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Argonne NATIONAL LABORATORY

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OFFICE OF SECRETARY
RULEMAKINGS AND
ADJUDICATIONS STAFF

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October 5, 2009

Secretary
U.S. Nuclear Regulatory Commission
Washington, DC 20555-0001

ATTN: Rulemakings and Adjudications Staff

Subject: Comments on Proposed Rulemaking on 10 CFR 50.46 (ECCS Acceptance Criteria) [RIN 3150-AH42]

Dear Secretary:

Argonne National Laboratory, with funding from NRC's Office of Nuclear Regulatory Research (RES), provided most of the data that form the basis for this proposed rule change. The rule change itself, however, is being formulated by NRC staff and Argonne has been involved only indirectly in this process. At this stage of the rulemaking process, the principal investigators at Argonne might be able to provide some perspective that could be helpful in moving forward. We have discussed this with our sponsor and they have encouraged us to send our comments directly to the rulemaking docket. The comments below are numbered according to the questions in Section IV of the Advance Notice of Proposed Rulemaking (Federal Register Vol. 74, No. 155, August 13, 2009).

#1. Objective 1 contemplates a rule covering all fuel cladding material without regard to its composition. It should be emphasized that the data base for zirconium-based cladding is extensive and the recent data cited in the ANPR build on a massive foundation. For example, a review paper by Hache & Chung (NSRC 2000) cites 37 significant published papers and reports on work performed before the recent research was begun. No such data base is cited for the contemplated general cladding material and it is likely that none exists. Such a rule would permit a relatively small proprietary data base to be developed by a manufacturer, and such data could bypass the peer-review process and hearing exposure that now assure the quality of the zirconium-alloy data base. Argonne is concerned that the contemplated generalization could allow future cladding criteria to circumvent open scientific scrutiny.

#3. The NRC request for vendor-calculated maximum time spans with cladding surface temperature above 1200°F (649°C) for the full range of piping break sizes and NSSS/ECCS design considerations may not lead to useful information that can be incorporated into a criterion that would preclude embrittlement due to breakaway oxidation. Argonne worked closely with NRC-RES, the Electric Power Research Institute, major nuclear vendors, and international research and regulation experts during the breakaway oxidation testing. In particular, we were seeking guidance on a maximum test time that would be LOCA-relevant. As we could not get a consensus on this, we set the maximum test time at 5000 s based on the feedback we did receive. It appears to us that of the two approaches being considered by NRR, the one specifying a time period for which breakaway oxidation did not occur would be easier to assess and regulate. Furthermore, the search for the minimum breakaway time and corresponding isothermal

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temperature is a very tedious experimental process requiring an extensive data base. Argonne performed such an investigation for one cladding material and repeated the study three times using different cladding heats and new test trains. Based on our experience with this cladding material, the minimum breakaway time occurred over a very narrow temperature range (970-985°C) with a corresponding large scatter in hydrogen pickup. However, the results at 1000°C were much more consistent and reproducible. In addition to a specified test time, NRC may want to consider also specifying a test temperature (e.g., 1000°C).

Argonne observed serious breakaway oxidation in a relatively short period of time (about 500 seconds) for only one cladding material, an older variety of a Zr-1%Nb alloy (E110) used only in Russian-designed VVER reactors. However, in studying that material, it became clear that materials used in the U.S. could be degraded by changes in manufacturing processes that by themselves would not appear deleterious. Enough testing has been done to know how to detect unacceptable breakaway oxidation, but a thorough study covering numerous possible temperature histories during a LOCA has not been performed. Therefore, it appears to us that a simple screening test for a specified oxidation time and temperature would be more compatible with the current level of understanding of breakaway oxidation.

#4. As proposed under Objective 2, the rule would require the retention of cladding ductility, defined as ≥1.00 percent permanent strain, and the applicant would interpret strain data to obtain an oxidation limit (CP-ECR) that could then be used in a LOCA analysis. The "1.00 percent" should be changed to "1.0 percent" due to measurement uncertainty. The second decimal place would require measurement accuracy down to the micron level.

As Argonne used a large data base for five cladding materials (including three cladding alloys) and expert judgment subjected to independent peer review to derive the ≥1.0% permanent strain criterion, Argonne endorses this ductility criterion. However, it should be noted that: a) permanent strain cannot be measured accurately for rings that fail with more than one crack or with a single wide crack; b) it is highly unlikely that a ring will fail precisely at 1.0%; and c) the shape of the permanent strain vs. oxidation level (CP-ECR) curve is required to determine the ductile-to-brittle transition oxidation level by means of interpolation. While Argonne has no reservations regarding the ≥1.0% permanent strain criterion, it does have reservations regarding the skill and experience of other organizations in sample preparation (oxidation and quench), in conducting ring compression tests properly, in stopping tests quickly after the first significant load drop, and in data assessment and interpretation. Argonne did write a procedure for conducting oxidation-and-quench tests, for conducting ring-compression tests, and for data assessment and interpretation [ML090900841]. Included in this document is the relationship between offset strain, which can be determined for every ring tested, and permanent strain, which can only be determined accurately for rings that fail with a single, tight, through-wall crack. However, the issue of *ensuring* data quality and proper data interpretation by means of independent peer review has not yet been adequately addressed.

We understand that there is a need to create a more performance-based rule than the previous one. The use of a sliding scale for the oxidation limit already provides this improvement. For example, a cladding material such as M5, which absorbs very little hydrogen during its burnup lifetime, would have a much higher oxidation limit than, say, Zircaloy-4, which absorbs a lot of hydrogen. Based on trends deduced from Argonne data, the sliding scale of oxidation limit vs. hydrogen content should apply to new Zr-based cladding alloys as well as to the specific Zircaloy-4 (Zr-1.3%Sn), ZIRLO (Zr-1%Sn-1%Nb) and M5 (Zr-1%Nb) cladding materials tested. The Argonne data allow for direct comparison between Zry-4 and ZIRLO oxidation limits at comparable hydrogen contents. The data for high-burnup M5 appear to be consistent with the Zry-4 and ZIRLO trend data. However, additional data for pre-hydrided M5 and pre-hydrided Zircaloy-4 with 100-200 wppm hydrogen would increase the confidence level of using the sliding scale for new cladding alloys.

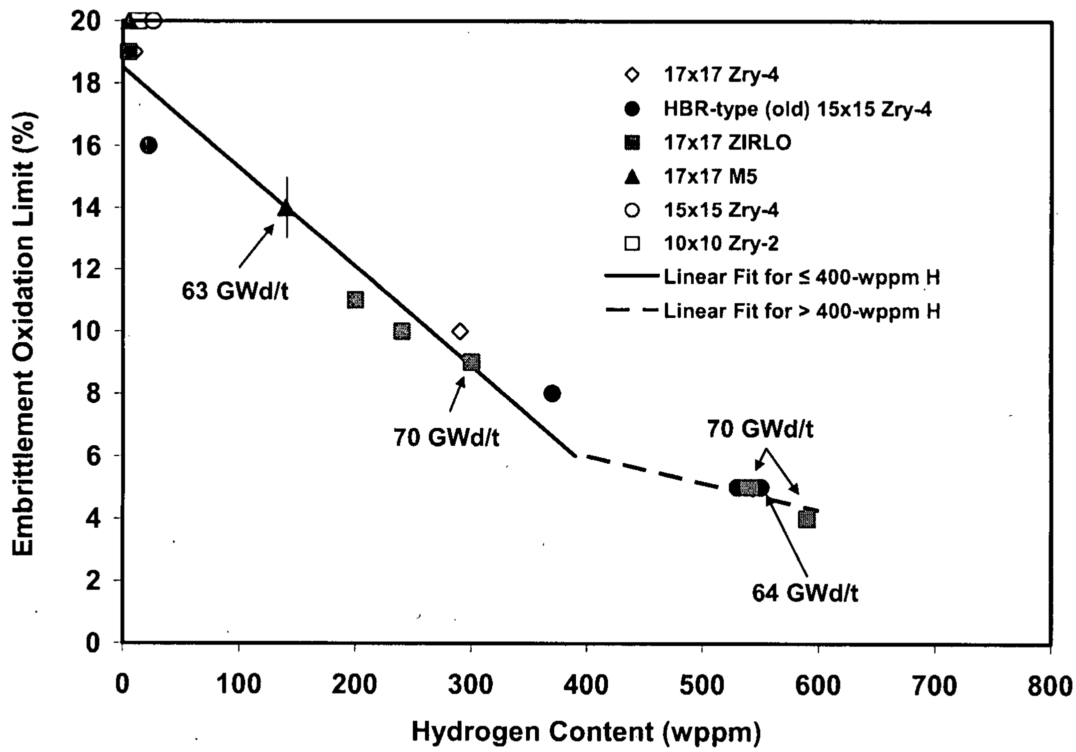
The primary circumstance that is likely to produce a significantly different sliding-scale oxidation limit would occur if an applicant proposes to limit peak cladding temperature to a value lower than 2200°F. In this case, we support using a criterion of 1.0% permanent strain and other procedures that could be specified in a Regulatory Guide. However, revised limits produced in such a case would be less susceptible to error if the applicant's procedures were shown to produce a limit at 2200°F that was not higher than the limit that is fixed in the rule and has had the benefit of open scientific peer review.

Paragraph b in Question 4 is not entirely accurate. In an April 17, 2009 report [ML091200702], Argonne used a refined method to convert raw data to pre-test hydrogen content in the metal for all high-burnup cladding materials used to determine post-quench ductility. This report was not mentioned in the ANPR and the corrected values were not used in Fig. A of the ANPR. We recommend that this detailed report be added to the list of "Available Supporting Documents." As the earlier summary report [ML090690711, March 30, 2009] contains hydrogen content values not fully corrected for the additional mass in the corrosion and fuel-cladding-bond layers, that report should be disregarded. Included in the April 17th report are high post-quench-ductility values (>43% offset strain) for high-burnup samples with about 600-wppm hydrogen after 4% oxidation level (CP-ECR). However, for high hydrogen content, the corresponding low CP-ECR values resulting in ductile-to-brittle transition are quite sensitive to heating rate and peak oxidation temperature, which must be kept in mind when interpreting the results. Argonne used LOCA-relevant heating rates for these tests. Low CP-ECR values were reached before the specimens reached 1200°C. This situation further underscores the desirability of data interpretation by the data taker coupled with an independent peer review.

With regard to Paragraph c of Question 4, Argonne would like to comment on the issue of data scatter from ring-compression testing. The so-called data "scatter" is much less than the scatter in hydrogen pickup along high-burnup fuel rods. Relatively little data scatter was observed for post-quench ductility of as-fabricated cladding and cladding with uniform hydrogen concentration. For pre-hydrided and high-burnup rings with large azimuthal variations in hydrogen content (e.g., 450 to 730 wppm), measured offset and permanent strains showed expected variation dependent on the orientation of the maximum hydrogen content relative to the locations of maximum stress. For high-burnup rings that were subjected to local hydrogen analysis following ring compression testing, through-wall cracks were observed to occur at locations of maximum hydrogen content. Thus, variations in ring-compression results were more systematic than random. However, large azimuthal and axial variations in hydrogen content were seen repeatedly in specimens taken from real fuel rods with average hydrogen content >400 wppm. Therefore, some statistical treatment of a large data base is warranted for a manufacturer's data in the development of a hydrogen uptake model. With regard to ring compression tests, five data points at each oxidation level would be sufficient to characterize the embrittlement threshold for cladding with large azimuthal variation in hydrogen content.

ANL has reservations regarding the use of Figure A in Appendix A. This figure shows raw data for offset strain, which were taken from earlier reports [ML082130389 and ML090690711]. Although appealing in that the figure indicates that a large data base was generated, the figure shows offset strain values even though the ductility criterion (≥1.0%) proposed by NRC is in terms of permanent strain. Without the relationship given in ML090900841 for offset vs.

permanent strain, it is not clear how the reader would understand the significance of the raw data. Also, Figure A invites an NRC reviewer or a licensee to use their judgment to determine the embrittlement threshold. These judgments have already been made by Argonne researchers who performed the tests and are familiar with each individual data point. The judgments have also been subjected to international peer review. The risk of using a figure such as Figure A is already apparent from the inclusion of several data points (particularly the low CP-ECR values near zero hydrogen concentration) that should not influence the result because of their low quality. The high-quality data set that should be used for "HBR-type (old) 15×15 Zry-4" is summarized on page 46 of ML082130389 and presented in detail in ML090900841. The appropriate figure to use in this instance is shown below. The figure contains two new data points for pre-hydrided ZIRLO with 200- and 240-wppm hydrogen. One high-burnup data point for ZIRLO has been added to the figure based on data presented in the April 17th report (Table 4 in ML091200702): 4% CP-ECR at 592-wppm hydrogen and 1132°C peak cladding temperature. Based on results at 5.1% CP-ECR (448 wppm, 1162°C, ductile) and 6.3% (540 wppm, 1176°C, brittle), it is clear that high-burnup ZIRLO with ≈600-wppm hydrogen would be brittle at 5% CP-ECR and ≈1160°C. Thus, the transition CP-ECR for which ductility is maintained is 4%.



Argonne welcomes an independent peer review of this figure and its data interpretation and would support any modification of this figure that results from such a review process. In particular the lines drawn to represent the best linear fits to the high-burnup data at ≤400-wppm hydrogen and >400-wppm hydrogen could be revised or eliminated from this figure.

#6. Section I of the ANPR (Background) says that research results revealed that alloy composition has a minor effect on embrittlement. Within the ductile range (≥1.0% permanent strain), Argonne observed more variability is strain values for the same cladding alloy (e.g., Zircaloy-4) fabricated using different manufacturing methods than observed for different alloys with similar manufacturing histories. However, no significant variation was found in the ductile-to-brittle transition oxidation level for four modern cladding materials with three different alloy compositions. Argonne believes that the high-temperature embrittlement of new, as-fabricated, dilute Zr-based cladding alloys would occur within the narrow range of 18-20% CP-ECR if proper testing methods and data assessment were used. As the determination of the embrittlement threshold for as-fabricated cladding alloys is relatively straightforward, Argonne is in favor of giving vendors the option of generating such data. To the extent that such data may fall below 18-20% CP-ECR, it is more likely that such variation would be due to test conditions (e.g., very rapid heating rate) and data interpretation (e.g., reliance on offset strain) than to alloy composition.

Paragraph c under Testing Considerations describes the possibility of using pre-hydrided fresh cladding as a surrogate for irradiated cladding in tests for post-quench ductility. In Paragraph c and in the appendices, it is implied or stated that the NRC has not yet accepted the use of this surrogate material. Although additional demonstration of the surrogate's adequacy is desirable, recent ANL data described herein (see figure on page 4) further support the validity of this surrogate.

#7. Current research has shown that the susceptibility to breakaway is affected by alloy ingot preparation and cladding surface conditions. Although there is some understanding of these particular effects, no one knows whether other process changes might also affect breakaway. Vendors occasionally make changes in ingot suppliers and cladding fabricators, which may impact breakaway oxidation behavior. Changes in surface finish processes may also affect breakaway oxidation behavior. Because zirconium alloy preparation and tubing fabrication remain somewhat of an art, Argonne does not expect that all process variables that might affect breakaway could be identified *a priori*. As such, periodic breakaway-oxidation screening tests are recommended.

Sincerely,

Michael C. Billone

Michael C. Billone, Principal Investigator, NRC-sponsored LOCA Program

Ralph Meyer

Ralph O. Meyer, Nuclear Engineer STA

Rulemaking Comments

From:	Billone, Michael C. [billone@anl.gov]
Sent:	Monday, October 05, 2009 6:42 PM
To:	Rulemaking Comments
Cc:	Meyer, Ralph; Scott, Harold; Flanagan, Michelle; Voglewede, John
Subject:	ANL Comments on ANPR (RIN 3150-AH42)
Attachments:	ANL Comments on LOCA ANPR.pdf

Enclosed are comments from Argonne National Laboratory (ANL) on the ANPR (RIN 3150-AH42).

The comments are written in letter format addressed to Secretary, U.S. Nuclear Regulatory Commission, Washington, DC 20555-0001, ATTN: Rulemakings and Adjudications Staff.
We also intend to send a paper copy by postal mail to this address.

However, the enclosed PDF version will be much easier to incorporate as an electronic file.

Please accept the enclosed comments.

Thank you,

Michael. C. Billone

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From: "Billone, Michael C." <billone@anl.gov>
To: "Rulemaking.Comments@nrc.gov" <Rulemaking.Comments@nrc.gov>
CC: "Meyer, Ralph" <rmeyer@anl.gov>, "'Scott, Harold'" <Harold.Scott@nrc.gov>,
"'Flanagan, Michelle'" <Michelle.Flanagan@nrc.gov>, "'Voglewede, John'"
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Date: Mon, 5 Oct 2009 17:42:22 -0500
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