan Atomic Energy Research Institute. The program consists of Zircaloy/steam oxidation test, mechanical property test of cladding, tube burst test and integral thermal shock test for evaluating the failure bearing capability of oxidized cladding upon quenching. Several types of Zircaloy cladding samples are used for these tests, they are as-received cladding tube, simulated high burnup fuel cladding and high burnup PWR fuel cladding tube. High temperature oxidation test with pre-hydrided sample has shown that the influence of absorbed hydrogen on oxidation rate is not significant for the time range expected in a LOCA and hydrogen concentration below about 800ppm. The integral thermal shock test with pre-hydrided cladding tube has indicated almost no influence of absorbed hydrogen on the threshold value defined by oxidation condition, ECR, for cladding failure by quenching under no restraint condition. On the other hand the threshold ECR is obviously reduced by absorbed hydrogen for the restraint condition.

* ECR : Equivalent Cladding Reacted (Proportion of oxide layer thickness assuming that all of absorbed oxygen forms stoichiometric ZrO₂)

1. Introduction

For the better fuel cycle economy, the maximum fuel burnup has been extended to high levels in many countries. The maximum discharge burnup of fuel assembly is currently limited to 55GWD/t for BWRs and 48GWD/t for PWRs, respectively, in Japan and the research and development has been extensively conducted by industries to aim at a higher level of burnup. With the burnup extension, various changes in a fuel rod occur which might reduce the margin of fuel reliability and safety. Waterside corrosion and accompanied hydrogen absorption as well as irradiation damage become significant in the Zircaloy cladding tube, besides FP gas accumulation in fuel pellets and chemical bonding between pellet and cladding tube progresses at high burnup. These phenomena could influence on fuel behavior under a LOCA condition. In particular, degradation of cladding mechanical property due to corrosion and hydrogen pickup is considered the key factor influencing the integrity of fuel rod under a LOCA.

Current safety criteria for LOCA were established during the late 60's and early 70's in most countries. These criteria were mainly based on experiments
conducted with fresh fuel cladding in the USA. In Japan, the criteria were once revised in 1981 based on newly accumulated knowledge on LOCA including a finding of so-called inner surface oxidation phenomena that could cause severe embrittlement of cladding. Although burnup effect was generally taken into account for the revised criteria, the level of fuel burnup was rather low at that time.

High burnup fuel behavior under a LOCA condition is a subject of great concern in many countries, consequently, extensive research programs have been performed in France, the United States, and Japan, etc. [1-5] A systematic research program is being conducted at JAERI, which aims at a wide range of data base for evaluating the influence of burnup extension on fuel behavior under LOCA conditions. The outline of the research program and recent results are described in the present paper.

2. Outline of Program

Several separate experiments are conducted to study fuel cladding behavior under a LOCA condition. They are oxidation rate measurement, mechanical property test, and rod burst test to investigate high burnup effect such as waterside corrosion, hydrogen absorption, and neutron irradiation on the fuel behavior. In order to evaluate the failure-bearing capability of oxidized cladding during a LOCA, an integral thermal shock test is performed with simulated fuel rods. The integral test simulates a temperature history of fuel cladding during LOCA, in which the test rod is experienced rapid heating-up, ballooning and burst, steam oxidation at high temperature and rapid cooling or quench by flooding water. Several types of samples, which are pre-oxidized, pre-hydrided, neutron irradiated in a research reactor and segments from spent fuel rods, are used in the present experiments to access separate effect. The schedule of present research program is shown in Table 1. The oxidation tests, mechanical tests and the integral thermal shock tests have been performed with non-irradiated cladding tubes so far. Preparations for the tests with irradiated cladding tubes are progressed in parallel. As a part of the program, a computer code is developed to analyze the fuel rod behavior under LOCA conditions including possible cladding failure on quenching. Data from the experiments will be incorporated into the computer code. The program will be once summarized in five years, then the tests will be continued targeting at the higher burnup range in the long term research program of JAERI, Advanced LWR Fuel Performance and Safety Research Program, ALPS.

3. High burnup effect on oxidation behavior of Zircaloy-4 in steam

Extensive studies were performed to precisely evaluate the oxidation rate of Zircaloy in high temperature steam for the LOCA safety analysis [6-11]. Obtained kinetics data show relatively good agreement each other, though the kinetics by Baker-Just is generally considered to overestimates the reaction for the temperature range above 1273K. In the previous studies the oxidation rates were determined mainly for the temperature range above 1273 K, while a limited number of studies were conducted for the lower temperatures range from 773 to 1273 K [11-13]. The effects of burnup extension on the oxidation rate, namely the effect of existence of oxide layer on cladding surface and hydrogen absorption during normal operation, have not yet sufficiently understood.
Isothermal oxidation tests in steam with low-tin (1.3wt% Sn) Zircaloy-4 has been conducted in the present study for the temperature range from 773 to 1573 K. The oxidation generally obeyed a parabolic rate law at temperatures above 1273 K (Fig.1) for the temperature and time conditions examined. On the other hand the reaction generally obeyed a cubic rate law in the temperature range from 773 to 1223. However, for the limited short time range up to 900 s, the oxidation at 773 through 1223K could be roughly estimated to obey a parabolic rate law. Then the parabolic rate constant was estimated for each test temperatures, which is plotted in Fig.2 with Arrhenius type equations determined for three temperature range.

The temperature dependence of the parabolic rate constant is described by:

\[
K_w = 3.66 \times 10^{-1} \exp(-170/RT) \quad \text{for } 1323 \text{ to } 1573K, \\
K_w = 9.15 \times 10 \exp(-230/RT) \quad \text{for } 1173 \text{ to } 1273K, \\
K_w = 1.72 \times 10^{-3} \exp(-126/RT) \quad \text{for } 773 \text{ to } 1123K,
\]

where \(K_w\) is in \(\text{g/cm}^2\text{s}\).

The discontinuity in the temperature dependence of parabolic rate law constant at about 1273K is possibly attributed to an allotropic phase transformation of ZrO\(_2\).
The possible effect of pre-existed oxide film on the oxidation rate during a LOCA is also one of concerns to be evaluated. As-received low-tin Zircaloy-4 cladding tube was oxidized at about 720 K in flowing oxygen to form 40 to 50 μm thick surface oxide film and subjected to subsequent isothermal oxidation test at temperatures ranging from 973 to 1473 K. The weight gain due to subsequent isothermal oxidation in steam at 1273, 1373 and 1473 K, is plotted as a function of oxidation time in Fig.3. The data for as-received cladding samples are also plotted for comparison. The weight gain of pre-oxidized sample is obviously smaller than that of as-received sample except for the test conditions of long time oxidation at 1373 and 1473K. Similar retarding effect of pre-oxidation on high temperature oxidation has been observed in the tests with spent fuel cladding[3,4].

The effect of absorbed hydrogen on oxidation rate was also investigated in the present study. Low-tin Zircaloy-4 cladding samples with hydrogen concentration of 200 to 1600wtppm were subjected to isothermal oxidation tests. The influence of preexisting hydrogen on the oxidation rate varied depending on temperature as well as hydrogen concentration. The largest influence was observed in the oxidation at 1223 and 1273 K. The proportion of sample weight gain, pre-hydrided to as-received, oxidized at 1273 K is plotted as a function of hydrogen concentration in Fig. 4. The figure shows that preexisting hydrogen generally enhances oxidation at this temperature and it is more remarkable at higher hydrogen concentrations. The present test result indicated about 9% increase of oxidation rate at maximum. However, the enhancement of oxidation rate due to preexisting hydrogen is considered to be not significant within the conditions of postulated LOCA and hydrogen concentration in high burnup fuel cladding.

Fig.3 Comparison of weight gain as a function of oxidation time between pre-oxidized and as-receives Zircaloy-4

Fig.4 Ratio of weight gain between pre-hydrided and as-received samples as a function of hydrogen content for the

4. Mechanical property change of Zircaloy-4 due to temperature transient
The amount of absorbed hydrogen increases with burnup extension and hydrides tend to accumulate at the cladding periphery [14], which might reduce the ductility of fuel cladding [15-17]. The temperature escalation in a LOCA transient causes microstructure change of cladding such as recovery of damaged structure, re-crystallization, phase transformation and redistribution of hydrides, affecting the mechanical properties of cladding tube. In order to investigate such effects on the mechanical property of Zircaloy cladding experienced high temperature transient, ring-like specimens cut from high burnup PWR fuel cladding with fast neutron fluence of $9 \times 10^{22} \text{n/m}^2$ and hydrogen concentration of about 200 wppm were examined in a hot cell. Specimens were heated to the temperatures between 673 and 1173K and annealed for 0 to 600s prior to the ring tensile test at room temperature. Figure 5 shows the correlation between crosshead displacement to failure and annealing temperature. Each numeral in the figure denotes the holding time at annealing temperature. The crosshead displacement to failure corresponds to the ductility of specimen, and it changed depending on annealing temperature and time. The specimen annealed at 873K showed the rapid recovery with time and the highest ductility data was measured at this temperature. It can be mainly attributed to recovery and re-crystallization of irradiated Zircaloy. On the other hand, for the temperatures above 1073K where the phase transformation from $\alpha$ to $\alpha+\beta$ occurred, the recovery of ductility was not so remarkable as the case at 873K, and once recovered ductility was reduced with increase of annealing time. The microscopic observation confirmed that these specimens experienced the transformation to the $\alpha+\beta$ phase and accumulated hydride beneath the outer oxide layer uniformly re-distributed in the wall thickness with comparatively random orientation above 973 K.

5. Integral thermal shock test of simulated fuel rod

The Japanese fuel safety criteria for LOCA were once revised in 1981 based on newly accumulated knowledge and findings. One of the important findings was so-called inner surface oxidation phenomena which was found in the rod-burst test in steam [18]. This experiment with as-received fuel cladding revealed that steam oxidation occurred inside the ruptured cladding could cause severe embrittlement due to a large quantity of hydrogen absorption in Zircaloy cladding. This clearly indicated not only oxidation but also additional hydrogen absorption could greatly influence on the embrittlement in case of cladding rupture. Consequently, current criteria are not based on the concept of zero ductility of cladding that was deduced from the experimental data of 60's, but they are mainly grounded on the failure threshold value defined by oxidation condition. ECR, which were
determined in the integral LOCA simulation tests of rod-burst, steam oxidation and rapid cooling by flooding water under the condition of rod-restraint[19].

In the present study the same type integral tests with pre-hydried Zircaloy cladding have been conducted to investigate the effect of hydrogen on failure-bearing capability of the oxidized cladding tube on quenching. Schematic drawing of the test apparatus is shown in Fig.6, which is composed of a tensile test machine, quartz-made reaction tube, infrared image furnace, steam generator and water supply system for flooding. The apparatus can simulate the sequence of fuel rod behavior during a LOCA. Details of the experimental procedure is described elsewhere [5].

The present integral thermal shock test aims to investigate both effect of geometry change of cladding tube, i.e. decrease in thickness, and preexisting hydrogen on failure-bearing capability of cladding upon quenching under conditions of restraint or no restraint. The samples examined so far are as-received 17-17 PWR type Zircaloy-4 tubes and pre-hydried tubes containing 400 to 600wtppm hydrogen.

Fig.7 shows the failure maps based on the test results under no restraint condition for both as-received and pre-hydried samples. The oxidation amount, ECR, of each data was calculated by the Baker-Just equation and both the reduction in cladding wall thickness due to ballooning and double-sided oxidation after cladding rupture were taken into account in the calculation. The open circles and triangles in the figure denote the cases of “survived”, while those of closed indicate “failed”. The threshold value of ECR between failure and non-failure seems to be around 60% for the as-received cladding tube. This is equivalent to that obtained in the previous study with 14-14 type cladding [19], indicating no significant influence of wall thinning with the change of fuel design. The threshold value for pre-hydried cladding tubes is nearly the same as that for as-received cladding. This indicates the hydrogen effect on the failure-bearing capability of cladding tube can be negligible under no restraint condition for the present sample condition of hydrogen content up to 600ppm.

The pre-hydried rods always fractured at the position apart from the rupture opening of cladding. Hydrogen analysis along the tube length showed remarkable difference in hydrogen concentration between rupture opening and other

![Fig.7 Failure maps relative to oxidation amount and oxidation temperature based on the test results under no restraint condition; (a) as-received and (b) pre-hydried cladding samples](image-url)
Fig. 8 Failure maps relative to oxidation amount and oxidation temperature based on the test results under restraint condition; (a) as-received and (b) pre-hydrided cladding samples
positions. Although no additional hydrogen absorption was analyzed at the rupture opening, plenty of hydrogen up to about 2000wtppm was measured at the fracture position, suggesting no significant effect of preexisting hydrogen on the failure during the test under no restraint condition.

During a LOCA transient a fuel rod shall be axially expanded with temperature increase and then be shrunk with decrease in temperature. The oxide formation and its growth on the cladding surface might enhance the fuel rod expansion. The shrinkage of fuel rod could be restricted to some extent in a fuel assembly due to possible mechanisms, and it is greatly dependent on the fuel design.

The actual condition of restraint imposed on a fuel rod during a LOCA is hardly evaluated and there can be no standard condition. Then, as the first approach to investigate the effect of restraint, integral thermal shock tests have been conducted under full restraint condition, though it is very conservative. Failure maps obtained from the tests under restraint condition for both as-received and pre-hydrided samples are shown in Fig. 8 with those obtained under no restraint condition for comparison. The influence of restraint was obviously seen in the both maps. The threshold ECR value is estimated to about 20% for the tests with as-received claddings. This result agrees very well with the previous data obtained in the experiment with 14-14 type cladding by authors. On the other hand, the influence of restraint on the fuel rod failure by quenching was much remarkable in the test with pre-hydrided cladding containing 400 to 600 wtppm hydrogen. The pre-hydrided rods always fractured at the position of rupture opening in this case. Although no additional hydrogen absorption was analyzed at the rupture opening, it can be considered that the amount of preexisting hydrogen at the ruptured position might be enough to influence the failure-bearing capability of fuel cladding under restraint condition.

6. Summary

A systematic research program on high burnup fuel behavior under LOCA conditions is being conducted at JAERI. The results obtained so far are summarized as follows;
- The parabolic rate law constants of Zircaloy/steam oxidation were determined for the wide temperature range from 773K to 1573K for the limited short time range up to 900 s.
• The influence of absorbed hydrogen on the Zircaloy oxidation rate varied depending on the temperature and hydrogen concentration. The influence was not significant within the conditions of postulated LOCA and hydrogen concentration in high burnup fuel cladding.

• The mechanical property of pre-hydrided or irradiated Zircaloy-4 cladding tube was remarkably degraded after short time annealing at temperatures above α to α+β phase transformation temperature of Zircaloy.

• The integral thermal shock test with pre-hydrided cladding tube has indicated almost no influence of absorbed hydrogen on the threshold value defined by oxidation condition, ECR, for cladding failure by quenching under no restraint condition. On the other hand the threshold ECR is obviously reduced by absorbed hydrogen for the restraint condition.

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2. N. Waeckel et al., Ibid.
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Tests with higher burnup fuel cladding in ALPS program

Table 1. Schedule of research program on high burnup fuel behavior under LOCA conditions
Discussion:

Question from the audience: Have you measured hydrogen content after annealing?

Answer by H. Uetsuka: No, we did not.

Question by H.M. Chung: During the 1973 ECCS hearing, retention of post-quench ductility (i.e., need to avoid zero ductility) was the primary rationale that eventually led to the establishment of the 17%-oxidation and 1204°C-peak-cladding-temperature limits. That rationale in turn was based on the recognition of the fact that it is difficult to quantify well the stresses and forces exerted on the cladding, such as thermal hydraulic and mechanical forces and the stress associated with constraints on the cladding. In contrast, as you stated, the basis of the oxidation criterion in Japan in early 1980s was the result of the JAERI thermal-shock quenching tests on fully constrained cladding. Now that your new results today indicate that threshold oxidation is %12-13%, a level significantly lower than the previous level of %17%, it seems that the rationale of the 1973 ECCS Hearing is justified. Could you kindly comment on this observation? Can you give an insight on what will be the primary rationale in dealing with cladding embrittlement in future in Japan?

Answer by H. Uetsuka: The JAERI's finding of inner surface oxidation and high content hydrogen absorption after rod rupture clearly indicated zero-ductility of cladding was possible at the oxidation condition far below the Japanese oxidation criteria 15%ECR. This is why the revised Japanese criteria does not depend on zero-ductility concept. It is clear that the condition of full constraint is too conservative. However, our tests results with fresh fuel cladding clearly showed 15% ECR is still OK even under full constraint condition. Although, as you pointed out, our new test results with prehydried cladding under fully restricted condition indicated lower threshold level of ECR than 15%, I do not think the rationale of the 1973 ECCS Hearing is justified. The essential point of future discussion in Japan may be on how we evaluate the stress associated with constraints on the cladding.

Question from the audience: What type of failure did you observe in restricted and unrestricted tests?

Answer by H. Uetsuka: We always observed a guillotine type failure in the restricted condition, while in unrestricted condition claddings were fractured into pieces in some tests oxidized at very high ECR conditions.

Question by G. Hache: Protective effect of preoxidation strongly depends on way how you did the preoxidation. Can you comment on that?

Answer by H. Uetsuka: I completely agree with it. The presented results show the protective effect of rather intact oxide film. The oxide formed on cladding surface during reactor operation may show less protective effect. We are planning to investigate it.
9. Evaluation of Fuel Rod Axial Forces During LOCA Quench
Nicolas Waeckel*, Rosa Yang**, Robert Montgomery**, Patrick Jacques*

Paper summary

Regulatory background

In 1972 US-NRC set a limit that ensures long term core coolability in case of a Loss of Coolant Accident (LOCA). Such limit is based on the zero ductility limit of the cladding measured after the LOCA transient, at low temperature (150 °C). As a result, the well known LOCA criteria have been defined : to avoid an excessive cladding embrittlement that could impair the core coolability, the peak cladding temperature (PCT) during the LOCA transient should remain below 1204 °C and the equivalent cladding reacted (ECR) should remain below 17 %.

These limits were set using unirradiated cladding at a time when high burnup and the related cladding hydriding were not of concern. NRC proposed the criteria to address “the unknown processes that might take place during a LOCA”.

By “unknown processes”, the staff meant the external forces that may appear during the quench phase (such as the axial forces resulting from a possible rod-to-grid lock-up) or after the LOCA transient (such as seismic loading or handling forces).

It is interesting to note that, from a practical point of view, the seismic events that may occur after a LOCA may not be a real safety concern for two reasons :
- first, the decay heat is low after a few days, therefore, even if the most embrittled rods (i.e. only a small fraction of the rods) collapse, the core will likely remain coolable (cf TMI).
- Second, it is now possible to assess, much better than 30 years ago, the seismic loading to the rods. Conservative calculations show that impacts between the rods and the grids would generate stresses in the clad well below the values corresponding to the zero ductility limit.

The handling forces should not be a concern : they affect one fuel assembly at a time, hence no core coolability issue.

The third “unknown process”, the fuel rod axial forces resulting from rods lock-up to the grids during the quench phase, is the focus of this paper. To address potential impact of these forces, JAERI performed a series of LOCA tests including fully constrained fuel rods1. The outcomes of these tests are two folds:

- an axial constraint reduces the failure limit upon quench from ECR in excess of 50% to ECR= 17% for as-received cladding. This result may suggest that applying an axial constraint during the quench phase is equivalent to using the post-quench ring compression tests which allowed NRC to determine the ECR=17% criterion

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209
axially constrained tests using pre-hydrided cladding (to simulate the pre-transient corrosion and hydriding) exhibit failure limit upon quench as low as 6%. If the previous statement about the equivalence between axially constrained LOCA tests and post-quench ring compression tests to define the LOCA limits is maintained, the LOCA criterion may be significantly reduced for high burnup fuel rods containing significant amount of pre-transient hydrogen. JAERI showed that a reduced experimental constraint allows a higher failure limit upon quench.

JAERI results lead to several questions: are the post-quench mechanical tests used by NRC in 1972 to determine the LOCA limits equivalent to the axial constraint during the quench phase, or one type of test is more conservative than the other? Are the fully constrained tests too conservative? If so, by how much?

These questions are the basis to better assess the degree of constraints the fuel rods may experience during a LOCA quench phase.

In this paper following two questions are addressed:
- Can the rods be locked-up at the grid locations during the LOCA?
- How strong the axial forces might be in case the rods are locked at the grid locations?

Rod-to-grid lock-up mechanisms

There are two possible causes for rod-to-grid linkage: clad ballooning in the grid cells or Zr-Fe eutectic formation at high temperature between Zr cladding and Inconel rod spring. The grid cells geometry is such that local clad strains of at least 15% are needed to lock the rods within the grids. REBEKA LOCA tests on rod bundles consistently show clad ballooning is lower than 5% at grid location in all tests compared to clad strains higher than 60% at mid-span. The result is due to an enhanced clad cooling at the grid location due to local flow turbulence and radiator fin effect. REBEKA tests results suggest the local clad temperatures were 200 °C lower at the grid location compared to the mid-span clad temperatures. Therefore, clad ballooning is not a possible scenario to lock the rods at grid locations.

During the high temperature phase of the LOCA transient, Zirconium Alloys may form an eutectic with the Fe contained in the inconel springs. Upon cooling, the eutectic may solidify and create a bonding between the clad and the spring. To address this issue, EDF performed separate effect tests to assess the temperature needed to form such an eutectic. Some as-received and pre-oxidized Zr samples were inserted in an Inconel crucible and heated in a furnace at 2 different heating rates until an incipient bonding is observed between the Zr sample and the inconel crucible. No incipient bonding was observed at clad temperatures and for holding times that might be expected during a prototypical LOCA transient at grid locations.

To conclude, experimental data suggest no rod-to-grid lock-up during a prototypical LOCA.

\[2\] K. Wiehr Rebeka tests "Bündelversuche untersuchungen zur Wechselwirkung zwischen aufblähenden Zircalloyhüllen und einsetzender Kernnotkühlung", EFK report May 1988
Axial forces assessment if locked-up is assumed

A finite element analysis has been performed to evaluate the fuel rod axial load within the assembly during quench if a rod-to-grid linkage is assumed. The Abaqus code has been used including 2-D beam elements representing the fuel rods, the guide tubes and the grids, high-temperature oxidation-induced growth, wall thinning due to oxide and oxide-rich alpha layer formation and either slip or fixed constraint at fuel rod-grid connection.

The analysis of JAERI single rod LOCA test with axial constraint (ref 1) with such a model shows that the model can reproduce the build-up of the axial force.

To analyze the axial load that may appear within a full-length 17X17 fuel assembly, 2-D model has been defined. The model consists of one row of 16 rods and 5 guide tubes connected to 6 grids and to the top and bottom nozzles. The temperatures of the fuel rods and the guide tubes are from LOCA system thermal-hydraulic analysis.

A total of four different cases have been conservatively investigated:

1. Base case: the guide tubes temperature follows the coolant saturation temperature in order to maximize the differential contraction between the fuel rod and the guide tubes. All the rods are locked to the spacer grids.
2. The guide tubes temperature is 100 °C above the coolant temperature. All the rods are locked to the spacer grids.
3. The guide tubes temperature is 200 °C below the fuel rod temperature. This temperature value, more realistic but still conservative, is based on Westinghouse calculations accounting for the radiation effect of the fuel rod bundle on the guide tubes. All the rods are locked to the spacer grids.
4. The guide tubes temperature conservatively follows the coolant saturation temperature (same as the base case) but an axial tolerance (1.1 mm) is assumed between the fuel rods and the spacer grid.

Based on the maximum clad strains expected at the grid location and the grid cells geometry analysis, this assumption is well justified.

The very conservative cases (1) and (2) give maximal axial forces lower than 300 N between grids 2 and 3. On the other hand, the more realistic but still conservative cases (3) and (4), does not lead to any axial forces.

Conclusions

Conservative analysis show low values of axial forces if rod lock-up is assumed. The calculated maximum axial load is 5 times lower than the fully constrained load at failure upon quench experienced in JAERI LOCA tests.

If more realistic guide tubes temperature history is used or if a small axial rod displacement (<1mm) is allowed, then no axial forces are calculated.

Moreover, experimental data suggest that the rod-to-grid linkage is very unlikely. Because of low clad temperatures at grid location, the clad ballooning strains at the grid location are too low to create any lock-up or any Zr-Fe eutectic formation.

Therefore, it has been demonstrated that fully constrained LOCA separate effect tests are atypical and overly conservative.

5 Calculations with unconstrained fuel rods shows that the maximum relative axial displacement during the quench period is only 1.5 mm at the grids 4 and 5 levels. Therefore, any axial move of the rods greater than 1.5 mm is sufficient to avoid any axial load on the rods.
Analysis of Fuel Rod Axial Forces During LOCA Quench

Nicolas Waeckel
Rosa Yang
Robert Montgomery
Patrick Jacques

LOCA Topical Meeting
OECD-SEG March 22-23 2001

OUTLINES

• Background
• Can the rods be locked-up at the grids locations?
  – Experimental data
• Analytical approach
  – Assume the rods are locked-up, how strong are the resulting axial forces?
• Conclusions
Current criteria are based on post quench mechanical tests

- In 1972 the NRC staff decided to set a limit that ensures **long term** coolability
- Such limit is based on the zero residual ductility limit of the cladding measured after the LOCA transient, at **low temperature**

$$PCT = 1204 \, ^\circ C \quad ECR<17 \%$$

- The criteria proposed were intended to address "**unknown** process that might take place during a LOCA"

![Diagram showing temperature and time with stages of LOCA transient and post-LOCA events](image)

**Unknown process that might take place during a LOCA**

- During LOCA events
  - axial forces induced by the rod-to-grid possible linkage

- Post LOCA events
  - seismic
  - hydraulic forces
  - handling

LOCA Topical Meeting Aix en Provence March 22-23 2001

Robert Fast Program
Influence of axial restraint level on failure limit (JAERI's results)

Need to better quantify axial loading that may apply during the quench phase of a prototypical LOCA transient

Two questions need to be answered

- Can the rods lock-up to the grids during LOCA?

- How strong the axial forces might be in case the rods are locked at the grid location?
Rod-to-grid lock-up mechanisms

- There are 2 possible causes for rod-to-grid linkage:
  
  1. Clad ballooning in the grid cells
  
  2. Zr-Fe eutectic formation between Zr cladding and inconel rod spring

How much ballooning is needed to lock rods in the grid cells?

- More than 28% strains are needed at the edge of the cells
- More than 21% strains are needed at the dimples location
- 15% strains are needed at the spring location
REBEKA LOCA tests on bundle

- REBEKA results consistently show clad ballooning < 5% at grid locations in all tests

No rod-to-grids lock-up due to clad ballooning

- Clad ballooning is limited at the grid location
  - Experimental data support clad deformations lower than 5% at the grids location
- Clad cooling is enhanced at the grid location
  - flow turbulence
  - radiator fin effect
  - Rebeka tests results suggest clad temperatures 200 °C lower at the grid location compared to the mid span clad temperatures

Clad ballooning scenario not possible to lock the rods at grid locations
Is eutectic formation a possible scenario to lock the rods?

- At high temperature Zirconium Alloys may form an eutectic with the Fe included in the Inconel rod springs
  - During cooling, the eutectic may solidify and create bonding
- Experiments have been performed to assess the temperature needed to generate an eutectic between Zr and Inconel

Calorimetric tests results

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Calorimetric experiments results

- No incipient bonding below 1200 °C

Melted Zr-Fe eutectic droplet unlikely to stay at grid location

- Lower clad temperature at the spring location
- Melted Zr-Fe eutectic droplet (if any) will slip down

No local bonding between the rod and the springs
Assume the rods are locked ...

- Experimental data suggest NO rod-to-grid linkage expected during a prototypical LOCA

- How strong are the axial forces if locked-up is assumed?
  - Perform FE analysis to evaluate the potential fuel rod axial load within the assembly during quench

Analytical Approach

- Use ABAQUS coupled with Zr-4 constitutive model to analyze assembly response during heat-up and quench
  - 2-D beam element representation of fuel rods, grids and guide tubes
  - Includes high temperature oxidation-induced axial growth
  - Wall thinning due to oxide and oxide-rich alpha layer formation
  - Either slip or fixed constraint at fuel rod-grid connection
  - Temperature of fuel rods and guide tubes from LOCA system thermal-hydraulic analysis
Model Verification

- Analysis of JAERI Thermal Shock Test
  - Single Rod Model
    - Analyze Heat-up and Quench
    - Verify numerical approach and constitutive model

Axial Displacement Results

Unconstrained Expansion and Contraction Results in JAERI Program
Evolution of Axial Strain - JAERI Test

![Axial Strain History Graph]

Axial strain (mm)

Time (sec)

Axial Constraint Applied

Axial Elongation by Oxidation (Leistikov Model)

αΔT + p_oxide

During Heatup and Oxidation

Axial Force Response - JAERI Test

![Axial Force Graph]

Fracture Force from JAERI Experiments

ECR > 15%

Start of Cooling/Quench

Time (s)
Analysis of JAERI Thermal Shock Test

- Model Verification
  - Analysis results demonstrate that methodology is reasonable
  - Can reproduce build-up of axial load as in JAERI tests

Full-Length Fuel Assembly Axial Load Analysis

- Model and Conditions
  - Mechanical properties used in analysis from MATPRO
  - Two-dimensional FE model of 17x17 Assembly
    (based on W V5H dimensions)

- Analysis Results
  - Axial Displacement
  - Axial Force
17x17 Fuel Assembly 2-D Model

Guide Tube Locations

Temperature Locations from CATHARE

Maximum and minimum temperature histories between grids 4 and 5

Quench Period for Region Between Grids 4 and 5
Maximum relative axial displacement is < 1.5 mm (unconstrained fuel rod)

Relative Axial Displacement History Between Grids 4 and 6 (Unconstrained Fuel Rod)

Max axial displacement < 1.5 mm

Quench period for region between grids 4 and 5

Axial Constraint Analysis Cases

- Rod-grid Lock-up assumed to occur at 800 °C (during the ballooning phase)

- Total of 4 different cases with all rods locked between two spacer grids
  1. Base case
     - Guide tube temperatures ~ Coolant saturation temperature (Tsat)
     - Solid lockup between fuel rod and grid spacer
  2. Guide tubes +100°C above saturation temperature
     - Solid lockup between fuel rod and grid spacer
  3. Guide tubes -200°C below fuel rod temperature
     - Based on Westinghouse calculations
     - Solid lockup between fuel rod and grid spacer
  4. Axial tolerance (±1 mm) between fuel rod and grid spacer
     - Guide tube temperatures ~ Coolant Saturation Temperature
Fuel rod - grid Maximum Allowed Displacement

Guide tubes strips
springs
wings

Maximum allowed axial displacement

±1 mm allowed displacement

Relative axial displacement

Non-Linear Fuel Rod-to-Grid Interaction
## Analysis Summary

<table>
<thead>
<tr>
<th>Analysis Case</th>
<th>Fuel Rod Axial Force</th>
<th>Axial Location</th>
</tr>
</thead>
<tbody>
<tr>
<td>Base case</td>
<td>290 N, 180 N</td>
<td>Grid Span 2-3, Grid Span 5-6</td>
</tr>
<tr>
<td>Guide tubes temp $T_{sat} + 100^\circ C$</td>
<td>280 N, 174 N</td>
<td>Grid Span 2-3, Grid Span 5-6</td>
</tr>
<tr>
<td>Guide tube temp $T_{fuel} - 200^\circ C$</td>
<td>~0</td>
<td></td>
</tr>
<tr>
<td>1 mm axial displacement at fuel rod-grid connection</td>
<td>~0</td>
<td></td>
</tr>
</tbody>
</table>

## Axial Force Distribution in Fuel Rods

Note: Plots show the maximum tensile force between grids.

- All rods stuck at 2 grids, Guide tube temperatures = $T_{sat}$
- All rods stuck at 2 grids, Guide tube temperatures = $T_{sat} + 100$ K
- All rods stuck at 2 grids, Guide tube temperatures = Fuel Rod Temp - 200 K
- 1 mm axial gap at rod-grid interface, Guide tube temperatures = $T_{sat}$
Fuel Rod Axial Force Distribution

Conclusions

- Fuel Assembly Axial Forces
  - Conservative analytical approach shows low values of axial force if rod lock-up is assumed
  - No axial force if a realistic guide tube temperature history is used
  - No axial forces if a small axial rod displacement (<1 mm) is allowed

- Experimental data suggest NO rod-to-grid linkage
  - clad ballooning strains at grid location too low to create lock-up
  - local clad temperature at grid location too low to allow for a Zr-Fe eutectic formation

LOCA separate effect tests with axial constraint during quench are NOT prototypical and too conservative
Discussion:

Question by R. Meyer:
Why do you not assume that the constraint comes the pellet stack internally when the tube bursts, shortens, and bends?"

Answer by N. Waeckel:
Well, the purpose of this presentation is to deal with the external forces that may apply during the LOCA transient and may have an impact on the overall behavior of the fuel rods. The constraint coming from the pellet stack is an internal force. The possible impact of these internal forces is (or will be) already included in the LOCA criteria based on integral tests such as the LOCA criteria tests at Argonne or the in-pile tests at Halden.(see also C. Vitanze's comment below)

Question by H.M. Chung:
It seems that you rather strongly discount the primary rationale of the 1973 ECCS Hearing (i.e., retention of post-quench ductility). Would you have a sound credible justification for this view of yours? In fact, the 1973 rationale was in part based on the recognition of the fact that it is difficult to accurately quantify the state and magnitude of the stresses exerted on fuel rod cladding in an assembly at the time of reflood quenching.

Answer by N. Waeckel:
I do not discount the primary rationale of the 1973 ECCS Hearings (i.e. retention of post-quench ductility to ensure long term core coolability). That was a wise and reasonable decision. I am just saying that the "unknown processes that may take place during a LOCA" (that justified the 1973 residual ductility based criteria) are now better known. Thirty years after the ECCS hearings it is now possible to determine more accurately the external forces that may impact the overall behavior of the fuel rods. We have shown that the axial constraint that occurs at the time of reflood are not significant. A postulated post-LOCA seismic event will not impair the core coolability either, for 2 main reasons: (1) the seismic loading on the fuel rods are very low, well below the yield stress of the cladding (i.e. rod behavior is not affected by a possible loss of the residual ductility of the cladding) and (2) after a short period of time the decay heat is such that there is no longer a safety issue, even if some part of the core collapses. At last the handling forces cannot be a safety issue since one fuel assembly at a time is concerned.

Comment by H.M. Chung:
I would also like to comment that the 0.3-J impact criterion we have proposed (in NUREG/CR-1344, 1980) was primarily based on the recognition of fact that cladding near ductile-brittle-boundary regime retains significant level of fracture toughness. 0.3-J impact energy corresponds to approximately 10 times of the impact energy that a burst and oxidized cladding must have to withstand the quenching thermal shock under unconstrained condition.

Answer by N. Waeckel:
0.3J impact energy represents also approximately 10 times the impact energy generated by a seismic event. Therefore the set of criteria defined in 1973 provide significant margins.
Comment by C. Vitanza: You mentioned the issue of internal forces and I would like to bring your attention that the Halden dryout test IFA 613 contains useful information. The test was run up to 1000-1100 °C clad temperature, followed by rapid quenching. The cladding was considerably embrittled but did not fail.
10. Synthesis of an EDF and FRAMATOME ANP Analysis on Fuel Relocation Impact in Large Break LOCA
Michel Lambert, Yann Le Hénaff, EDF/SEPTEN,
Jean-Luc Gandrille, Framatome ANP, France

Paper summary

During a Large Break (LB) LOCA, there is a rapid loss of coolant, inducing depressurization and loss of cooling of the fuel rods. Considering the hottest fuel pin, simulated with conservative assumptions, the following phenomena happen:
- increase of the stress (depressurization),
- cladding strain and burst,
- violent gap depressurization of the broken fuel pins,
- and possible relocation of fragments of the pellets in the burst area.

Locally (a few centimeters), geometry, and residual power are therefore potentially modified. Qualitative analysis shows that dominant parameters are:
- the fill up ratio of the balloon by the fragments,
- the balloon diameter in the affected area of the pin,
- the thermal conductivity of the fragmented fuel material,
- and the heat transfer between fragments and cladding.

Calculations are performed using the CATHARE LB version of the CATHARE code (this version is based on CATHARE 2 V1.3L_1 version with additional models specific to LB LOCA). The code has been approved by French Safety Authority for Large Break safety analysis.

The impact of relocation is estimated using conservative assumptions for the transient,
- penalizing size and location of the break (stagnation point in the core),
- SERMA + 2 $\sigma$ low for residual power,
- Penalizing maximum local power.

but realistic approach for relocation models (mainly balloon size, filling rate and heat transfer models). In assembly creep experiment, an azimuthal hot spot is observed, so the Keusenhoff's azimuthal temperature difference model, qualified on REBEKA experiment, is used to calculate the balloon size.

Calculation results show a limited impact on the Peak Clad Temperature (PCT) ($\sim$30°C). Therefore, it can be concluded that relocation phenomenon should not be taken into account in safety regulation, considering that:
- the effect of relocation on cladding temperature is low, even for the more penalizing transient, simulated with conservative assumptions (except for relocation phenomenon which is simulated with more realism),
- overall conservatism in the LB safety analysis is higher than relocation penalty,
- the relocation phenomenon, if it exists, is limited to the burst area (a few centimeters), and to the hot rods.
Synthesis of an EDF and FRAMATOME ANP analysis on fuel relocation impact in Large Break LOCA

Michel Lambert (EDF/SEPTEN)
Yann Le Hénaff (EDF/SEPTEN)
Jean-Luc Gandrille (FRAMATOME ANP)

Relocation in Large Break LOCA

Plan

- introduction
- description of the phenomenon
- conductivity model
- burst strain
- effect on cladding temperature
- conclusion
Relocation in Large Break LOCA

Introduction

- During LB LOCA, with the conservative conditions of the Safety Reports:
  - fluid depressurisation
    - stress
    - cladding strain → cladding burst
  - violent gap depressurisation
  - possible relocation of fragments of pellet in burst area

- Main question for the plant operator:
  - Is it necessary to consider the relocation phenomenon in safety studies?

Relocation in Large Break LOCA

Description of the phenomenon

- main parameters:
  - fill up ratio
  - balloon diameter
  - Conductivity of the fragmented area
  - Exchange between fragments and cladding
Relocation in Large Break LOCA
Conductivity of the fragmented area

- Many conductivity correlations describing an heterogenous area exist.
- The Imura-Yagi model (ICARE2 model) is the best one with regard to the temperature level and fragment size.
Relocation in Large Break LOCA
Burst strain

- Realistic approach:

  - In assembly creep experiment, existence of an azimuthal hot spot.
    (hot-side-straight effect)
    - FRAMATOME/CEA-Grenoble qualified an hot spot model
      (keusenhoff) on REBEKA experiment

  - In Keusenhoff model, the azimuthal hot spot temperature difference is governed by the eccentricity parameter.
Relocation in Large Break LOCA Calculation

* To estimate the effect of relocation, CATHARE GB (LB) was used.

* CATHARE LB is approved by French safety authorities.
  - Based on CATHARE 2 V1.3L_1 version developed by CEA-Grenoble.
  - Specific models developed by FRAMATOME for LB LOCA (refill phase, cross-flow, fuel models ...).

Relocation in Large Break LOCA Calculation

* Relocation is not taken into account in the present safety regulation.
  ⇒ a realistic approach can be use

* With realistic LB LOCA transient : no relocation
  ⇒ Conservative transient to estimate relocation impact

* Description of the reference case (3 loops PWR) :
  - conservative transient
    - penalizing size break
    - residual power SERMA+2ο
    - penalizing maximum local power (FQmax)
  - relocation model : realistic approach
Relocation in Large Break LOCA

Calculation

- EDF considers that relocation phenomenon should not be taken into account in safety regulation:
  - The effect of relocation on cladding temperature is weak even with conservative assumptions.
  - All the others conservatisms in the LB safety calculations are higher than relocation penalty.
  - Relocation phenomenon doesn't exist with realistic conditions.
  - The rupture node is not necessarily the hot spot node.
  - The relocation is limited to the burst area (a few centimeters) and to the hottest rods.
HIGH BURNUP UO₂ FUEL LOCA CALCULATIONS TO EVALUATE THE POSSIBLE IMPACT OF FUEL RELOCATION AFTER BURST

C. GRANDJEAN, G. HACHE, C. RONGIER
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CEN Cadarache, FRANCE.

Abstract
A literature review, conducted at IPSN, of available results of past LOCA in-pile experiments with irradiated fuel has revealed that irradiated rods behavior was significantly different from that of unirradiated rods under similar conditions.

Particularly, as suggested from the results from PBF-LOC, FR2 and FLASH5 experiments indicating a general occurrence of the relocation of fragmented fuel within the ballooned cladding, a main concern was raised regarding the possible impact of fuel relocation on peak clad temperature and local oxidation rate.

In view of obtaining some insight into the fuel rod performance following fuel relocation in a PWR high BU UO₂ rod under LOCA, calculations were being performed, using the French CATHARE-2 code with specific modifications in the fuel routines so as to describe a fuel accumulation after burst in the ruptured mesh of the rod cladding.

Main results indicate that the peak clad temperature may increase significantly, but still remains below the ECCS acceptance limit on PCT. On the other hand, the maximum cladding oxidation rate may exceed the 17% acceptance limit when the initial (in service) oxidation rate is cumulated with the transient oxidation rate. However, alternative embrittlement criteria based on residual thickness of ductile metal, such as the Chung and Kassner criteria, indicate a fair remaining margin to the thermal shock embrittlement limit, whereas the handling embrittlement limit may be exceeded.

1 INTRODUCTION
In the following of the studies that were jointly conducted by IPSN and EDF in order to investigate the behavior of high burnup fuel cladding under LOCA conditions, IPSN has been re-examining the problem of Loss-of-Coolant-Accidents with consideration of specific aspects related to fuel and cladding irradiation, so as to identify the remaining needs for further studies and experimental data.

These concerns have led IPSN to initiate new studies in order to provide the answers to pending questions regarding the behavior of irradiated rods and assemblies under LOCA conditions.

In a preliminary step, in view of obtaining some insight into the fuel rod performance following fuel relocation in a PWR high BU UO₂ rod under LOCA, calculations were being performed, using the French CATHARE-2 code with specific modifications in the fuel routines so as to describe a fuel accumulation after burst in the ruptured mesh of the rod cladding.

2 BACKGROUND
2.1 Irradiated fuel rod behavior
2.1.1 Literature review
There exists a few number of available results of such experiments with irradiated fuel rods under LOCA conditions: main issues were found in results from the PBF-LOC tests in the USA[1,2], the FR2 tests in Germany[3], and the FLASH5[4] test in France.
A process of fuel relocation was clearly evidenced from the experimental observations made in these tests series: in all irradiated rods of the PBF-LOC, FR2 and FLASH5 tests, fuel relocation has occurred as a result of slumping of pellets fragments from upper locations into the swollen region of the burst cladding. Fuel relocation phenomena is not restricted to high burnup fuel since fuel fragmentation occurs as soon as low burnup levels (it was thus noticed on LOC5-7B rod, fresh rod pre-conditioned up to 48 MWd/t).

A main question concerns the instant of fuel relocation occurrence in these experiments. It is not easy to make it perfectly clear for most of tests but, in FR2 tests E3 and E4 that were specially instrumented for that purpose, it was demonstrated that the fuel movement initiation occurred at the time of cladding burst, possibly initiated by the pressure difference between rod plenum and coolant channel with assistance of gravity slumping.

The fuel movement was probably favored in PBF-LOC and FR2 experiments where the fuel-cladding gap was not totally closed, due respectively to low burnup or to the inverted rod internal-external pressure difference during initial irradiation at low temperature. A tight bonding between fuel and clad was supposed to counteract the fuel motion inception. However, in FLASH5 experiment with high burnup fuel (50 GWD/t), and in spite of a low clad ballooning (not higher than 16%) post-test examinations have shown that fuel fragments were no more stuck to the cladding: the transient temperature rise combined to clad deformation may be sufficient to suppress fuel-cladding bonding.

2.1.2 Main concern

For irradiated fuel rods, as observed in the PBF-LOC results, the clad deformation is expected to be larger than for fresh rods, as a result of a more uniform temperature distribution associated to pellet-clad gap reduction following clad creepdown during rod irradiation. The increase in clad deformation will leave more space for fuel fragments to relocate. Since the fuel fragmentation is clearly associated to burnup, with finer fragments at higher BU, a pellet stack slumping is likely to occur after burst resulting in more or less compact filling of clad balloons. A major question is then what could be the impact on peak clad temperature and final oxidation ratio of the local increase in lineic and surfacic power and of the associated local decrease in fuel-clad gap?

It should be emphasized that this question is particularly important for UO₂ fuel at beginning-of-life and for MOX fuel at end-of-life where power generation is not reduced unlike for UO₂ fuel.

2.1.3 Early evaluations

The State-of-the-Art Review performed by P.D. PARSONS et al. for CSNI/PWG-2 and published in 1986[5], thus after PBF-LOC and FR2 tests completion, reports two calculation studies addressing the impact of fuel relocation on peak clad temperature.

2.1.3.1 Calculations in Sweden

The first one was conducted in 1978-79 in Sweden by Bergquist[6], within the frame of the ECCS evaluation for the Ringhals 3 power plant. It consisted of a series of parametric transient calculations, performed with the TOODEE-2 code, so as to evaluate the response on clad temperature with/without fuel relocation in the balloon after rod burst.

The main assumption for fuel relocation was a uniform redistribution of fuel in the deformed meshes of the balloon with a density taken to 50% of the theoretical density in the base case, a fuel thermal conductivity of 0.6 W/m/K and heat exchange between fuel and cladding dealt with an exchange coefficient of 5000 W/m²/K. In the reference case, the hoop strain of the most deformed mesh of the balloon was 42%, and the peak clad temperature (PCT) without fuel relocation did not exceed 2000°F (=1093°C).

Calculations with fuel relocation showed that the evolution of clad temperature in the ruptured mesh is essentially dependent of the power rating in that mesh, in relation with fuel average density:

240
- at 43 kW/m linear power (Fq = 2.09), the clad temperature evolution remains of classical shape, with a PCT around 2050°F (1121°C);

- at 47.87 kW/m linear power (Fq = 2.32) the clad temperature evolution exhibits a significant rate increase around 2000°F with a subsequent temperature escalation after 45 s in the transient;

- at 43 kW/m linear power, but with a 60% fuel theoretical density in the balloon (instead of 50%), the clad temperature evolution again exhibits a significant rate increase after 45 s, reach of the 2200°F limit around 62 s, followed by subsequent temperature escalation.

Although these early calculations had to be considered with large reservations, it may look surprising that they were not much discussed nor compared to counter-calculations, in consideration of the possible importance of calculated trends with respect to safety analysis.

2.1.3.2 INEL Calculations

The second evaluation was a steady state thermal analysis of a ballooned fuel rod following a fuel redistribution, the amount of which based on PBF-LOC tests results. This analysis was performed by T.R. Yackle[7] as a response to a NRC request; it is also mentioned by Broughton in the PBF-LOC3/LOC5 test report [1].

Fuel redistribution in the ballooned cladding is modeled by a series of up to 7 concentric rings of different width to take account of large particles of original fuel and small particles of additional fuel, neighbor rings being separated by gas gaps. Only radial heat transfer is considered, with a rod power corresponding to ANS decay heat 100 s after scram (~ 3% original power), and a flat radial power profile. A cladding surface heat transfer coefficient of 60 W/m²/K was assumed, a fuel thermal conductivity constant at 2.6 W/m/K and no radiative transfer between fuel particles.

The amount of fuel redistribution has been determined from the results of the PBF LOC-3 and LOC-5 tests. A line fit through the available data of fuel relative increase as function of cladding relative volume increase indicates an average filling ratio closed to 0.65. Three calculations have then been considered, corresponding to clad strain of 0, 44 and 89%. The following table gives the temperatures at fuel centerline and clad outside surface obtained in these three calculations.

<table>
<thead>
<tr>
<th>Clad strain (%)</th>
<th>T_clad (K)</th>
<th>T_cent002 (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>1095</td>
<td>1180</td>
</tr>
<tr>
<td>44</td>
<td>1120</td>
<td>1620</td>
</tr>
<tr>
<td>89</td>
<td>1320</td>
<td>2450</td>
</tr>
</tbody>
</table>

For the worst case, with 89% clad strain allowing the redistribution of 160% additional fuel, the outside clad temperature is 225 K larger than for the reference case (without deformation), while the corresponding increase on maximum fuel temperature is 1270 K.

The conclusion that was drawn at that time appears presently quite surprising, as it was stated that "fuel relocation into a balloon (with conditions such those calculated) will not pose a significant problem during a LOCA since both fuel and clad temperatures remain well below the corresponding melting points"...It may be thought that the relatively close occurrence of the TMI-2 accident is likely to explain such shift of concerns from LOCA to Severe Accident issues.

3 IPSN CALCULATIONS

3.1 Reference code and calculation procedure

The French CATHARE-2 code has been chosen as a base tool due to its capability to provide a best-estimate evaluation of the thermal-hydraulic evolution in hot assembly as well as in mean core subchannels. The code organization allows to run stand-alone calculations of the fuel module (the

241
CATHACOMB module) in order to provide rapidly information about the behaviour of specific fuel rods subjected to a given set of hydraulic conditions; these hydraulic conditions will thus not be influenced by the behaviour of such specific rods. The hydraulic conditions may be retrieved from a previous CATHARE whole calculation, or may be that of the current CATHARE computation with which the stand-alone fuel module is carried out in parallel.

For the purpose of this study, the calculations were performed as follows:
- in a first step a whole CATHARE-2 computation was run, for a large break LOCA transient occurring on a typical French PWR, with an input deck corresponding to a fresh fuel “mean core” rod and a high burnup UO₂ fuel “hot assembly” rod; this computation provided the hydraulic file used as input in the following calculations;
- in a second step a series of stand-alone CATHACOMB calculations were run, using the previously created hydraulic file, for a specific hot rod of the hot assembly, and with the inclusion of specific modifications in some fuel routines in order to simulate fuel relocation after burst in the ruptured mesh; these modifications will be briefly described in the following.

3.2 Code version

The CATHARE-2 V13L code version was used as starting version, according to the known improvements implemented in this version to calculate the reflooding phase of the LOCA transient.

Slight modeling improvements have been added in the fuel routines so as to compute at the end of each time step the oxidation weight gain (using both Cathcart-Pawel and Baker-Just rate laws) as well as the thickness of α-Zr[O] oxidation layer; the former variable allows a direct calculation of the equivalent oxidation rate ECR, while the latter variable allows to derive the remaining thickness of the central β-Zr layer.

3.3 Basic input options for the initial CATHARE-2 whole calculation

3.3.1 Accident transient conditions

The main assumptions are in agreement with those retained in the Standard Safety Report for 900 MWe French PWRs:

- double ended break on cold leg of the loop bearing the pressurizer,
- core at 102% nominal power at accident initiation,
- residual power = ANS71 + 20%.

3.3.2 Fuel rods description

Basically, three fuel rods may be described for a CATHARE calculation:

- the mean core rod, with a weight of \((N_{as} - 1) \times N_{rpa}\)
  - where \(N_{as}\) is the number of assemblies in the core and \(N_{rpa}\) the number of active rods per assembly
- the hot assembly mean rod, with a weight of \(N_{rpa}\)
- one (or several) hot rod(s) in the hot assembly

Each of these rods is described in terms of geometry, cladding oxide thickness profile, power profile. A typical axial meshing with 40 meshes was chosen.

In agreement with the options retained in sensitivity studies performed at EDF some years ago, the lineic power of the mean core rod was chosen as that of beginning of life (BOL) while only the hot assembly rods were chosen irradiated to 57 GWj/Th; the ratio \(F_{th}\) of hot rod power to mean core rod power was chosen to 1.28 (as compared to the 1.55 value for BOL case), and the ratio \(F_{hp}\) of hot rod power to hot assembly mean rod power was kept to 1.05 identical to BOL case.
Prior to the LOCA initiation, the reactor core is then supposed to be subjected to a transient evolution that brings the three above mentioned rods respectively to: 68.20 kW, 83.25 kW and 87.41 kW, and with a truncated cosine axial power profile.

The irradiated rods bear an external oxide layer on the zircaloy clad, with a thickness profile typical of 4 cycles irradiation, the maximum thickness reaching 106 μm at 2.79 m elevation. The pellet-cladding gap is supposed to be closed in the irradiated rods at transient initiation. The internal pressure in hot conditions in irradiated rods is significantly higher (~15.6 Mpa) than in mean core fresh rods.

Two hydraulic channels are associated to the mean core and hot assembly rods. The thermo-mechanical behaviour of the hot rod(s) is influenced by the hot assembly channel hydraulics during the blowdown and refill phases and by the mean core channel hydraulics during the reflooding phase.

3.4 Reference case behaviour (without fuel relocation)

A reference calculation without fuel relocation was first performed for the hot rod of the hot assembly. It must be pointed out that a best-estimate treatment of the clad ballooning and burst for the irradiated rod was not searched here: the standard clad deformation and burst models for fresh fuel were kept unchanged in CATHARE.

However, in consideration of the results of the TACIR experiments (oxidation and quenching tests) on irradiated cladding [8], having clearly indicated that the initial oxide scale was no more protective for high temperature oxidation, it was chosen to suppress the protective effect of the initial oxide scale towards transient oxidation of the clad, that is normally active in the standard oxidation model of CATHARE.

The rod cladding appeared to rupture at 30.2 seconds on mesh 24 (elevation 2.15 m) with a hoop strain of 56.3%. All the following results, unless explicitly stated, will refer to the ruptured mesh elevation.

Figure 1 displays the evolution of the fuel centerline and clad outside temperatures: the clad outside temperature rises to a maximum of 970°C while the fuel centerline temperature remains below 1100°C during the heatup phase.

Figure 2 displays the equivalent cladding reacted ECR evolution, as calculated with Cathcart-Pawel rate law, for the ruptured mesh and the two neighbor meshes. For the non ruptured meshes, due to unprotected oxidation on the external face only, the oxidation rate ECR is increased by about 1.7% in absolute value, while on the ruptured mesh, due to two-sided oxidation, the ECR rises from an initial value at 9.2% to 12.6% at the end of the transient.

Figure 3 compares the equivalent cladding reacted ECR evolutions, as calculated with Cathcart-Pawel and Baker-Just rate laws, for the ruptured mesh. It can be noticed that both correlations give very close results in the corresponding range of clad temperature. Acceptance criterion on clad maximum oxidation rate (<17%) is clearly well satisfied.

Figure 4 displays the evolution of the remaining thickness of the clad β-Zr layer, showing the sharp drop in thickness (from ~520 to ~330 μm) corresponding to clad ballooning up to rupture, followed by a slow decrease corresponding to high temperature oxidation. The final thickness remains just above 300 μm, indicating a fair remaining resistance to thermal shock embrittlement, with reference to embrittlement criterion proposed by Chung and Kassner[9], while the handling limit proposed by these authors is just reached.

3.5 Fuel relocation case

3.5.1 Basic assumptions and modeling options

With reference to the FR2 experimental results discussed before in section 2.1.1, we assumed that fuel pellets crumbling and relocation occurred immediately after the cladding burst, leading to a partial
filling of the inside volume of the ballooned cladding ruptured mesh. This volume was calculated as that of a cylindrical volume with clad inner radius at burst and mesh height. It was then assumed that this ruptured mesh volume was filled homogeneously with fuel fragments up to an user's input filling rate (= ratio of dense fuel volume to new mesh volume). A base calculation was performed with a filling ratio of 61.5% corresponding to a value measured in the FR2 experiment E5. Two other calculations were conducted with values of the filling ratio of 40% and 70% in order to evaluate the sensitivity of the results to this main parameter.

The fuel fragments were assimilated to spherical particles with user's input diameter. These fragments are in contact with the cladding, leading thus to a closed fuel-cladding gap.

The effective thermal conductivity of the fuel fragments was derived from the Imura[10] correlation and taking into account the radiative transfers between particles according to the Yagi theoretical model. The resulting model, so-called "Imura-Yagi" model, had been implemented in the SFD code ICARE2 of IPSN after it had been validated against Sandia DC1 experiment.

According to the results of the TAGCIR experiments mentioned in previous section, the protective effect of initial oxide scale towards LOCA transient oxidation was again suppressed in all the following calculations involving fuel relocation.

3.5.2 Results of the base case (61.5% filling ratio)

A basic calculation was performed with a filling ratio of 61.5% corresponding to a local value measured on a sample taken from the ballooned region (with 67.5% total circumferential elongation) of the FR2 experiment E5. The particle diameter was taken as the average size determined in FR2 experiments, i.e. 2.7 mm.

Figure 5 displays the evolution of the fuel centerline and clad outside temperatures: the clad outside temperature reaches a maximum level around 1100°C while the fuel centerline temperature remains below 1200°C during the heatup phase.

Figure 6 shows the evolution of the oxidation rate ECR, as calculated with Cathcart-Pawl rate law, for the ruptured mesh and the two neighbor meshes. It appears that the oxidation rate rises from 9.2% to near 18% on the rupture/relocation mesh, while the increase in ECR does not exceed 2% on the neighbor meshes. Figure 7 displays the evolution of ECR values at ruptured mesh, as calculated with Cathcart-Pawl and Baker-Just rate laws; compared to the corresponding curves for the calculation without fuel relocation (figure 3) a clear distinction can now be made between both evolutions, corresponding to the increase in clad temperature. The maximum value of ECR calculated with Baker-Just rate law is 19.4%, thus exceeding the current acceptance limit.

Figure 8 displays the evolution of the remaining thickness of the clad β-Zr layer, showing the same sharp drop as in reference case corresponding to clad ballooning up to rupture, followed by the decrease corresponding to high temperature oxidation, with a final thickness just below 250 μm. Since the maximum oxygen content at this temperature level remains below 0.9 wt %, it appears that the Chung/Kassner criterion would be satisfied for the thermal shock limit but not for the handling limit.

3.5.3 Sensitivity to the balloon filling ratio

Finally, comparative calculations were performed with filling ratio values of 40% and 70%, the latter value corresponding to the fuel void fraction measured by gamma decay counts in some PBFT-LOC experiments. The particle diameter was kept at the same value as in the previous calculation (2.7 mm).

Figure 9 displays the evolution of clad outside temperature with increasing value of the balloon filling ratio: for 40% filling the temperature level is similar to that of reference case without fuel relocation whereas peak clad temperature reaches 1144°C with 70% filling.

Figures 10 and 11 display the evolutions of the oxidation rate ECR, as calculated with Cathcart-Pawl, and of the remaining thickness of the clad β-Zr layer respectively, with increasing filling ratio
values. For highest filling value, the total oxidation rate ECR reaches 19.7% (22% with the Baker-Just rate law) while the β-Zr layer remaining thickness remains near 230 μm at the end of LOCA transient.

4 SUMMARY AND CONCLUSIONS

LOCA transient calculations have been performed with an adapted version of the French code CATHARE-2 in order to evaluate the possible impact of crumbling and relocation of irradiated fuel in the ballooned region of a cladding after burst.

Focus has been put on the sensitivity of peak clad temperature and final oxidation rate on the filling ratio of the ballooned cladding with fuel crumble.

The calculations do not intend to give a best-estimate view of the detail behaviour of high burnup fuel rod under LOCA transient. In particular, the thermo-mechanical properties of irradiated zircaloy were not available for the calculation of cladding deformation and burst with irradiated material.

The results indicate that for fuel relocation in the ballooned region with a filling ratio up to the values obtained in FR2 or PBF-LOC experiments, the peak clad temperature may increase significantly, but still remains below the ECCS acceptance limit (1200°C) on PCT.

On the other hand, the maximum cladding oxidation rate exceeds the 17% acceptance limit when the initial (in service) oxidation rate is cumulated with the transient oxidation rate and when the initial oxide layer is assumed no more protective for transient oxide growth. However, alternative embrittlement criteria based on residual thickness of ductile metal, such as the Chung and Kassner criteria, indicate a fair remaining margin to the thermal shock embrittlement limit, whereas the handling embrittlement limit appears exceeded.

The results of the present study give some insight into the possible impact of the crumbling and relocation of high burnup UO₂ fuel in a LOCA transient, a phenomena that was observed previously in in-pile experiments and which might significantly affect the late evolution of accident transient and associated safety issues. It must be pointed out that results of corresponding calculations with low burnup UO₂ or high burnup MOX fuels would have been more severe with regard to acceptance limits.

The results of the present calculation study give some support to the need for further experimental data, to be provided by irradiated fuel LOCA experiments involving fuel relocation. A best representativeness should be obtained with in-pile experiments, so as to maintain heat generation in fuel fragments whatever their displacement may be during the relocation process. Such experiments are currently under planning by Halden Reactor Project and by IPSN.
REFERENCES


3. E.H. KARB et al., LWR Fuel Rod Behavior in the FR2 in-pile Tests Simulating the Heat-up Phase of a LOCA. KFK 3346, March 1983


Large Break LOCA. Hot Rod. BU = 57 GWh/BU
No fuel relocation. No protective effect of initial oxide

Figure 1.  Figure 2.

Large Break LOCA. Hot Rod. BU = 57 GWh/BU
No fuel relocation. No protective effect of initial oxide

Figure 3.  Figure 4.
Figure 5.

Figure 6.

Figure 7.

Figure 8.
LB LOCA: Hot Rod. Burnup = 57 GWD/TH
Fuel relocation in ruptured mesh: Influence of filling ratio

Figure 9.

LB LOCA: Hot Rod. Burnup = 57 GWD/TH
Fuel relocation in ruptured mesh: Influence of filling ratio

Figure 10.

LB LOCA: Hot Rod. Burnup = 57 GWD/TH
Fuel relocation in ruptured mesh: Influence of filling ratio

Figure 11.
High Burnup UO$_2$ Fuel LOCA Calculations to Evaluate the Possible Impact of Fuel Relocation After Burst

C. GRANDJEAN, G. HACHE, C. RONGIER
IPSN, Cadarache, France

OECD Topical Meeting on LOCA Fuel Safety Criteria,
Aix-en-Provence, March 22-23, 2001
High Burnup Fuel LOCA Calculations to Evaluate the Possible Impact of Fuel relocation after Burst

BACKGROUND (1)

EXPERIMENTAL OBSERVATIONS

Main findings were provided by the results of: PBF-LOC, FR2, FLASH5 experiments:

- fuel relocation was observed in all irradiated rods as a slumping of fuel fragments from upper locations into the swollen region
- fuel movement initiation occurred at burst in E3 and E4 FR2 tests
- fuel motion, (favored in FR2 due to non closure of gap) is supposed to be counteracted by a tight fuel-clad bonding
  - bonding was not observed on FLASH5 (50 GWd/t) despite low clad ballooning (16%)

**Important issue:**
Fuel relocation ⇒ increases local power and reduces drastically pellet-clad gap

**impact on Peak Clad Temperature and Oxidation Rate?**

importance: UO$_2$ at BOL, MOX at EOL
Neutron radiographs of rod F1 (burnup 20 000 MWd/t_{U}).
Comparison between status pre-transient and post-transient.
High Burnup Fuel LOCA Calculations to Evaluate
the Possible Impact of Fuel relocation after Burst

BACKGROUND (2)

EARLY ANALYTICAL EVALUATIONS

*Bergquist* (Sweden, 1978-79) : Parametric transient calculations with TOODEE-2 code
- impact of fuel relocation after clad ballooning and burst / reference case without relocation
- main sensitivity to peaking factor ($F_q$) and density of relocated fuel ($\rho_{\text{reloc}}$)

**Results:**
- ref case ($F_q = 2.32$) w/o relocation $\rightarrow$ PCT $\sim 2000^\circ F = 1093^\circ C$
- relocation, $F_q = 2.09$, $\rho_{\text{reloc}} = 50\% \rho_{\text{theor}}$ $\rightarrow$ PCT $\sim 2050^\circ F = 1121^\circ C$
- relocation, $F_q = 2.09$, $\rho_{\text{reloc}} = 60\% \rho_{\text{theor}}$ $\rightarrow$ $T_{\text{clad}} \uparrow$ above 2150$^\circ F$ and subsequent escalation

*Backle* (INEL, 1980) : Steady state thermal analysis of a fuel rubble in clad balloon
- fuel relocation ratio extrapolated from PBF-LOC experiments
- relocated fuel modeled as a series of 7 concentric nodes with stagnant steam gaps
- power : ANS decay heat at 100 s ; flat radial power profile ;

**Results:** worst case : 89% cladding strain $\rightarrow$ 160% fuel redistribution

$\Downarrow$ $T_{\text{clad}} = 1320 \text{ K (} + 225 \text{ K)}$ and $T_{\text{cent-fuel}} = 2450 \text{ K (} + 1270 \text{ K)}$

**Conclusion:** fuel relocation = not a problem since both $T$ are well below melting points !!
IPSN Calculations: Large Break LOCA calculations with irradiated fuel rods

Code version
CATHARE2.V1.3L with specific modifications to:
- simulate fuel accumulation in the ruptured mesh after burst
- calculate oxidation rate ECR and $\beta$-Zr remaining thickness

Calculation Procedure
- 1\textsuperscript{st} step: whole CATHARE2 LOCA computation run without fuel relocation
  - provides the hydraulic conditions for following calculations
- 2\textsuperscript{nd} step: stand-alone fuel module (CATHACOMB) calculations
  - under imposed hydraulic conditions retrieved from previous step
  - without fuel relocation (reference case)
  - with simulation of fuel relocation after burst, according to user's input characteristics for the filling of the clad balloon
Whole CATHARE2 standard calculation:

- large break LOCA (double ended break on cold leg)
- mean core rod: fresh fuel
- hot assembly rods: irradiated fuel 57 GWd/t
  \[ \frac{P_{\text{hot rod}}}{P_{\text{mean core rod}}} = 1.28 \quad (1.55 \text{ at BOL}) \];  
  \[ F_q = 1.94 \quad (2.35 \text{ at BOL}) \].
- at accident initiation:
  - core at 102% of nominal power,
  - cosine axial power profile,
  - pellet-clad gap closed in irradiated rods,
  - rod internal pressure \( P_{\text{int}} = 15.6 \text{ Mpa} \)
High Burnup Fuel LOCA Calculations to Evaluate 
the Possible Impact of Fuel relocation after Burst

CHARACTERISTICS OF CALCULATIONS (2)

Stand-alone CATHACOMB calculations:

- suppression of the protective effect of initial oxide scale
  (according to the results of TAGCIR experiments on irradiated cladding)

- homogeneous filling of the balloon
  Filling ratio (= 1 - void ratio):
  - base case: 61.5% (value measured in FR2 experiment E5)
  - sensitivity study: 40% and 70%

- fuel fragments assimilated to spherical particles in contact with the cladding wall (res. gap = 1μm)
  particle diameter: 2.7 mm (= average value in FR2 experiments)

- thermal conductivity derived from a debris bed model, including convective and radiative heat transfer between fuel particles:
  IMURA/YAGI model, validated against the DC1 experiment (SNL)
Large Break LOCA. Hot Rod. BU = 57 GWj/tU

No fuel relocation. No protective effect of initial oxide

Rod burst at 30.2 s on mesh 24 (z = 2.15 m) with $\varepsilon_B = 56.3\%$
Large Break LOCA. Hot Rod. BU = 57 GWj/tU
No fuel relocation. No protective effect of initial oxide

Large Break LOCA. Hot Rod. BU = 57 GWj/tU
No fuel relocation. No protective effect of initial oxide
LB LOCA. Hot Rod. Burnup = 57 GWD/tU
Fuel relocation in rupt. mesh: 61.5% of balloon volume

CATHARE2 V1.3L priv
CLADEXTT (CELSIUS, Z215)

TCL_24

A fill=40%
B fill=61.5%
C fill=70%

LB LOCA. Hot Rod. Burnup = 57 GWd/tU

Fuel relocation in ruptured mesh. Influence of filling ratio

CATHARE2
V1.3L priv
LB LOCA. Hot Rod. Burnup = 57 GWD/tU
Fuel relocation in ruptured mesh. Influence of filling ratio

CATHARE2
V1.3L priv

Beta Layer Thickness (microns)

TIME (SECONDS)
High Burnup Fuel LOCA Calculations to Evaluate the Possible Impact of Fuel relocation after Burst

SUMMARY

- LOCA transient calculations have been performed with an adapted version of the French code CATHARE2 in order to evaluate the possible impact of crumbling and relocation of irradiated UO₂ fuel in the ballooned region of a cladding after burst.

- Focus has been put on the sensitivity of PCT and final oxidation rate on the filling ratio of the ballooned cladding with fuel crumble.

- Results indicate that with a filling ratio up to values obtained in FR2 or PBF-LOC experiments:
  - the PCT increases significantly but still remains below the 1200°C limit;
  - the oxidation rate ECR may exceed the 17% acceptance limit when initial and transient oxidation are cumulated and when initial oxide is considered no more protective for transient oxidation;
  - alternative embrittlement criteria based on residual thickness of ductile metal, such as the Chung and Kassner criteria, indicate a fair remaining margin to the thermal shock embrittlement limit, whereas the handling embrittlement limit appears exceeded;
CONCLUSIONS

* The results of the present study give some insight into the possible impact of the crumbling and relocation of high burnup UO\textsubscript{2} fuel in a LOCA transient, a phenomena that was observed previously in in-pile experiments and which might significantly affect the late evolution of accident transient and associated safety issues.

* Results of corresponding calculations with low burnup UO\textsubscript{2} or high burnup MOX fuels would have been more severe with regard to acceptance limits.

* These results bring some support to the need for further experimental data, provided by irradiated fuel LOCA experiments involving fuel relocation. A best representativity should be obtained with in-pile experiments so as to maintain the heat generation in fuel fragments whatever their displacement may be during the relocation process.
Discussion:

Comment by M. El-Shanawany: The UK carried out similar analysis (1988) using the computer code BART which included a number of models such as the grid rewetting effect that were not taken into account in your paper.

The UK analysis used information from a number of experiments such as KfK, PBF, UKAEA, Halden and URN. The analysis indicated that the peak clad temperature may increase but the temperature did not exceed the 1204 C limit.

Hence, it is not clear what is new in IPSN's analysis, and how your analysis is adding value to our understanding of fuel pellet fragment axial relocation.

(REF. : Calculations of the effect of pellet fragment axial relocation on the peak clad temperature during a loss of coolant accident in a pressurised water reactor. K. T. Routledge, M. El-Shanawany & D. Utton, Second UK National Heat Transfer Conference, 14-16 September 1988, University of Strathclyde, Glasgow )

Question from the audience: Was a rupture induced improving of cooling taken into account?

Answer by C. Grandjean: The improving of cooling associated to clad ballooning and rupture is not taken into account in the standard version of CATHARE, nor in the version modified for the calculations presented here. Such influence may however have been taken into account in some calculations performed by vendors with their own evaluation models.

Question by R. Meyer: What assumptions made a 10-times difference in peak cladding temperature between your calculations and the previous paper's?

Answer by C. Grandjean: IPSN calculations differ from EdF calculations on some options in modelling, particularly heat transfer in post-DNB conditions, resulting in different burst time and burst strain, and on options for fuel relocation in ballooned area, mainly filling ratio and residual gap (or not) between fuel fragments and clad.

G. Hache added: There was too much azimuthal temperature variation in the EdF model.

Question by H.M. Chung: Tight pellet-cladding bonding, commonly observed in high-burnup cladding, is likely to strongly influence the degree of azimuthal temperature variation at burst, and hence, burst size, wall thinning, susceptibility to thermal-shock fragmentation, and fuel relocation. Would
the degree of azimuthal temperature variation at high burnup be larger or smaller than at low burnup, and why?*

Answer by C. Grandjean:

The azimuthal temperature gradient is reduced for irradiated fuel as a result of fuel fragmentation and relocation associated to cladding creepdown during reactor normal operation. Fuel relocation is supposed to be a bit more compact at high BU with possibly fine fragments interspersed among larger ones. However, fuel rearrangement has been observed to start with early fuel conditioning and the resulting effect on azimuthal temperature variation might not be much larger at high burnup than at low burnup.

Comments by J. R. Jones:

The assessments of heat transfer in the blockage region of the ballooned fuel assembly is sensitive to the dynamics of entrained droplets and in the UK we have found it necessary to depart from the mean-diameter approach in favour of a multi-group representation of the droplet spectrum.

In the early 1980s work was reported by Garlick et al. on the observation of relocation of fractured fuel pellets in simulated LOCA conditions. The degree of pellet relocation around the time of burst was modest, and certainly less that the reported simulations assumed.

I question whether the relocation observed post test in PIE had occurred later than the time of interest.

Let me pointed out that pellets can be conditioned by ramping to high reactor power, prior to test and relocation can be examined without the need to achieve high fuel burnups. This was confirmed by Dr. Wolfgang WIESENACK of Halden who did this for the IFA 54x test series.

Answer by C. Grandjean:

In the FR2 experiments, only two tests (E3 and E4) have been instrumented in such a way to allow to trace the inception of fuel relocation. These two tests have clearly demonstrated that the fuel stack collapse starts at the time of cladding burst.

Comments by C. Vitanza

Halden LOCA tests with internal and external thermocouples showed that it was difficult to have a completely uniform azimuthal temperature distribution. Even if one tries to get very uniform boundary conditions at the circumference, there are always small azimuthal temperature differences which tend to reduce the effect of balloning.
12. High Burnup Phenomena Affecting the Failure Mode of Fuel Rods During LOCA.
Hiroshi Hayashi, NUPEC, Japan

Paper summary

NUPEC has made irradiation tests on BWR and PWR assemblies of burnup of 50 and 55 GWD/t, respectively. Through power ramp tests of segmented rods irradiated 3, 4 and 5 cycles in BWR we have found that the ramp terminal power of the failed segment irradiated 3 cycles is about 610 w/cm and the power of the failed segment rods irradiated 4 or 5 cycles decrease to about 350 or 420 W/cm, respectively. The failure mode of the 3 cycles irradiated segment is a pin hole type due to PCI-SCC but both of 4 and 5 cycles irradiated segments are axial sprits starting from the outer surface of claddings. The fracture surface shows brittle features developed by the combination of the stress due to PCI and the hydrogen in the cladding. This significant decrease of the failure thresholds would come from the irradiation embitterment and the hydrogen effects.

BWR 8x8 fuel assemblies with segmented rods were irradiated up to 5 cycles in Fukushima Daini Nuclear Power Station No. 2 Unit. Ramp tests of the segment rods were made in JMTR and PIEs were executed at NFD.

Maximum linear heat rate of the assembly keeps above 300 w/cm in the first cycle, above 250 w/cm in the second and the third cycle and decreases to 200 w/cm in the fourth cycle and 80 w/cm in the fifth cycle.

The main results of PIE concerning high burnup fuel performance are as follows:

Thin and uniform oxide covers cladding tubes and the thickness is about 10 to 20 micrometers during from the first cycle to the fifth cycle. The hydride in claddings distribute uniformly in the first, the second and the third cycle and gradually increases from the inside to the outside of claddings in the fourth and the fifth cycle. Hydrogen content in claddings is 25 ppm during from the first to the third cycle and 50 ppm in the fourth cycle and 100 ppm in the fifth cycle, but less than that of PWR claddings.

Bonding layer between claddings and pellets is observed in the samples irradiated for more than three cycles. The thickness of the bonding layer is about 10 to 20 micrometers and does not change with burnup. The rim structure at the pellet peripheral is observed when the pellet average burnup is over 35 GWD/t and the thickness of the rim structure increases linearly with burnup. Thermal conductivity calculated using the data of thermal diffusibility of pellets at 60 GWD/t decreases to about 80 % of un-irradiated one. Fission gas release rate is less than 5 % up to 50 GWD/t of rod average burnup except for 2 fuel rods, which have the higher linear heat rate of 355w/cm.

PWR assemblies with segmented rods have been irradiated at ENDESA's Vandellós 2 Nuclear Power Plant in Spain. PIE and power ramp tests of segmented rods were executed at Studsvik in Sweden.

The main results of PIE concerning high burnup fuel performance are as follows:

Oxide thickness of low-Sn Zircaloy-4 claddings irradiated for 4 cycles is about 100 micrometers and that of MDA and ZIRLO claddings is about 50 micrometers. Hydrogen content in the cladding metal depends on the oxide thickness and the maximum value is above 800 ppm in the case of low-Sn Zircaloy-4 irradiated to 58 GWD/t. Yield strength of irradiated claddings is greater than that of un-irradiated claddings and is almost constant up to fluence of 1.15x10^23 n/m². Total elongation of claddings decreases with fluence but is still 4%. Total elongation decreases with the hydrogen content in claddings. Concerning the rim structure of pellets, rim formation starts from over 30 GWD/t local burnup and the increasing rate of the rim width of large grain pellets is lower than that of normal pellets.

High burnup phenomena affecting LOCA failure mode would be:

* Cladding degradation due to hydrogen absorption and irradiation embitterment
* Bonding layer between pellet and cladding
* RIM formation in the region of pellet peripheral
* Decreasing of thermal conductivity of pellet.
High Burnup Phenomena Affecting the Failure Mode of Fuel Rods during LOCA

Hiroshi Hayashi   (NUPEC *, Japan)

* NUclear Power Engineering Corporation
Irradiation History of Step LUAs

Irradiation Period (Days)

EOC Peak Pellet Burnup (GWd/t)

1st cycle  | 2nd cycle | 3rd cycle | 4th cycle | 5th cycle

16  | 31  | 44  | 57  | 61  

Linear Heat Rate (W/cm)
Visual Appearance and Cross-sections of Oxides on Fuel Rods (Step II Fuel)

Local Burnup: ca 60 GWd/t

<table>
<thead>
<tr>
<th>Fuel Stack</th>
<th>Gas Plenum</th>
</tr>
</thead>
<tbody>
<tr>
<td>![Fuel Stack Image]</td>
<td>![Gas Plenum Image]</td>
</tr>
</tbody>
</table>

- crud oxide
- oxide

- Zry2

- 20 m
Burnup Dependence of Maximum Oxide Thickness

- $\alpha$: Step II (uniform)
- $\beta$: Step I (nodular)
- $\gamma$: 8x8 (nodular)

Local Burnup (GWd/t)
Hydrogen Content as a Function of Irradiation Period

\[ \omega : \text{Step II} \]
\[ i : \text{Step I} \]
\[ \epsilon : 8 \times 8 \]
\[ \alpha : \text{published} \]

\[ \uparrow : \text{Max.} \]
\[ \downarrow : \text{Min.} \]

Hydrogen Content (ppm)

Irradiation Period (days)
Cross-sectional Metallography at Pellet-Cladding Interface

Pellet → Cladding

1 cy, 16 GWd/t
2 cy, 31 GWd/t
3 cy, 43 GWd/t
4 cy, 56 GWd/t
5 cy, 60 GWd/t

50 m
SEM Images of High Burnup Fuel Pellet Periphery

Local Burnup; ca 60GWd/t

Polished Surface  Ruptured Surface
Burnup Dependence of Rim Width

Width of Porous Region at Pellet Periphery (m)

\( \square : \) Step II
\( \triangle : \) Step I
\( \times : \) 8x8

Pellet Burnup (GWd/t)
Burnup Dependence of Thermal Conductivity

Thermal Conductivity
(Norm. to un-irrad. UO$_2$)

+10%
-10%

α : UO$_2$
χ : (U,Gd)O$_2$

Burnup (GWd/t)
Burnup and Power Dependence of Fission Gas Release

Rod Average Burnup (GWd/t)  Maximum Linear Heat Rate (W/cm)
Typical Irradiation History of Segment Rod

Fuel Assembly Loading History in Vandillos 2
Axial Distribution of Oxide Thickness on 6th Segment
Measured by ECT
Max. Oxide Thickness Measured by ECT
Metallograph after 4-cycle Irradiation
Mechanical Properties of Cladding Tube
Mechanical Properties of Cladding Tube

Fast Neutron Fluence $\sim 10^{25} \text{ n/m}^2, E>1\text{ MeV}$
Elongation vs Hydrogen Content

- Low-lin Zircaloy-4: Dogbone
- Low-lin Zr-4+Texture control: -
- MDA+Texture control: -
- ZIRLO: -
- ZIRLO+Texture control: -

Gage Length: Dogbone=33mm, Tube=50mm
Ube

Hydrogen Solubility limit at Test Temp.
Relationship Between Bumup and Width of Rim Region
Discussion:

Question by R. Yang: Did you observe any difference in the ductility dependence of Zr-2 and Zr-4 on H and fluence? For the Zr-2 materials presented, the H level did not change much from three to five cycle (<150ppm), yet the ductility seem to change significantly. Is it due to fluence? In other words, the ductility of Zr-4 depends more on H and Zr-2 depends more on fluence?

Answer by H. Hayashi: Hydrogen contents in the cladding of BWR goes up from 25 ppm at 3 cycle irradiated to 100 ppm at 5 cycle irradiated. These values are smaller than hydrogen contents in PWR claddings. But the cladding temperature of BWR is lower than that of PWR and we found the strong effect of hydrogen contents on the ductility of irradiated Zircaloy-2. In this sentence, „ductility“ means not only the mechanical properties but also the ramp terminal power of the failed fuel rod at the power ramp.

Question by H.M. Chung: Almost all the hydrides in ZIRLO you have shown belong to the outside of cladding In low-Sn Zircaloy-4 hydrides are observed in the midwall area also. How do you explain it?

Answer by H. Hayashi: One difference between them is the amount of hydrogen in the cladding. Oxide thickness of ZIRLO and Low-Sn Zircaloy-4 are about 50 micronmeter and 100 micronmeter, respectively. So, the hydrogen absorbed in low-Sn Zircaloy-4 might be double of that of ZIRLO. Another difference is the fabrication process of the claddings. This ZIRLO cladding was fabricated by the process controled method to modify the textures.

Question by G. Hache: Mr.Kaminura's results on hydrogen content differ from yours Why?

Answer by H. Hayashi: After his presentation at SEG FSM meeting on June 2000 we have made the activity to distinguish the hydrogen contents in the cladding metals and the oxide by modify the measurement procedure. In my presentation the hydrogen content means amounts of hydrogen only in the cladding metals.

Question by C. Vitanza: The thermal conductivity decrease with burnup is not so pronounced. At what temperature was it measured?

Answer by H. Hayashi: The thermal diffusivity were measured in the temperature range from about 220 to 1600 degree centigrade. The thermal conductivity is calculated using the data of 1000 degree centigrade.
13. Results of the Experimental Research on High Burnup VVER-type Fuel Behaviour in LOCA Conditions

Smirnov V.P. et al., RIAR, Russian Federation

Paper summary

Currently, most fuel producers aim at providing the average fuel burn-up of about 60 MWd/kgU. Some producers predict to manufacture fuel with average burn-ups of 70-80 MWd/kgU in future. It requires to validate fuel safety and reliability. In the 70-80 MWd/kgU for LWR was validated up to the average burn-up of about 40 MWd/kgU. However, the burn-up increase resulted in the significant changes in the fuel and cladding properties: appearance of rim-layer; accumulation of the significant amount of fission products in the fuel, decrease in the cladding ductility due to radiation failures as well as oxidation and hydrogenation.

It requires to reconsider the behaviour of the fuel rods of this type under accident conditions and first of all under design-basis accident conditions. According to this task, in the 90s a group of companies under the leadership of ITRM RRC "KI" and SSC VNIINM carried out experimental research of the VVER spent fuel behaviour in RIA conditions. One of the experiment's most significant results is that high safety of the VVER high burn-up fuel as compared with the PWR and BWR under RIA conditions is shown. Currently RIA experiments on BIGR reactor are being carried out.

Less attention was paid to the experiments on Russian spent fuel behaviour under LOCA conditions. Some results on spent fuel properties and the evaluation of embrittlement safety criteria research were published. Recently, some relevant work in this respect was initiated by three institutions supported by "TVEL", "MSZ" and "NZHK" joint companies. The main experimental research results on high burn-up fuel rod behaviour under LOCA conditions are the following:

Relative fission product release increases from 15 % to 100 % with a temperature rise from 1000 to 2500°C. Fuel with burn-up of about 60 MWd/kgU is characterized by monotonously increasing FGR in the temperature range from 1000 to 1950°C. Fuel with burn-up of about 16 MWd/kgU is characterized by almost zero FGR up to 1600°C. FGR kinetics at temperatures above 1950°C for all burn-ups don't differ.

The oxidation rate of irradiated VVER-type claddings slightly exceeds the unirradiated claddings oxidation rate during first few minutes of oxidation at temperature 1000°C. At higher temperature level this difference reduces and at the temperature 1200°C the insignificant reverse effect is detected.

Short-term mechanical properties of irradiated Zr-1%Nb claddings are characterized by high ductility (δ ≥ 5 %, δ ≥ 20 %) even at room temperature. At typical LOCA temperatures (T>600°C) the irradiated cladding mechanical properties don't differ from the properties of unirradiated claddings. Experimental substantiation of the embrittlement criterion confirmed that Zr1%Nb claddings don't fail (during quenching, during disassembly, during transportation out of hot cell, during handling) up to 1100°C. Further experiments should be continued in temperature range up to 1200°C.

Different types of tests were carried out with the purpose of defining the rupture parameters (hoop strain, temperature, pressure, time) in typical for first and second LOCA stages loading conditions.
The Results of the Experimental Research of the VVER High Burn-up Fuel Behaviour Under LOCA Conditions

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Abstract

The report considers the principal experimental results on VVER and RBMK spent fuel behaviour under accident conditions. Main experimental research directions were following:

- properties of the high burnup fuel;
- validation of the embrittlement safety criteria;
- properties of the materials and FA elements under accident conditions;
- Zr1%Nb unirradiated and irradiated VVER-type fuel rod claddings behavior in accidents with loss of the coolant (LOCA) loading simulating conditions.

1. Introduction

Currently most fuel producers aim at providing the average fuel burn-up of about 60 MWd/kgU [1]. Some producers predict to manufacture fuel with average burn-ups of 70-80 MWd/kgU in future. It requires to validate fuel safety and reliability. In the 70-80\textsuperscript{a} fuel safety for LWR was validated up to the average burn-up of about 40 MWd/kgU. However, the burn-up increase resulted in the significant changes in the fuel and cladding properties: appearance of rim-layer; accumulation of the significant amount of fission products in the fuel, decrease in the cladding ductility due to radiation failures as well as oxidation and hydrogenation.

It requires to overthink afresh the behaviour of the fuel rods of this type under accident conditions and first of all under design-basis accident conditions. According to this
task in the 90th a group of companies under the leadership of IRTM RRC "KI" and SSC VNIINM carried out experimental research of the VVER spent fuel behaviour in RIA conditions. One of the most significant results of it is that high safety of the VVER high burn-up fuel as compared with the PWR and BWR under RIA conditions is shown. Currently RIA experiments on BIGR reactor are being carried out.

Less attention was paid to the experiments on Russian spent fuel behaviour under LOCA conditions. Some results on spent fuel properties and evaluation of embrittlement safety criteria research were published. Lately some relevant work in this respect has been started by three institutions supported by "TVEL", "MSZ" and "NZHK" joint companies. The results obtained during the last years are presented.

2. The results of experiments

The first stage of the experimental research of the high burn-up fuel rods behaviour in LOCA conditions required experimental data on the following directions:

- fission gas release from the fuel depending on temperature and time;
- evaluation of possibility to use the kinetic relations describing oxidation of unirradiated claddings for irradiated;
- getting more precise data on kinetic relations describing cladding oxidation depending on burn-up;
- measurement of the mechanical properties of the irradiated claddings;
- substantiation of the license embrittlement criterion (1200°C, 18% ECR);
- determination of the claddings rupture parameters (hoop deformation, pressure, temperature and time) in the first and second LOCA stage loading conditions.

Data on all of these directions are submitted.

Fission gas release

Fission gas release was studied on the electrically heated facility [2]. Fragments of the pellets and fuel rods were tested in different environments and at different temperatures (tab. 1). For example, experimental results characterizing FGR kinetics in the inert environment depending on temperature are shown in figs.1 and 2. The analysis of the results proves that they mainly coincide with the data obtained for the PWR fuel. The results can be used for improving codes designed for the analysis of fuel behaviour under accident conditions and calculation of the dynamics of the cladding internal pressure change. In addition to fission gas release research the fuel structure change at high temperatures was investigated.
Oxidation kinetics

The kinetics of the irradiated cladding oxidation was studied using electrically heated facility [3]. The research was carried out by RIAR and VNIINM specialists. The specimens were manufactured from the VVER-type spent fuel rods. The experiment conditions are presented in tab. 2. Some results are shown in fig.3. It also presents the data on two-side oxidation of Zry-4 unirradiated claddings [4].

Kinetics of the Zr1%Nb unirradiated cladding oxidation coincides well with VNIINM data [5].

The results prove that in the temperature range (1000-1100)°C rate of Zr1%Nb irradiated claddings oxidation increases insignificantly during the first few minutes as compared with unirradiated claddings. However, this difference disappears after a while. The oxide film structures of both irradiated and unirradiated Zr1%Nb claddings are the same and change due to oxidation temperature. Comparison of the oxidation kinetics for Zr1%Nb and Zircaloy alloys showed that up to 10 minutes the Zr1%Nb oxidation rate increases insignificantly at temperatures (1000-1100)°C and at 1200°C the oxidation rate for both alloys is the same. Comparison of the oxidation kinetics for irradiated and unirradiated Zr1%Nb claddings reveals the following:

- the irradiated claddings oxidation rate slightly exceeds the oxidation rate of unirradiated claddings during the first few minutes of oxidation in the temperature range (1000-1100)°C;
- at 1200°C oxidation rate of irradiated claddings becomes less than the oxidation rate of unirradiated claddings.

Mechanical properties of the claddings

The technique was adjusted and the research of mechanical properties of the VVER claddings (ring samples) in the temperature range (20-900)°C was carried out [6]. The research was carried out by RIAR and IRTM RRC "KI" specialists. The temperature dependence of the short-term mechanical properties of the VVER-1000 claddings both unirradiated and irradiated up to burn-up of about 50 MWd/kgU are presented in fig.4.

Main results are following:

- VVER irradiated claddings preserve high ductility in the temperature range (20-900)°C;
- at the temperature of irradiation defect annealing the mechanical properties of irradiated claddings don't differ from the mechanical properties of unirradiated claddings.
Validation of the embrittlement criterion for Zr1%Nb claddings

The research was carried out by RIAR and VNIINM specialists. The regime of thermal shock tests is presented in fig.5.

The simulators were made from spent fuel rods. The two-side oxidation of the claddings was realized in the experiments. The oxidized simulators were rapidly moved to the vessel, filled with distilled water of room temperature by time less than 1 s. In all experiments the specimens were of the same geometry (fig.6).

The simulators with oxidized claddings were tested for ability to withstand the thermal shock (quenching) and to keep the mechanical strength after LOCA sufficient for further manipulation with FA (disassembly, transportation out of hot cell, handling).

The experimental parameters and some experimental results are given in tab. 3. The pre-test and post-test appearance of the simulators are shown in fig. 7. The thermal shock tests results are presented in fig.8.

The main result of the carried out thermal shock tests is the experimental confirmation of the embrittlement criterion "1200°C PCT - 18 % ECR" for Zr1%Nb fuel rod claddings of VVER-type.

Rupture of the VVER-type irradiated claddings (during disassembly, during transportation out of hot cell, during handling) oxidized at (800 - 1100)°C temperatures had place outside of the cladding's allowable state region (1200°C - 18% ECR). No claddings were ruptured upon quenching. The expected maximal permissible temperature for VVER-type irradiated claddings will be not less than 1100°C. The experiments with the purpose of substantiation of the embrittlement criterion parameters values for VVER-type irradiated Zr1%Nb claddings are being continued.

Research of the fuel rod cladding rupture parameters in the first LOCA stage conditions

The first LOCA stage is characterized by quick heating of the cladding at a rate of 150-200°C/s and rapid decrease in the coolant pressure. The danger of this stage is in potential cladding rupture due to excessive internal cladding pressure. The task of the experiments is to define the claddings rupture parameters (hoop strain, temperature, pressure, time) typical for this accident stage. The experiments include two stages. The first stage includes rupture tests on irradiated and unirradiated claddings loaded with internal pressure. The results of these tests, shown in figs.9, 10, allowed to determine the following:

➢ the cladding rupture temperature monotonously decreases depending on pressure. The rupture parameters of irradiated and unirradiated claddings don’t differ;
claddings rupture hoop strain decreases monotonously in the temperature range of (800-1000)°C and then monotonously increases in the temperature range of (1000-1200)°C. The behaviour of irradiated and unirradiated claddings is the same.

The second stage of the experiments was carried out to prove that behaviour of the fuel rod claddings with complicated geometry and with complex loading will correlate with the results of two-axial tests. Up to now no attempts were undertaken to simulate this accident stage. That's why a special facility was developed where fuel rods are directly heated (fig.11). Four irradiated fuel rod simulators were tested under conditions simulating the first LOCA stage (figs.12, 13, tab. 4).

The results of these tests are shown in fig.14-18 and in tab. 5. In every test the cladding diameter was reduced and then the cladding was subjected to ballooning when the sign of the "cladding-coolant" pressure difference changed (fig.15-18). Three of four fuel rods (with greater initial pressure) had depressurized during the experiment (tab. 5). Analysis of the obtained results hasn't been completed yet. The obtained data show that rupture parameters (hoop strain, temperature, pressure, time) is relatively independent from pre-history of the loading (fig.19). However, it can be assumed that hoop strain of the claddings depends on the loading pre-history.

**Fuel rod cladding rupture parameters in the second LOCA stage**

One more special testing facility was developed to study long-term LOCA stage. The radiant fuel rod simulator heating method was used in these experiments. Fuel rod simulators made from VVER-440 spent fuel rods were tested. Fuel burn-ups were about 54-60 MWd/kgU.

The post-test parameters are shown in tab. 5. The experimental conditions and some results are presented in tab. 6. The simulator temperature and pressure difference distribution are shown in fig.20.

The analysis of the obtained results showed that the claddings had not ruptured up to temperature 1100°C in the experiment where the external pressure exceeded the internal or the internal pressure insignificantly exceeded the external.

In the experiments with exceeding internal "ballooning", longitudinal cracks formed and fission product released into the coolant (tab. 6). Data on influence of high temperature creep on the cladding rupture parameters in LOCA conditions were obtained in these tests. The dependence of time to rupture on the rupture temperature at relatively constant pressure difference was defined. These claddings properties must be taken into account in the temperature range (700 – 900)°C.

Four integral experiments with VVER-type fuel rods were carried out under LOCA conditions simulating the core drying (tab. 7). Among them there was only one experiment with one spent fuel rod located in the centre of seven element fuel rod assembly [6].
The purpose of the tests was to compare the behaviour of unirradiated and irradiated fuel rod claddings.

Unirradiated fuel rods were equipped with thermocouples (both the fuel column and cladding). "Fresh" pellets were with U-235 enrichment of 2% to make the linear power of "fresh" and spent fuel approximately the same.

The fuel rod №4 fuel column temperature and fuel rod №2 cladding temperature are shown in fig.21. The coolant pressure during the tests exceeded the fuel rod internal pressure. The maximal temperature of the fuel and cladding was within the range of 900...1000°C. The drying period was about 30 minutes.

The post-test cladding mechanical properties which were obtained using the ring specimens are shown in tab.8.

The behaviour of unirradiated and irradiated claddings under LOCA conditions (temperature up to 900°C and exceeding external pressure) is identical.

Conclusion

Main experimental research results on high burn-up fuel rod behaviour under LOCA conditions are the following:

1. Relative fission product release increases from 15 % to 100 % with temperature rise from 1000 to 2500°C. Fuel with burn-up of about 60 MWd/kgU is characterized by monotonously increasing FGR in the temperature range from 1000 to 1950°C. Fuel with burn-up of about 16 MWd/kgU is characterized by almost zero FGR up to 1600°C. FGR kinetics at temperatures above 1950°C for all burn-ups don't differ.

2. The oxidation rate of irradiated VVER-type claddings slightly exceeds the unirradiated claddings oxidation rate during first few minutes of oxidation at temperature 1000°C. At higher temperature level this difference reduces and at the temperature 1200°C the insignificant reverse effect is detected.

3. Short-term mechanical properties of irradiated Zr-1%Nb claddings are characterized by high ductility (δ\text{uniform}=5 %, δ\text{total}=20 %) even at room temperature. At typical LOCA temperatures (T>600°C) the irradiated cladding mechanical properties don't differ from the properties of unirradiated claddings.

4. Experimental substantiation of the embrittlement criterion confirmed that Zr1%Nb claddings don't fail (during quenching, during disassembly, during transportation out of hot cell, during handling) up to 1100°C. Further experiments should be continued in temperature range up to 1200°C.
Different types of tests were carried out with the purpose of defining the rupture parameters (hoop strain, temperature, pressure, time) in typical for first and second LOCA stages loading conditions.

References


3. Кунгурцев И. А., Смирнов В. П., Жителев В. А., Стулина Л. Н., Чесанов В. В., Пимонов Ю. И., Кузьмин И. В., Звир Е. А., Андреева-Андриевская Л. Н., Соколов Н. Б. Исследование кинетики окисления при температуре 1000 °C в паро-аргоновой среде образцов оболочки твэла ВВЭР-440, отработавшего до выгорания 42.2 МВт-сут/кг U. Сборник докладов 5-й межотраслевой конференции по реакторному материаловедению, Т. 1, ч. 1, стр. 270-280, Димитровград, 1997.


Parameters and results of the fission product release experimental research on VVER spent fuel

<table>
<thead>
<tr>
<th>Environment</th>
<th>Temperature range, °C</th>
<th>Burn-up, MWd/kgU</th>
<th>Specimen</th>
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<tbody>
<tr>
<td>argon</td>
<td>1000 - 2200</td>
<td>15,8</td>
<td>Fuel pellet fragment</td>
</tr>
<tr>
<td>helium</td>
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<td></td>
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<td>800 - 2000</td>
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<td></td>
<td>1000 - 2400</td>
<td>59,8</td>
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</tr>
<tr>
<td>argon</td>
<td>1000 - 2100</td>
<td>64,0</td>
<td>Fuel rod fragment</td>
</tr>
<tr>
<td>argon</td>
<td>1250, 1450</td>
<td>15,8</td>
<td></td>
</tr>
<tr>
<td>air</td>
<td>600 - 1200</td>
<td>36,8 *</td>
<td>fuel pellet fragment</td>
</tr>
<tr>
<td></td>
<td>600 - 2200</td>
<td>0 *</td>
<td></td>
</tr>
<tr>
<td>argon + steam</td>
<td>600-1300</td>
<td>55,2</td>
<td>fuel rod fragment and</td>
</tr>
<tr>
<td></td>
<td></td>
<td>51,7</td>
<td>fuel pellet fragment</td>
</tr>
<tr>
<td></td>
<td></td>
<td>50</td>
<td></td>
</tr>
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</table>

* specimens are subjected to additional irradiation in the MIR reactor
Fig. 1: Dependences of Kr-85 release on time.
Fuel pellet fragments with burn-up of 59.8 MWD/kgU

Fig. 2. Temperature dependence of Cs release
Parameters of the oxidation experiments on VVER-type Zr1%Nb claddings

<table>
<thead>
<tr>
<th>Burn-up, MWd/kgU</th>
<th>Isothermal temperature, °C</th>
<th>Isothermal exposure, min</th>
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<td>47.1</td>
<td>800</td>
<td>5, 30, 53, 60, 120, 300, 420</td>
</tr>
<tr>
<td>0</td>
<td>800</td>
<td>5, 30, 60, 120, 400</td>
</tr>
<tr>
<td>42.2</td>
<td>1000</td>
<td>0.2, 3, 4, 5, 7, 10, 20, 25, 30, 40, 60</td>
</tr>
<tr>
<td>0</td>
<td>1000</td>
<td>10, 30, 60</td>
</tr>
<tr>
<td>42.2</td>
<td>1100</td>
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</tr>
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<td>1100</td>
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</tr>
<tr>
<td>48.0</td>
<td>1200</td>
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<td>1200</td>
<td>0</td>
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<td>48.0</td>
<td>1200</td>
<td>4.85</td>
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<td>47.1</td>
<td>1200</td>
<td>6.10</td>
</tr>
<tr>
<td>48.0</td>
<td>1200</td>
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<td>1200</td>
<td>15</td>
</tr>
<tr>
<td>48.0</td>
<td>1200</td>
<td>30</td>
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<tr>
<td>0</td>
<td>1200</td>
<td>0.2, 3, 4, 6, 10, 12, 15, 30</td>
</tr>
<tr>
<td>49.8</td>
<td>1300</td>
<td>2.5, 15</td>
</tr>
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Fig. 3. Dependence of weight gain of Zr-1%Nb claddings on time, burnup and temperature
Fig. 4. Dependence of mechanical properties of unirradiated and irradiated (48.5 MWd/kgU) VVER claddings (ring specimens)
Fig. 5. Temperature regime of thermal-shock tests

Fig. 6. Fuel rod simulator

1 — simulator head, 2 — cladding, 3 — fuel column, 4 — fuel column catch
Fig. 7. Pre-test and post-test VVER-1000 simulator appearance (T=1100 °C)
<table>
<thead>
<tr>
<th>№</th>
<th>Specimen type</th>
<th>Isothermal temperature, °C</th>
<th>Burn-up, MWd/kgU</th>
<th>Isothermal exposure, min</th>
<th>ECR, %</th>
<th>Specimen state</th>
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<td>VVER</td>
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<td>14</td>
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<tr>
<td>4</td>
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<td>1000</td>
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<td>40</td>
<td>14</td>
<td>non-failed</td>
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<tr>
<td>5</td>
<td>VVER</td>
<td>1000</td>
<td>47,1</td>
<td>40</td>
<td>14</td>
<td>non-failed</td>
</tr>
<tr>
<td>6</td>
<td>VVER</td>
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<td>40</td>
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<td>7</td>
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<td>8</td>
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<td>9</td>
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<td>49,2</td>
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<tr>
<td>10</td>
<td>VVER</td>
<td>1085</td>
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<td>&gt;20</td>
<td>failed</td>
</tr>
<tr>
<td>11</td>
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<td>1085</td>
<td>49,2</td>
<td>60</td>
<td>&gt;20</td>
<td>failed</td>
</tr>
<tr>
<td>12</td>
<td>VVER</td>
<td>1100</td>
<td>50,1</td>
<td>40</td>
<td>21</td>
<td>failed</td>
</tr>
<tr>
<td>13</td>
<td>VVER</td>
<td>1100</td>
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<td>30</td>
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<td>failed</td>
</tr>
<tr>
<td>14</td>
<td>VVER</td>
<td>1100</td>
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<td>15,9</td>
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<td>37</td>
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<td>700</td>
<td>21,8</td>
<td>420</td>
<td>&lt; 4,5</td>
<td>non-failed</td>
</tr>
<tr>
<td>19</td>
<td>RBMK under SG</td>
<td>700</td>
<td>21,8</td>
<td>420</td>
<td>&lt; 4,5</td>
<td>non-failed</td>
</tr>
<tr>
<td>20</td>
<td>RBMK between SG</td>
<td>700</td>
<td>22,6</td>
<td>420</td>
<td>&lt; 4,5</td>
<td>non-failed</td>
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<tr>
<td>21</td>
<td>RBMK under SG</td>
<td>700</td>
<td>22,6</td>
<td>420</td>
<td>&lt; 4,5</td>
<td>non-failed</td>
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<td>22</td>
<td>RBMK between SG</td>
<td>700</td>
<td>21,8</td>
<td>300</td>
<td>&lt; 4,5</td>
<td>non-failed</td>
</tr>
<tr>
<td>23</td>
<td>RBMK under SG</td>
<td>700</td>
<td>21,8</td>
<td>300</td>
<td>&lt; 4,5</td>
<td>non-failed</td>
</tr>
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<td>24</td>
<td>RBMK between SG</td>
<td>700</td>
<td>22,6</td>
<td>300</td>
<td>&lt; 4,5</td>
<td>non-failed</td>
</tr>
<tr>
<td>25</td>
<td>RBMK under SG</td>
<td>700</td>
<td>22,6</td>
<td>300</td>
<td>&lt; 4,5</td>
<td>non-failed</td>
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<tr>
<td>26</td>
<td>RBMK between SG</td>
<td>750</td>
<td>21,8</td>
<td>300</td>
<td>&lt; 4,5</td>
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<td>27</td>
<td>RBMK under SG</td>
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<td>21,8</td>
<td>300</td>
<td>&lt; 4,5</td>
<td>non-failed</td>
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<tr>
<td>28</td>
<td>RBMK between SG</td>
<td>750</td>
<td>22,6</td>
<td>300</td>
<td>&lt; 4,5</td>
<td>non-failed</td>
</tr>
<tr>
<td>29</td>
<td>RBMK under SG</td>
<td>750</td>
<td>22,6</td>
<td>300</td>
<td>&lt; 4,5</td>
<td>non-failed</td>
</tr>
</tbody>
</table>
Fig. 8. Failure map for Zr-1%Nb irradiated claddings after thermal-shock tests

Fig. 9. Rupture pressure as a function of temperature
Fig. 10. Rupture hoop strain of irradiated claddings as a function of temperature

1,8 - container with argon; 2,4,6,7,9,14,17,20 - valve; 3 - bulb for pressure rate regulation; 5 - bulb for pressure stabilization; 10 - pressure detector; 11 - housing; 12 - heated screen; 13 - fuel rod simulator; 15 - thermocouple; 19 - bulb for gas drop; 21 - aerosol filter

Fig. 11. Scheme of electrically heated testing facility designed for study of the first LOCA stage
Fig. 12. Fuel rod-coolant pressure difference
(LOCA scenario)

Fig. 13. Temperature of unirradiated fuel simulator as a function of time
### Parameters of the first LOCA stage scenario-1

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Simulator number</th>
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</thead>
<tbody>
<tr>
<td>Fuel rod pressure, MPa</td>
<td>1</td>
</tr>
<tr>
<td>Average fuel rod burn-up, MWd/kgU</td>
<td>5</td>
</tr>
<tr>
<td>Time $\tau_0$ at $\Delta P_{10-s}=0$, s</td>
<td>10.3</td>
</tr>
<tr>
<td>Cladding temperature corresponding to time $\tau_0$, °C</td>
<td>860</td>
</tr>
<tr>
<td>Fuel rod-coolant pressure difference at $\tau=0$ s, MPa</td>
<td>11</td>
</tr>
<tr>
<td>Fuel rod-coolant pressure difference at $\tau=0.1$ s, MPa</td>
<td>6</td>
</tr>
<tr>
<td>Fuel rod-coolant pressure difference at $\tau=30$ s, MPa</td>
<td>4.5</td>
</tr>
</tbody>
</table>

![Simulator 2](image1) ![Simulator 3](image2) ![Simulator 4](image3)

**Fig. 14. Claddings appearances and their cross-sections in the depressurization areas**
Fig. 15. Outer diameter distribution
along the length of unirradiated simulator 1

Fig. 16. Outer diameter distribution
along the length of unirradiated simulator 2
Fig. 17. Outer diameter distribution along the length of unirradiated simulator 3

Fig. 18. Outer diameter distribution along the length of unirradiated simulator 4
Fig. 19. The dependence of claddings rupture pressure on temperature
### Table 5

**Post-test rupture (depressurization) parameters of unirradiated simulator claddings**

<table>
<thead>
<tr>
<th>Rupture parameter</th>
<th>Simulator number</th>
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<tbody>
<tr>
<td></td>
<td>1</td>
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<tr>
<td>Cladding state</td>
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<tr>
<td>Pressure difference, MPa</td>
<td>-</td>
</tr>
<tr>
<td>Cladding temperature, °C</td>
<td>-</td>
</tr>
<tr>
<td>Pre-depressurization time, s</td>
<td>-</td>
</tr>
<tr>
<td>Maximal hoop strain, %</td>
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</tr>
<tr>
<td>Ballooning coordinate, mm</td>
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</tr>
<tr>
<td>Crack length, mm</td>
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</tr>
<tr>
<td>Crack width, mm</td>
<td>-</td>
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### Table 6

**Conditions of the LOCA simulating experiments with single fuel rods**

<table>
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<tr>
<th>Exp. Nr</th>
<th>External pressure, MPa</th>
<th>Specimen temperature, °C</th>
<th>Internal pressure, MPa</th>
<th>Fuel burn-up, MWd/kgU</th>
<th>Time to rupture, s</th>
<th>FGR, %</th>
<th>Kr-85</th>
<th>Cs-134</th>
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<td>0.1</td>
<td>59.2</td>
<td>-</td>
<td></td>
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<tr>
<td>2</td>
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<td>1100</td>
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<td>-</td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>3</td>
<td>0.1</td>
<td>800...1200</td>
<td>1.5</td>
<td>56.5</td>
<td>485</td>
<td>1.5</td>
<td>3.2</td>
<td>2.4</td>
<td></td>
</tr>
<tr>
<td>4</td>
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<td>700</td>
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<td>1836</td>
<td>0.6</td>
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<td>3.9</td>
<td>60.5</td>
<td>1440</td>
<td>1.3</td>
<td>4.2</td>
<td>2.9</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>0.1</td>
<td>1000</td>
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<td>54.4</td>
<td>2</td>
<td>3.9</td>
<td>3.6</td>
<td>2.0</td>
<td></td>
</tr>
</tbody>
</table>
Fig. 20. Dependence of temperature and internal pressure on time

- a - temperature, b - pressure, 1-6 - experiment number
<table>
<thead>
<tr>
<th>Experiment number</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of unirradiated fuel rods in the FA</td>
<td>18</td>
<td>19</td>
<td>19</td>
<td>6 and 1 spent one</td>
</tr>
<tr>
<td>Coolant parameters:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>• pressure, MPa;</td>
<td>12</td>
<td>12</td>
<td>4</td>
<td>6</td>
</tr>
<tr>
<td>• temperature on outlet, °C</td>
<td>~320</td>
<td>~320</td>
<td>~80</td>
<td>~270</td>
</tr>
<tr>
<td>• flow rate, kg/s</td>
<td>0.04</td>
<td>0.04</td>
<td>0.04</td>
<td>0.008</td>
</tr>
<tr>
<td>FA power, kW:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>• steady state</td>
<td>500</td>
<td>500</td>
<td>500</td>
<td>100...120</td>
</tr>
<tr>
<td>• LOCA</td>
<td>≤100</td>
<td>≤100</td>
<td>≤70</td>
<td>~35</td>
</tr>
<tr>
<td>Maximal fuel rod temperature, °C</td>
<td>550*</td>
<td>1200</td>
<td>720</td>
<td>920</td>
</tr>
<tr>
<td>Maximal temperature exposure, min</td>
<td>72</td>
<td>3</td>
<td>25</td>
<td>6</td>
</tr>
<tr>
<td>Post-test FA state</td>
<td>non-failed</td>
<td>ruptured</td>
<td>ruptured</td>
<td>ruptured</td>
</tr>
</tbody>
</table>

* - short-term temperature of 950 °C was reached on one of the fuel rods.
Fig. 21. Temperature regime of FA tests under LOCA conditions
<table>
<thead>
<tr>
<th>Fuel rod</th>
<th>Specimen coordinates, mm</th>
<th>Test temperature, °C</th>
<th>Ultimate stress $\sigma_{br}$ MPa</th>
<th>Yield stress $\sigma_{0.2}$ MPa</th>
<th>Uniform elongation $\delta_{uniform}$ %</th>
<th>Total elongation $\delta_{total}$ %</th>
</tr>
</thead>
<tbody>
<tr>
<td>7</td>
<td>315-345</td>
<td>20</td>
<td>472</td>
<td>423</td>
<td>7.2</td>
<td>25.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>350</td>
<td>272</td>
<td>233</td>
<td>7.6</td>
<td>28.2</td>
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<tr>
<td>770-800</td>
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<td>350</td>
<td>323</td>
<td>279</td>
<td>6.7</td>
<td>17.6</td>
</tr>
<tr>
<td>320-360</td>
<td>20</td>
<td>380</td>
<td>328</td>
<td>11</td>
<td>31.6</td>
<td></td>
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<tr>
<td></td>
<td></td>
<td>350</td>
<td>217</td>
<td>187</td>
<td>10.8</td>
<td>33.8</td>
</tr>
<tr>
<td>4</td>
<td>775-808</td>
<td>20</td>
<td>434</td>
<td>365</td>
<td>7.6</td>
<td>15.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>350</td>
<td>265</td>
<td>222</td>
<td>8.5</td>
<td>26.0</td>
</tr>
</tbody>
</table>
Discussion:

Question by H.M. Chung: How much were the hydrogen contents of the cladding specimens?

Answer by N. Sokolov: For 4-year fuel cycle the average value of hydrogen content is 30-60 ppm, we measured the maximum hydrogen content: 100 ppm.

Question by R. Yang: Your LOCA transient is rather long - 10 minutes. Is it typical for VVERs?

Answer by N. Sokolov: The duration of high temperature transient is the same in PWR as in VVER. It is 200 seconds. The coolant pressure in the above mentioned integral experiment is rather high (from 4 to 12 MPa). This coolant pressure corresponds to so called Small Break LOCA. For simulation of a Large Break LOCA in integral experiment it is necessary to complete some modernization of the loop.

Question by G. Hache: In 1996 Russian colleagues presented results of one integral test. Was it the same test as you have presented here?

Answer by N. Sokolov: Maybe 10 integral experiments have been carried out during last 10 years. I don't remember the result of what test was presented in 1996.
14. IPSN Analysis Of Experimental Needs Requested for Solving Pending LOCA Issues
A. Mailliat, IPSN, France

Paper summary

In France and in other countries, a permanent evolution of the light water reactors (LWR) is observed since the seventies. The evolution deals with the reactor designs (900 MWe/3 loops, 1300MWe/4 loops, N4, future EPR). It is also related to the fuel management and burnup increase (3 cycles, 4 cycles, 39 GWd/tU, 47, 52, 60 GWd/tU in the next future). This evolution affects the fuel itself (UO₂, MOX, Gd fuel), the cladding (Zircaloy, Zirlo, M5) and the control rods (Ag-In-Cd, B₄C). As a consequence of these modifications, there is a permanent necessity for reassessing the reactor safety studies which implies improving the associated knowledge and upgrading the corresponding calculation tools. Such a need is not specific to the French situation. For the studies associated with the continuous evolution of the reactor operation, the safety authorities requirements are both related to the design basis accidents and the severe accidents. They have to appreciate to which extent their analyses and criteria might be modified by the burnup increase and the type of fuel. In France, under safety considerations, it was requested prior to any generic authorisation of discharge burn-up extension, that the high burn-up fuel behaviour be validated, with the support of appropriate R&D tests results, under accidental conditions, particularly under Loss-of-Coolant-Accident (LOCA) conditions.

For many years, IPSN and several other safety organisations have applied a three-tier method for their reactor safety researches. The first step consists of computer code developments from the existing data bases. The second step involves small-scale, out-of-pile experiments, which provide the additional data bases requested by the code developments and their preliminary assessments. But, as the reactor phenomenology cannot be totally reproduced in such small scale experiments, a third step consisting of integral in-pile experiments using real materials is essential for comprehensive accident analyses. Their results allow the final code assessment in terms of reactor applicability and simulation completeness. This in-pile part of a programme assures that the investments done for code developments and small scale experiments will produce profits in terms of reactor safety.

Studies performed in IPSN and elsewhere pointed out that high burnup may induce specific effects, especially those related with fuel relocation. Uncertainties exist regarding how much these effects might affect the late evolution of the accident transient and the associated safety issues. IPSN estimates that a better knowledge of specific phenomena are required in order to resolve the pending uncertainties related to LOCA criteria.

IPSN is preparing the so called APRP-Irradié (High Burnup fuel LOCA) programme. One of the important aspect of this programme is In-Pile experiments involving bundle geometries in the PHEBUS facility located at Cadarache, France. A feasibility study for such an experimental programme is underway and should provided, by the end of this year, a finalised project including cost and schedule aspects.

The main objectives of the in-pile experiments will be to investigate the behaviour of fuel and cladding with conditions representative of the reactor during LOCA sequences. The main parameters to investigate are:

- the nature of fuel (UO₂, MOX, Burn-up),
- the nature of test rods : refabricated from actual PWR irradiated rods
- the fuel-clad thermomechanical coupling (i.e. fuel relocation)
- thermal azimuthal gradients (main factor affecting cladding strain and blockage ratio)
- thermal-hydraulic aspects (i.e. quenching, coolability of blocked arrays)

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4 Mean value per assembly
These tests should be realised with 9 high burn up rods with a ring of 12 or 16 fresh fuel rods which will provide a representative thermal environment in order to ensure representative strains and subsequent phenomena. A blowdown phase will be simulated depending on its impact on the relocation process deduced from the previous studies. Finally, additional axial stress during quenching due to rod blockage in the assembly should be simulated during these tests.
IPSN Analysis of Experimental Needs
For Solving LOCA Pending Issues

Alain MAILLIAT, Claude GRANDJEAN, Georges HACHE
Institut de Protection et de Sûreté Nucléaire
Département de Recherches en Sécurité
C.E., CADARACHE, France

SUMMARY

Studies performed in IPSN and elsewhere pointed out that high burnup may induce specific effects, especially those related with fuel relocation. Uncertainties exist regarding how much these effects might affect the late evolution of the accident transient and the associated safety issues. IPSN estimates that a better knowledge of specific phenomena are required in order to resolve the pending uncertainties related to LOCA criteria. IPSN is preparing the so called APRP-Irradié (High Burnup fuel LOCA) programme. One of the important aspect of this programme is In-Pile experiments involving bundle geometries in the PHEBUS facility located at Cadarache, France. A feasibility study for such an experimental programme is underway and should provided, by the end of this year, a finalised project including cost and schedule aspects.

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LOCA Related Safety Criteria

The current regulatory safety criteria for LOCA, still in use in most countries, are derived from the ECCS acceptance criteria that were issued by USAEC in December 1973 and published in the Code of Federal Regulations (10.CFR50, part 50.46) as "Acceptance Criteria for Emergency Core Cooling Systems for Light-Water-Cooled-Nuclear Power Reactors". The criteria are stated as 5 requirements, concerning the calculated performance of the cooling system under the most severe loss-of-coolant accident conditions. These requirements address:

1. Peak Cladding Temperature (PCT)
   shall not exceed 2200 F (= 1477 K);

2. Maximum Cladding Oxidation Rate (Equivalent Cladding Reacted =ECR)

\(^1\) Mean value per assembly
shall nowhere exceed 17% of the cladding thickness before oxidation (but after cladding swelling with or without rupture);

3. Maximum Hydrogen Generation
total amount shall not exceed 1% of the hypothetical amount generated by the reaction of all the metal in the cladding cylinders surrounding fuel;

4. Coolable Geometry
calculated changes in core geometry shall leave the core amenable to cooling;

5. Long-term Cooling
after any operation of the ECCS, the core temperature shall be maintained at an acceptably low value and decay heat removed for the extended period of time required by long-lived radioactivity;

The oxidation limit (ECR<17%) was based on the available results of different series of quenching experiments, performed on as-received cladding only. Furthermore, the statement of the requirement on Maximum Cladding Oxidation implies that this requirement applies to non corroded (i.e. unirradiated) cladding at the initiation of LOCA transient.

Consequently, the French practice for design studies involving LOCA calculations with irradiated fuel consists to apply the 17% limit to the cumulated oxidation rate, including initial corrosion during in-reactor operation and high temperature oxidation (after clad swelling and rupture) during LOCA transient.

Uncertainties and Pending Issues

In the aftermath of the ABC LOCA criteria release, numerous studies were undertaken world-wide in order to improve the basic knowledge of the physical phenomena intervening in LOCA transients, so as to allow a better prediction with realistic models. Beyond the numerous experimental investigations that were conducted on unirradiated rods or cladding, either in-pile or out-of-pile, there exists a few number of available results of such experiments with irradiated material. Following is a very short review of the current knowledge on clad and fuel rod behaviour gained from experiments on irradiated material, that will introduce the pending questions and critical issues for irradiated fuel behaviour in LOCA.

Clad behaviour

An important progress in knowledge relative to irradiated clad behaviour has been obtained from the results of the French EDF/IPSN [1,2] program (TAGCIR and HYDRAZIR tests), addressing the oxidation kinetics and quench bearing capability of irradiated zircaloy. The main outcome concern:
- the protective effect of corrosion oxide scale;
- the oxidation kinetics of irradiated zircaloy;
- the resistance to quench loads of irradiated zircaloy;
- the effect of high hydrogen content, as a result of internal hydriding during LOCA transient.

Relative to oxidation kinetics and quench behaviour, a comprehensive understanding of all involved phenomena and of their inter-related influences is not yet achieved and leaves still pending questions, most of them being not specific to high BU fuel. One important question is the influence on clad quenching resistance of axial constraints that may result from differential contractions upon quench between guide tubes and a fuel rod blocked in spacer grids as a result of ballooning or chemical interaction: such an effect had been evidenced on past tests at JAERI[3] on unirradiated rods and should therefore be expected to some extent on irradiated rods.
Rod behaviour

There exists a few number of available results of experiments with irradiated fuel rods under LOCA conditions: the main outcome were found in results from the PBF-LOC tests[4,5] in the USA, the FR2 tests[6] in Germany, and the FLASH5 test[7] in France; they concern the two following phenomena:

A) Fuel relocation

In all irradiated rods of the PBF-LOC, FR2 and FLASH5 tests, fuel relocation has occurred as a result of slumping of pellets fragments from upper locations into the swollen region of the burst cladding. Fuel relocation phenomena is not restricted to high burn-up fuel since fuel fragmentation occurs as soon as low burn-up levels (it was thus noticed on LOC5-7B rod, fresh rod pre-conditioned up to 48 MWd/t).

It was demonstrated on FR2 tests E3 and E4 that the fuel movement initiation occurred at the time of cladding burst, driven by the pressure difference between rod plenum and coolant channel with assistance of gravity slumping. The fuel movement was possibly favoured in FR2 experiments where the fuel-cladding gap was not totally closed; due to the rod inverted internal-external pressure difference during initial irradiation at low temperature. A tight bonding between fuel and cladding is supposed to counteract the fuel motion inception. However, in FLASH5 experiment with high burn-up fuel (50 GWd/t), and in spite of a low clad ballooning (not higher than 16%) post-test examinations have shown that fuel fragments were no more stuck to the cladding: the transient temperature rise combined to clad deformation may be sufficient to suppress fuel-cladding bonding.

B) Cladding deformation

In PBF-LOC experiments 2 couples of rods (2 unirradiated $+$ 2 irradiated) were simultaneously tested in the same test train. Available data for comparison, although in very limited number due to technical problems, clearly indicate significant differences in the deformation behaviour of irradiated versus unirradiated rods:

- a higher circumferential rupture strain for irradiated rods (a factor greater than 2 relatively to unirradiated rod strain for maximum values) and more axially extended;
- a wall thinning affecting almost all the circumference of irradiated rods, thus indicating low azimuthal temperature differences as compared to unirradiated rods;

These differences in behaviour have been attributed to the lower temperature differences on the clad of irradiated rods, circumferentially and axially, as a result of the pellet-clad gap reduction due to clad creepdown during rod irradiation.

The results of FR2 experiments do not indicate any dependence of circumferential strain on maximum azimuthal temperature difference at burst elevation, thus appearing inconsistent with PBF-LOC and out-of-pile results. However a thorough examination of tests conditions has shown that this unexpected result could be explained as the consequence of non prototypic test conditions (single rod within cold narrow shroud limiting rod bowing, axial stresses due to temperature profile and a large plenum spring) and of the unrepresentative pre-irradiation conditions. As a conclusion the important effect evidenced by PBF-LOC tests results should not be disregarded in consideration of FR2 results.

A better understanding of the specific phenomena identified above leads to raise some complementary questions regarding:

1) Clad Deformation
   - influence of hydrogen pick-up and other irradiation effects on ballooning and burst behaviour?

2) Fuel relocation
   - instant of fuel movement at high burn-up, with possible delay due to fuel-clad bonding?
   - filling ratio of clad balloon at high burn-up, with fragmentation of UO2 rim or MOX clusters?
   - filling ratio for large cladding deformations?
- effect on peak clad temperature and final oxidation ratio of the local increase in lineic and surfacic power and of the local decrease in fuel-clad gap resulting from fuel accumulation?

*Note that this question is particularly important for end-of-life MOX fuel where power generation is not reduced, unlike for UO2 fuel.*

Related questions should be considered additionally, relative to flow blockage behaviour of highly deformed cladding with possibly relocated fuel and the embrittlement potentials associated to fuel fragmentation:

3) Flow blockage

- what is the maximum flow blockage ratio that leaves coolable an irradiated rods bundle?

*The 90% value derived from results of flooding experiments (FEBA, SEFLEX et al) on unirradiated rods arrays is questionable since these experiments did not take account of any fuel relocation and associated effects.*

- does the maximum flow blockage ratio attainable with an irradiated rods array remain below the maximum coolable value indicated above?

*There is presently a complete lack of data allowing to answer this*

- what flow blockage configuration would be worst coolable with occurrence of fuel relocation?

*In other words, is the coplanar flow blockage still the worst coolable case?*

D) Embrittlement of high BU fuel rod

- potential for fuel swelling under fission gas expansion due to overheating in central fragments?

- potential for PCMI constraints upon quench, when clad balloon is closely filled with fine fuel fragments?

The IPSN APRP Irradié Project

For many years, IPSN and several other safety organisations have applied a three-tier method for their reactor safety researches. The first step consists of computer code developments from the existing data bases. The second step involves small-scale, cut-of-pile experiments, which provide the additional data bases requested by the code developments and their preliminary assessments. But, as the reactor phenomenology cannot be totally reproduced in such small scale experiments, a third step consisting of integral in-pile experiments using real materials is essential for comprehensive accident analyses. Their results allow the final code assessment in terms of reactor applicability and simulation completeness. This in-pile part of a programme assures that the investments done for code developments and small scale experiments will produce profits in terms of reactor safety.

This three-tier method is applied by IPSN for the various research programmes devoted to reactor safety, design basis accidents, RIA and severe accidents programmes. The in-pile part of the LOCA programme, with the collaboration of Japan (JAERI), consisted of 20 LOCA tests run from 74 to 84 [10, 16]. It provided a large spectrum of results for the assessment of codes like CATHARE, FRETA (J), RELAP (US), etc. The studies devoted to the early phase of core degradation during a severe accident, made with the collaboration of Germany (GRS, IRE) and Spain (CSN, CIEMAT), included the CSD programme in the Phébus facility: six experiments where run from 78 to 89, see [17, 20]. The results were used to validate the ICARE (F), KESS (G) and SCDAP (US) codes. Since 93, the Phébus-FP programme [21, 24] is under way in collaboration with the European Commission, the US (NRC), Canada (COG, AECL), Japan (NUPEC, JAERI), Korea (KAERI) and Switzerland (PSI, HSK). The results of the first three tests are used to validate the codes in the field of core degradation, FP and structure materials transport and chemistry in the primary circuit and the reactor containment. The outcome of the programme demonstrates the importance of such in-pile experiments. Actually, the results of Phébus-FP lead to change or improve several models in most of the codes used by the nuclear safety community to get more realistic estimates of core degradation and hydrogen
production, radioactive material deposits in the primary circuit components and iodine chemistry both under gaseous and aqueous forms.

Returning now to the LOCA question, and the current testing programmes dealing with irradiated material keep limited to out-of-pile experiments:

- Separate effect quench tests on irradiated cladding (TAGCIR tests) in France;
- Oxidation tests on irradiated cladding and integral type experiments (ballooning / burst / oxidation / quench) on irradiated rods at ANL (USA) [8] with the support of an important programme of mechanical tests;
- Oxidation tests and burst tests on irradiated cladding, and integral type experiments (ballooning / burst / oxidation / quench) on irradiated rods at JAERI (Japan) [9] again with the support of separate effect mechanical testing

But these programmes, will not solve the previously mentioned uncertainties because these ones are mainly associated with the combined behaviour of fuel and cladding under representative conditions of the reactor conditions during the LOCA transient.

Based on the long fruitful experiences of a three-tier method, the so called APRP Irradié programme, providing the third tier in terms of the In-Pile experiments, should provide the missing part of the databases required for code assessments in terms of reactor applicability and simulation completeness. This programme is prepared in a coherent way with the present international efforts in order to validate, and possibly update, the results obtained from separate effects tests and previous limited in-pile tests.

THE MAIN OBJECTIVES

The main objectives of the in-pile experiments will be to investigate the behaviour of fuel and cladding with conditions representative of the reactor during LOCA sequences. The main parameters to investigate are
- the nature of fuel (UO2, MOX, Burn-up),
- the nature of test rods: refabricated from actual PWR irradiated rods
- the fuel-clad thermomechanical coupling (i.e. fuel relocation)
- thermal azimuthal gradients (main factor affecting cladding strain and blockage ratio)
- thermal-hydraulic aspects (i.e. quenching, coolability of blocked arrays)

IN-PILE TESTS

In-Pile experiments are a necessity according to the following points.

In pile tests provide the unique way to maintain the correct heat generation, which simulates the residual power, in the fuel fragments whatever are the relocations induced by the ballooning and/or the burst of the rod.

This heat generation correctness is one of the conditions for having realistic estimates of the relocation consequences in terms of equivalent clad reacted, peak clad temperature and hydrogen uptakes inside the balloon. All these aspects impact the strength of the rod during the quenching phase and the residual ductility of the rod after the LOCA transient.

During the blowdown phase of the LOCA transient, there is much less heat generation in the fuel and the clad coolant heat transfer is drastically reduced. Therefore, the fuel stored energy is redistributed in the pellet and the clad. This redistribution simultaneously produces a decrease of the pellet centreline temperature from 1500°C down to, say, 1000°C and an increase of both the pellet rim and clad temperatures from 300°C up to 1000°C. Due to these temperature transients, the central part of the pellet will experience a contraction while the rim and the clad will undergo an expansion. Fuel mechanical stresses and fragmentation could be induced by these adverse effects. It has to be kept in mind that during usual experiments, for which a blowdown phase is not reproduced, clad and fuel
temperatures are simultaneously increased without producing any comparable thermomechanical transient. In-pile tests including a blowdown phase provide the way to get a definitive answer regarding the additional fuel fragmentation prior to the relocation and how much this refragmentation process affects the amount and the characteristics of the relocated fuel.

**BUNDLE GEOMETRY**

In addition to the requirement associated to heat generation mentioned above, a bundle geometry is an second important condition to produce realistic data. During the early stage of the LOCA transient the fuel rods experience the ballooning and burst processes. For such phenomena, a bundle geometry is a necessity to get a correct azimuthal temperature field around the fuel rods because this parameter is a crucial one to produce a realistic balloon size. In addition, the radial interactions between adjacent fuel rods need to be included because they modify the size and shape of the balloons. Finally, the amount of relocated fuel being associated with the size and the shape of the balloon, it means that realistic data will require a bundle geometry.

Now, considering the late stage of the transient, when quenching takes place, a bundle geometry is also a necessity to produce valuable results. It is also necessary to represent the axial and radial stresses induced by the grids and the adjacent rods which might restrain the rod contraction during quenching.

Since it is hardly conceivable to carry out one type of experiments that will address all pending questions with any chance to provide some usable results, it appears more appropriate to perform two kinds of in-pile experiments:

1) Separate effects tests; addressing phenomenological aspects, in order to confirm or correct and extent the previous results relative to:
   - rod deformation,
   - fuel relocation,
   - the resulting resistance to thermal shock loads, with or without effect of clad axial constraining;

   These tests should be realised with a single irradiated rod with a ring of 8 fresh fuel rods which will provided a representative thermal environment in order to ensure representative strains and subsequent phenomena. In addition these In-Pile separate effects tests should be include a blowdown phase providing representative conditions for the residual power transient during a LOCA sequence.

2) Bundle experiments, with a more integral character, addressing the aspects of:
   - impact of blowdown phase
   - flow blockage
   - quenching behaviour and coolability,

   These tests should allow to check the absence of unexpected phenomena or unexpected coupling between foreseen processes, and finally provide data for the validation of reactor computational tools.

These tests should be realised with 9 high burn up rods with a ring of 12 or 16 fresh fuel rods which will provide a representative thermal environment in order to ensure representative strains and subsequent phenomena. A blowdown phase will be simulated depending on its impact on the relocation process deduced from the previous studies. Finally, additional axial stress during quenching due to rod blockage in the assembly should be simulated during theses tests.

Such a programme should take place in the PHEBUS Facility. By this way IPSN will take advantage of the know-how accumulated when the previous LOCA programme with fresh fuel was run. Furthermore,
a new LOCA programme in the Phébus facility will take advantage of the R&D efforts made for the subsequent programmes in terms of high activity material measurements. Tomography technique is one of the examples which can be given how such efforts provide practical applications in the new programme. Fuel relocation studies will be made easier through this technique which provides the 3D location and determine the nature of a material fragment inside a bundle. Presently, fragment size less than less than 100 microns can be located. Further enhancement of the existing technique will increase the resolution providing several points inside the clad with an oxide/metal discrimination.

CONCLUSION

Studies performed in IPSN and elsewhere pointed out that high burnup may induce specific effects, especially those related with fuel relocation. Uncertainties exist regarding how much these effects might affect the late evolution of the accident transient and the associated safety issues. IPSN estimates that a better knowledge of specific phenomena are required in order to resolve the pending uncertainties related to LOCA criteria.

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IPSN Analysis of Experimental Needs for Solving LOCA Pending Issues

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OECD LOCA Topical Meeting
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Introduction

Studies performed in IPSN and elsewhere pointed out that high burnup may induce specific effects, particularly clad enhanced deformation and fuel relocation.

Uncertainties exist regarding how much these effects might significantly affect the late evolution of the accident transient and the associated safety issues.

Better estimates of the specific phenomena identified here after are required in order to resolved the pending uncertainties related to LOCA criteria.
SUMMARY of UNCERTAINTIES and PENDING QUESTIONS

1 FUEL RELOCATION

FRESH FUEL

High Burnup FUEL

FRESH FUEL ROD
Test B 1.1

FUEL PELLET

35 GWd/τU ROD
Test G 3.2

PELLET DEBRIS

Réunion XYZ du abc
1 FUEL RELOCATION

Neutron radiographs of rod F1 (20 GWD/tU)
Pending Questions: The Parameters?

- Instant of fuel movement at high burnup?
- Are they any delay due to fuel-clad bonding?
- Filling ratio of clad balloon at high burnup, with fragmentation of UO2 rim or MOX clusters?
- Fragment sizes, corresponding conductivity?
**Pending Questions: The Consequences?**

Effect on:
- peak clad temperature
- final oxidation ratio
- hydrogen uptake

due to:
- the increase of power
- the decrease in fuel-clad gap resulting from fuel accumulation

⇒ Note that this question is particularly important for end-of-life MOX fuel where power generation is not reduced, unlike for UO2 fuel.
What is the maximum flow blockage ratio that leaves coolable an irradiated rods bundle?

The 90% value derived from results of flooding experiments (FEBA, SEFLEX et al) on unirradiated rods arrays is questionable since these experiments did not take account of any fuel relocation and associated effects (lineic and surfacic power increase, gap reduction).

Does the maximum flow blockage ratio attainable with an irradiated rods array remain below 90%?

There is presently a complete lack of data allowing to answer this question.
Uncertainties

**Pending Questions: The Consequences?**

- Which is the impact on failure during quenching if the fuel rod cannot totally accommodate the stresses due to rod blockage in the assembly?

**Pending Questions: The Consequences?**

- Which is the residual ductility of the cladding after quenching?
Such uncertainties have led IPSN to initiate new studies on the behaviour of irradiated rods and assemblies under LOCA conditions to investigate the combined behaviour of fuel and cladding under representative conditions.

This APRP-Irradié programme under project will include:

- Existing and future out-of-pile separate effects tests,
- In-pile specific experiments.
- Code developments
The main objective of the in-pile experiments

Investigate the combined behaviour of fuel and cladding with conditions representative of the reactor ones during LOCA

The parameters to investigate:

The nature of fuel (UO2, MOX, Burnup),

The fuel-clad thermomechanical coupling (i.e. fuel relocation)

Thermal azimuthal gradients (main factor affecting cladding strain and blockage ratio)

Thermohydraulic aspects (i.e. quenching, coolability of blocked arrays)
IN-PILE experiments are necessary requirement to fulfil the above representativity objectives:

- In-pile tests provide the unique way to maintain the heat generation in the fuel fragments whatever are the fuel movements induced by the relocation at burst.

- Heat generation correctness is one of the conditions for having representative estimates of the relocation consequences for ECR and PCT.

- In pile tests with a blowdown phase provide the way to get the correct fuel temperature profile transient which induce pellet radial stresses and possible effects on the amount and the characteristics of the relocated fuel.
BUNDLE experiments are necessary to fulfil representativity objectives:

To get correct azimuthal temperature field around the tested fuel

- Temperature field correctness is crucial to produce realistic balloon size
- Radial interactions between adjacent fuel rods impact the balloon size and shape
- Balloon size and shape impact the amount of relocated material
BUNDLE experiments are necessary to fulfil representativity objectives:

- To represent the axial stresses induced by the grids which restrain rod contraction during quenching
- To get a realistic value of the flow blockage and the associated heat generation induced by fuel relocation
- To get the realistic complex flow behaviour and quench front progression during reflooding
In-pile bundle experiments will complement previous in-pile tests programs (PBF-LOC, FR2, FLASH5), as well as In pile single rod or out-of-pile test programmes currently under progress or preparation.

Since it is hardly conceivable to carry out one type of experiments that will address all pending questions with any chance to provide some usable results, it appears more appropriate to perform two kinds of in-pile experiments:
Studies devoted to fuel relocation characteristics and fuel relocation consequences:

- 1 high burnup rod with a ring of 8 fresh fuel rods
- A blowdown phase which will include representative conditions for the residual power transient during a LOCA sequence.

Studies devoted to Flow blockage, Quenching behaviour and Coolability:

- 9 High burn up rods with a ring of 16 fresh fuel rods
- Additional axial stress during quenching due to rod blockage in the assembly
- Blowdown phase will be simulated depending on its impact on the relocation process deduced from the previous studies.
Such a programme should take place in the PHEBUS Facility. By this way IPSN will take advantage of the know-how accumulated when the previous LOCA programme with fresh fuel was run.

Between Years 76 and 83, 20 LOCA bundle tests were run in the PHEBUS Facility.
A new LOCA programme in PHEBUS FACILITY will take advantage of the R&D efforts made for the subsequent programmes in terms of high activity material measurements. To day we can measure the 3D location and determine the nature of a material fragment less than 100 microns in a bundle through tomography.

Enhancement of the existing technique will provide several points inside inside the clad with an oxide/metal discrimination. Such a tomography measurement associated to post test examination in hot lab will provided the requested information about relocation.
IPSN is preparing the so called *APRP-Irradié* programme devoted to high Burnup fuel LOCA pending issues.

One of the important aspects of this programme is In-Pile experiments with bundle geometries in the PHEBUS facility.

A feasibility study for such an experimental programme is underway.

A finalised project including the technical, cost and schedule aspects should be ready for discussion by end of year 2001.
Discussion:

Question by C. Vitanza: Large bundle tests are difficult and very expensive; will we need to go through them always for all new materials and high burnup levels?

Comment from the audience: It seems to be a too complex program for such an unlikely event like LOCA actually is.

Comment by J.R. Jones: I disagree with the EdF view, presented in the paper by M. Lambert et al. (see para 17) and repeated several times during the meeting, that fuel pellet axial relocation is a new issue that had not previously been addressed in a regulatory framework. In the UK we had been working on this since the late 1970s and this issue was explicitly addressed as part of the pre-construction licensing case for the Sizewell B reactor.

Comment by N. Waeckel: Since the fuel fragments relocation has been observed in several experiments, the relocation phenomenon, per say, is not a speculation. Nevertheless, its consequence, such as local enhanced cladding temperatures, has not yet been demonstrated, analytically or experimentally, and can still be considered as a speculation for two reasons: none of the experiments we know exhibited fuel relocation and showed local enhanced cladding temperature - none of the experiments gave information about the time of the fuel relocation. If it does not happen during the blowdown (i.e. before the high temperature oxidation phase) then it does not matter. It is plausible that some fragments may relocate during the quench phase when the rods are violently shaken by the coolant flow. This phenomena has been clearly shown on the movie recorded by JAERI during one of their integral LOCA tests. When fuel relocations occur during quenching phase, no adverse consequence is expected. The planned in pile Halden LOCA test on a single high burnup rod should confirm whether fuel relocation is a safety issue or not.

Question from the audience: Do you consider also small break LOCA tests in your programme?

Answer by A. Maillat: Yes we do.
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Aix-en-Provence, 22-23 March 2001

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