IRSTITUT DE RADIOPROTECTION ET DE SORETÉ NUCLÉAIRE

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Farouk Eltawila, Director Division of Systems Office of Nuclear Regulatory Research

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Direction de la prévention des accidents majeurs Subject: IRSN comments to RIL-801. Your Ref. Your letter dated July 14<sup>th</sup>, 2008. Our Ref.

Dear Dr E-Ettawila,

By your letter sent to me by electronic mail on July 14<sup>th</sup> 2008, you were requesting IRSN review and comments on two documents prepared by the NRC staff (Research Information Letter 801) and its contractor at ANL (NUREG/CR-6967).

These documents were to serve as a technical basis for potential revision of embrittlement criteria in. 10 CER 50.46 for postulated Loss-of-Coolant Accidents.

Your letter were also providing guestions that the public will be invited to address.

Enclosed you will find the IRSN review of the RIL-801 and IRSN position on the potential revision of the embrittlement criteria.

To summarized, IRSN considers; in view of the current state of knowledge, not reasonable to exempt the ballooned part of the core from satisfying conditions that would ensure clad integrity. IRSN thinks that more research work is needed to find an appropriate criteria and is ready to cooperate with the USNRC in this field.

Yours sincerely.

Michel SCHWARZ

Director, Major Accident Prevention Division

JUNSE BENEW Complife Memplale = ADM-013

F-RIDS=ADM-03 Que = P. Chifford (PMC3)

3P.3 13115 Saint-Paul-lez-Durance Cedex France

Mail to :

Fél. +33 (0)4:42:19:96:89 Fax +33 (0)4.42:19:91:57 michel.schwarz@irsn.fr

eadguarters 7, av. du Général-de-Gaulle 2140 Clamart witchboard +33 (0)1:58 35 88 88

S Nanterre 8 440 546 018



# Copies with enclosure:

IRSN

J. Repussard

D. Queniart M. Jorel

S. Cadet-Mercier

E. Scott de Martinville

A. Denys

JC. Micaelli F. Barré

C. Grandjean

## IRSN Comments to RIL-0801 and NUREG/CR-6967

## 1. General remarks

As a preliminary remark IRSN would like to point out that the **embrittlement** criteria is only one of the topics of the **acceptance** criteria. In this respect, IRSN believes that the amount of experimental data on high burnup fuel behavior in LOCA conditions, - already available or to be expected in the next coming years -, would justify to consider the revision of the whole ECCS acceptance criteria and related methodologies.

Enclosure 2 of the RIL discusses other fuel-related LOCA phenomena involving additional issues, the resolution of which being fully justified in a revision process. Among these issues, the coolability of core regions with high flow blockages resulting from clad ballooning is addressed by 10CFR50.46(b)(4). IRSN thinks that this issue should be revisited in light of the experimental data obtained after issuance of NUREG-0630 and with consideration of the axial relocation of fuel in clad balloons.

The axial relocation of fragmented fuel -with a potential for dispersal of fuel particles through burst opening -, although having no direct influence on embrittlement threshold, has been recognized for having possibly a significant impact on the accident transient, therefore on the LOCA analysis.

The revision of the acceptance criteria should also be closely related with the ongoing discussion on transition break size (TBS). Should break size regulation change in a near future, then significant impact might be expected on the conditions to be considered for LOCA analysis.

### 2. The key-questions

Focusing on the **embrittlement** criteria, the key-questions are:

- 1. Is the requirement of retaining (some) ductility in the cladding appropriate to represent the load bearing capability of the cladding under LOCA, without being overly restrictive ?
- 2. Can an embrittlement criteria ensure residual ductility over the whole rod length without excluding the balloon region ?

Due to the effect of secondary hydriding that leads to significant hydrogen absorption from the cladding inside in the burst region as a result of the dissociation of steam entering the burst opening, we know that the answer to Question 2) is NO. This is clearly recognized in page 4 of the RIL :" ... no criteria have been found that would ensure ductility in the cladding balloon " and recalled in page 5 of Enclosure 1: "The cladding does not retain ductility at these locations (of peak in hydrogen concentration), even though the oxidation in the balloon has been limited in accordance with the current regulation".

The related question is : How to handle the particular situation of no residual ductility in the ballooned region of the cladding ? The answer currently proposed in

the RIL appears somewhat uncertain and lacks technical basis: "However, loss of ductility in this **short portion** of a fuel rod should not lead to an uncoolable geometry as long as the amount of oxidation in the ballooned region remains limited in the current manner."

It must first be pointed out that this argument departs from the opinion of the AEC Regulatory Staff and of the Commission during the 1973 ECCS Hearing: "... we think it prudent to apply our criteria to all of the core and not to exempt any part." (see Appendix A for extended excerpts of opposing arguments).

Thirty five years later, might the original requirement of no clad fragmentation over the whole rod length be relaxed to allow an exemption over a **limited** part of the fuel rod cladding ? If a positive answer were to be envisaged this would certainly involve :

- strong requirements defining the extent of an "acceptable" fragmentation;
- a complementary evaluation of its possible impact on the core coolability and on radiological release;
- experimental data to validate the approach, in particular in the range of fuel burnups currently aimed at.

However, for the time being, on account of the expected high uncertainties remaining for a possible evolution towards an non-coolable situation, IRSN considers that the current state of knowledge is not sufficient to support such a position.

The NRC argument for an exemption of the ballooned region from the non-zero ductility criteria is based on the observed behavior of pressurized rod segments that were submitted to a simulated "integral" LOCA transient (OCL and ICL tests). Enclosure 1 (page 5) of the RIL indicates: "Although ductility in the ballooned region cannot be assured with reasonable limits, the ballooned region has **remained structurally intact** during these tests with limited oxidation." To some extent, this provides a partial answer to above Question 1), suggesting that the maintaining of the structural integrity could be a better requirement than the retention of ductility for the evaluation of the load bearing capability, insofar as it can address the ballooned region of the fuel rod. This is also recognized in the staff response to ACRS recommendation (1) following the ACRS meeting of Feb. 2, 2007, as reported in Enclosure 3 of the RIL: "The requirements would be aimed at ensuring that fuel maintains adequate structural integrity during a LOCA so that coolable geometry and long-term cooling capability are maintained...".

The maintaining of structural integrity can be addressed in a strength-based approach by testing the quench survivability under constraint conditions, as followed in the current Japanese approach ; prototypic bundle constraints should be best evaluated to define the constraint conditions.

The maintaining of structural integrity can also be addressed in a toughness-based approach using impact testing. Impact testing has already been widely used in the extensive test program conducted by Chung and Kassner at ANL in the late 70s.

Appendix B recalls recent opinions from NRC and EPRI showing that impact testing was still considered until recently to evaluate the cladding embrittlement while keeping consistent with the experimental methods used at ANL in the early 1980s

and that were relied upon in the 1988 technical evaluation of the current acceptance criteria.

However, no impact tests were performed in the ballooned region of the rods of integral tests in the current ANL test program.

The above Question 1 may therefore be considered as still pending, particularly with consideration of the negative answer to Question 2.

An alternative approach could be based on strength or toughness. However, a prior technical consensus should be obtained on the loading conditions being best representative of fuel rod loading conditions and on methods to demonstrate compliance. A key point is the selection of a testing method where the load application does not bias the location of the failure site and lets the sample fail at its weakest point.

In summary, in view of the current state of knowledge, IRSN would not support an embrittlement criteria that leaves under question the fragmentation of the cladding in the balloon region. In this respect, IRSN considers that further tests and analyses are needed to provide the appropriate experimental data required to complement the important ANL results reported in NUREG/CR-6967 and to establish a revised criteria. IRSN is currently examining alternatives to the ductility-based approach for a revised embrittlement criteria.

## 3. Small Breaks

In addition to the issues related to phenomena of *Clad ballooning/Flow blockage* and *Fuel relocation* discussed in Enclosure 2 of the RIL, important issues may be related to the impact of steam pressure on key phenomena such as cladding oxidation and secondary hydriding. These issues are still poorly informed, current experimental data being obtained from tests performed mostly under Large Break conditions (atmospheric steam pressure).

Thus, extending the criteria from the Large Break to Small Breaks conditions will require to extend the scientific basis, since :

- No high-pressure steam test was performed in the ANL program. Secondary hydriding under high-pressure steam conditions was never tested in the whole world, except on IE-019 rod of PBF/IE-5 test (irradiated cladding, fresh pellets).
- Very few hydrogen measurements after Zircaloy oxidation under high-pressure steam have been performed. No hydrogen measurement neither mechanical test has been performed after ZIRLO oxidation under high-pressure steam. No highpressure steam test was performed with M5. However, tests with E110 show very rapid oxidation and massive hydrogen uptake<sup>[1]</sup>.

<sup>1</sup> G. Hache, Oxidation of Zr Alloys in High Pressure Steam and some Results under Atmospheric Pressure, Nuclear Safety Research Conference, Washington (USA), 28-30 October 2002, NUREG/CP-0180, pp. 169-189

### 4. Answers to Questions in letter from F. Eltawila to M. Schwarz

In addition to the previous comments on the RIL-0801 and NUREG/CR-6967, we would like to give brief answers or comments to some of the questions listed in the letter from F. Eltawila to M. Schwarz :

I. Technical Basis

1. RIL 0801 Figure 1 provides the measured embrittlement threshold for all fresh and irradiated cladding specimens investigated during the ANL research program. Hydrogen dependent post-quench ductility regulatory criteria, similar to the lines on this figure, may be established from these experimental results. The linear fits in figure 1 of the RIL0801 are not in best accordance with the experimental results: due to the curved form of the embrittlement CP-ECR as a function of Hydrogen content (Figures 120 or 238 of NUREG/CR-6967), the linear fits will be overly conservative at > 600 wppm hydrogen and not conservative enough in the 300-500 wppm hydrogen range.

- a. Is the technical information presented within NUREG/CR-6967 sufficient in scope and depth to justify specific regulatory criteria applicable to all current zirconium cladding alloys? YES within the scope of a ductility-based approach which cannot address the balloon region of the fuel rod.
- b. Is the technical information presented within NUREG/CR-6967 sufficient in scope and depth to justify periodic testing on as-fabricated material? *YES*
- c. Is the technical information presented within NUREG/CR-6967 sufficient in scope and depth to address sensitivities to alloy composition, trace elements, manufacturing practices, fuel rod burnup, and transient temperature profile? *YES*
- 2. Section 2 of NUERG/CR-6967 details the experimental techniques and procedures employed at Argonne National Laboratory (ANL) to assess post-LOCA cladding properties.
  - a. Were the experimental techniques and procedures adequate for their intended purpose of defining acceptable fuel criteria (e.g., specimen preparation, specimen size, heating/cooling rates, ring-compression techniques, test temperature, acceptance criteria for post-quench ductility and breakaway oxidation, etc.)?

There is currently no clear consensus with other labs on the Ductile-to-Brittle (DTB) transition based on Offset Strain. Due to the shape of the Offset Strain curve as a function of CP-ECR (or H content) near the DTB, this could possibly induce significant variations in results obtained in different labs.

*The impact of heating rate - particularly important for short duration tests - has not been clearly evaluated.* 

No breakaway test was performed with high burnup nor with hydrided cladding.

- b. Is the technical information presented within NUREG/CR-6967 sufficient in scope and depth to address uncertainties related to and repeatability of measured results? *Uncertainty appears sufficiently limited from the observed scatter around the trend curves.*
- II. Performance-Based Testing Requirements
  - 1. Due to potential sensitivities to manufacturing processes, performance based testing may be required to characterize the LOCA performance of new cladding alloys.
    - a. Section 2.1 of NUREG/CR-6967 details all of the fresh and irradiated cladding specimens investigated during the ANL research program. Is the extent of the ANL material database sufficient to justify the applicability of experimental results to future cladding alloys?
      - Yes within the range of the current alloys composition.

- b. Conducting testing on irradiated specimens is more difficult and expensive than similar tests performed on unirradiated specimens. Does a sufficient technical basis exist to justify testing on hydrogen charged, unirradiated cladding specimens as a surrogate for irradiated fuel cladding? *YES, except for secondary hydriding occurring in integral testing.*
- 3. Due to potential sensitivities to manufacturing processes, routine testing may be required to verify material performance. Discuss the degree of difficulty for performing such testing along with benefits and limitations.

If the routine testing should be performed by manufacturers, a difficulty may arise in the need to freeze the testing procedures for use in different places and for extended periods of time.

#### III. Implementation

- 1. Implementing new regulatory criteria for 10 CFR 50.46(b) may necessitate further testing and new licensing activities (e.g., revised methods, updated safety analyses, etc.).
  - a. Discuss the cost-benefit for implementing new regulatory requirements similar to those discussed in RIL 0801.
  - b. Discuss the relationship between implementing planned changes to 10 CFR 50.46(a) (i.e., transition break size) and implementing new performance-based criteria in 10 CFR 50.46(b).

This relationship need to be clarified in relation with the on-going discussion on Transition Break Size.

- 2. Implementing hydrogen-based regulatory criteria may require the development of high confidence corrosion and hydrogen pickup models.
  - a. What type of information is needed to develop such models and is such information readily available?

Essentially hydrogen content as a function of burnup, available from measurements on irradiated fuel rods extracted from operating reactors.

b. What sort of performance indicators (e.g., pool-side measurements, hot cell examinations, etc.) could be used to validate models? *Not investigated by IRSN.* 

- 3. Crud deposits on the fuel cladding surface may affect fuel stored energy, fuel rod heat transfer, and cladding corrosion. *Not investigated by IRSN* 
  - a. What sort of role does plant chemistry and crud deposits play on these items?
  - b. How should normal and abnormal levels of crud deposits be addressed from a regulatory perspective?

# Appendix A

During the 1973 ECCS Rule-Making Hearing, the AEC Regulatory Staff wrote in its Concluding Statement [A1]:

"Westinghouse has suggested that because clad bulges have survived mechanical tests (Exhibit 1078, Appendix D; Exhibit 1151, Section 13), the bulges can be effectively ignored... The staff is not aware of any experimental information to support the conclusion that the bulges are more resistant to fragmentation. However, we suggest that... experiments of deformed and undeformed cladding could be performed to answer this question.

It has been pointed out that the bulged regions constitute a small fraction of the core and, therefore, need not be considered in LOCA analysis (Westinghouse, GE and B&W Concluding Statements). It is true that clad fragmentation of ONE local ruptured region of ONE pin would not impair the coolability of the core. However, the staff is not aware of any experimental information which would aid in quantitatively assessing the effect on core coolability of numerous ruptured regions with associated fragmentation. If the hot spot with its associated bulge were to be ignored along with other ruptured fuel elements, it is not clear that these other bulged regions would not fragment if the hot spot fragmented ; nor is it known to what extent fragmentation of a widespread rupture distribution would impair ECCS effectiveness. Therefore, until realistic quantitative assessments can be made, the staff believes that swelling and rupture should be treated in LOCA analysis as specified in Section II.H so as to preclude fragmentation of the core hot spot."

In December 1973, the Commission expressed the following opinion [A2]:

"All of the reactor manufacturers except Combustion Engineering objected strongly to the application of oxidation and maximum temperature criteria to the hottest spot of the cladding, especially to regions that are calculated to have swollen and burst open. It was argued that this represents only an extremely small fraction of the reactor core and that even if this small amount became fractured it would no harm. Another contention is that the criteria are so conservative that even if these hot spots were oxidized more than the criteria allow they would remain intact...

(These arguments) must be recognized as opinions as to what happen in a situation that has never yet occurred. Others are not so sure that a local failure would not be propagated more widely throughout the core... In view of the lack of experience in this hypothetical situation, we think it prudent to apply our criteria to all of the core and not to exempt any part".

### References

- A1. USAEC, Atomic Energy Commission Rule-Making Hearing, Concluding Statement of the Regulatory Staff, Docket RM-50-1, April 16<sup>th</sup>, 1973.
- A2. USAEC, Opinion of the Commission in the Matter of the Rulemaking Hearing on Acceptance Criteria for Emergency Core Cooling Systems for Light-Water-Cooled Nuclear Power Reactors, CLI-73-39, December 28, 1973.

# Appendix B

In Attachment 2 of the Research Information Letter 0202 [B1], NRC wrote :

"It was known that resistance to fragmentation was the underlying objective of the Commissioners in 1973, and resistance to fragmentation can also be characterized by toughness. Therefore, a toughness test was devised that consisted of a *pendulum impactor* that would strike the fuel rod in the ruptured region, precisely *where the hydrogen absorption* was worst...

Rods that survived the quenching with little margin were fragile and would fracture with a low *impact energy* of about 0.03 J at room temperature. A factor of 10 in *impact energy* was chosen as a measure of robustness, and rods with a measured total 17% oxidation or less were found to survive the 0.3 J impact at room temperature, provided the oxidation did not take place much above 2300°F. There was a de facto acceptance of these tests in 1981 as confirmation that the \$50.46 embrittlement criteria were conservative.

In a brief write-up dated April 8, 2002, Ralph Meyer added [B2]:

Cladding alloys that are different from Zircaloy may have different toughness, and the effects of burnup and corrosion might also affect toughness. The *impact test* can take all of these effects into account, and the result would be an oxidation limit that would ensure a significant margin of resistance to fragmentation beyond that needed to survive quenching, consistent with the opinion of the Commission at the conclusion of the 1973 ECCS hearing."

In a letter to Ashock C. Thadani dated September 9, 2003, EPRI argued that [B3]:

"...It is of utmost importance that the Argonne test program provide the same technical data as used in the mid-1980s evaluations to confirm the applicability of the cladding embrittlement criteria established in 1988 to high-burnup fuel... It is not evident that the program can accomplish this goal, because the current Argonne test matrix focuses heavily on thermal shock and ring compression tests and *contains no impact testing* 

...It is not clear how the ring compression tests would confirm the safety margin in the current criteria for post-LOCA conditions since this safety margin **was established using** *impact tests* during the 1980s assessment. To confirm the current criteria for post LOCA conditions after reflood, the impact tests are more applicable. The program test plan *should be revised to include impact tests* similar to those relied upon in the 1988 technical evaluation to support the current criteria for post-LOCA conditions."

It is also worth mentioning the opinion of R.E. Williford in a paper published in Nuclear Technology in September 1986 [B4]:

"The handling failure characteristics of oxygen-embrittled Zircaloy have frequently been based on the results of ring compression tests. However, significant differences in failure loads can be caused by sample length and by the experimental methods employed...Impact may be a better representation of maximum loads to be anticipated during rod removal by overload crane or accidental rod drop during removal, transport and storage."

#### References

- B1. A. C. Thadani, Research Information Letter 0202, Revision of 10 CFR 50.46 and Appendix K, June 20<sup>th</sup>, 2002.
- B2. R.O. Meyer, Toughness vs ductility, e-mail to V. Asmolov et al., April 8<sup>th</sup>, 2002.
- B3. D.J. Modeen, letter to A.C. Thadani, September 9<sup>th</sup>, 2003.
- B4. R.E. Williford, Safety Margins in Zircaloy Oxidation and Embrittlement Criteria for Emergency Core Cooling System Acceptance, Nuclear Technology, 74, , pp. 333-345, September 198