ANPR 50
(74FR40765)

2

October 5, 2009

TO: Rulemaking.Comments@nrc.gov

FROM: Ralph Meyer, USNRC (retired)
rmeyer@anl.gov

SUBJECT: Advance Notice of Proposed Rulemaking on 10 CFR 50.46 (ECCS Acceptance Criteria) [RIN 3150-AH42]

While still working for the NRC, I prepared responses to some comments that were received in reply to a notice in the Federal Register on July 31, 2008. That notice requested stakeholder comments on the adequacy of the technical basis and other issues related to potential new requirements in 10 CRF 50.46. The responses I prepared, with help from Michael Billone of Argonne National Laboratory, were circulated within NRC. However, the recent Advance Notice of Proposed Rulemaking makes reference to those stakeholder comments and places them on this docket. Therefore, it seems appropriate for our responses to be available also on this docket.

To summarize, some of the comments received from stakeholders were critical of the technical basis or suggested that rulemaking should not proceed. Some of those comments may not have merit, as can be seen from the following responses. The responses are provided below in Appendix A in the form of frequently asked questions and their answers.

Ralph Meyer

Ralph Meyer

DOCKETED
USNRC

October 5, 2009 (4:15pm)

OFFICE OF SECRETARY
RULEMAKINGS AND
ADJUDICATIONS STAFF

Template=SECY-067

DS10

Appendix A

Response to Frequently Asked Questions

Q1. Is there a safety issue for plants with approved LOCA analyses?

A1. There may be adequate margin in LOCA analysis to accommodate burnup effects, but plant-specific analysis may be needed to confirm this. Keep in mind that cladding oxidation must be limited in order to avoid cladding embrittlement and partial core collapse in the event of a LOCA. The 2003 fuel cleaning accident at the Hungarian Paks-2 power plant is an example of fuel assembly collapse under oxidizing conditions at relatively low temperatures (Fig. 1). Although these oxidizing conditions were extreme (approximately 7 hours at temperatures up to 1300°C), that accident shows the importance of an oxidation limit.

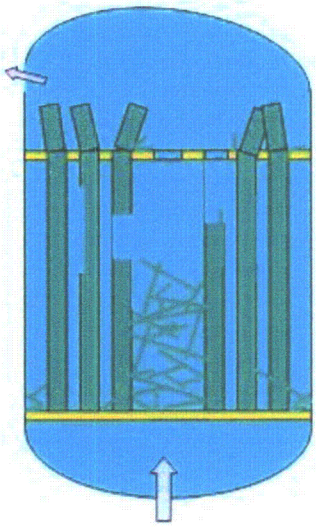


Fig. 1. Collapse of fuel with severely oxidized cladding in low-flow incident in a cleaning tank at Paks-2 plant in Hungary

Remaining below a ductility-based oxidation limit will preclude this kind of behavior, but the margin above the limit is not known. In the past, NRC's regulations have not identified a deviation in the oxidation level that would be of concern, but they did identify a change or error in cladding temperature of 50°F as significant and therefore reportable; see 10 CFR 50.46(a)(3)(i). Using the standard Cathcart-Pawel oxidation equations, it is easy to show that a change of 50°F is equivalent to approximately 2% oxidation (i.e., 2% CP-ECR) in the temperature range of interest. This 2% CP-ECR value can thus be used to test the significance of the new oxidation limits that have been found in NRC's research program.

An example that shows the significance of the new limits comes from tests on high-burnup ZIRLO-clad fuel from the North Anna plant (see Fig. 2). The specimen tested had a corrosion (i.e., pre-test oxidation) thickness of 40 microns and a hydrogen content of 550 wppm, well within the industry limits of 100 microns and 800 wppm, respectively. When tested with realistic, yet bounding, heat-up and cool-down rates, this material

exhibited a transition from ductile to brittle behavior at 5% CP-ECR. If this fuel had been analyzed according to NRC's latest requirements and recommendations, the oxidation limit would have been 13% (i.e., 17% from 10 CFR 50.46(b) then reduced to account for the corrosion thickness according to IN-98-29). The difference between 13% and 5% is large compared with the 2% significance test.

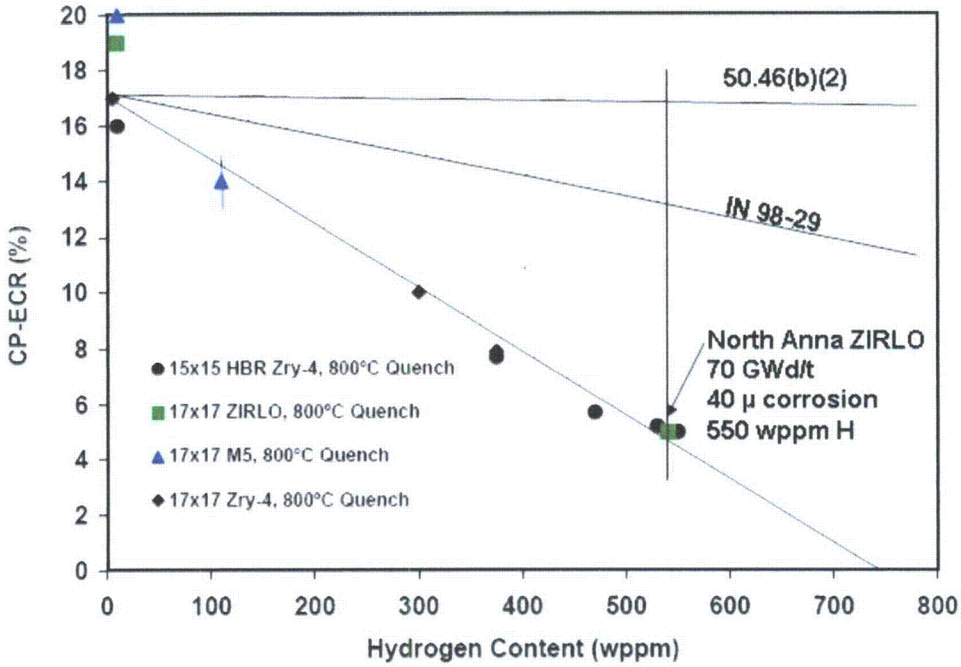


Fig. 2. Embrittlement threshold for North Anna ZIRLO-clad fuel rod and other relevant data compared with current licensing criteria.

However, there could have been mitigating features in the operating cycle and core design at the North Anna plant at the time this fuel was irradiated. For example, some postulated LOCA sequences might have had slower heat-up and cool-down rates than the bounding rates tested. This could increase the oxidation limit from 5% to about 9% in a sequence without quench based on other tests that were done. Further, if the fuel in question never exceeded LOCA cladding temperatures of about 1000°C, this might also increase the oxidation limit by several percent CP-ECR. On the other hand, many operating cycles are designed to maintain high power levels throughout at least the 2nd cycle by using burnable poisons and special core designs. Thus it is not certain that all fuel with significant corrosion and high hydrogen content would have low LOCA temperatures.

The proposed modification to 10 CFR 50.46(b) gives a hydrogen-dependent oxidation limit that is realistic, yet bounding. The proposed rule would also permit a licensee to provide additional data to justify a less-bounding limit for core designs and accident sequences when appropriate. Because of the large variation in accident sequences, core designs, and cladding performance under LOCA conditions, it may not be possible to assess the safety significance of the new criteria without plant-by-plant analyses. From the results discussed above, it is seen that the changes needed in the oxidation limit are significant and warrant attention.

Q2. Is a better understanding of balloon embrittlement needed before rulemaking?

A2. No, but the reason for this answer is not obvious.

The concern about balloon embrittlement involves a perceived problem with the special procedures for analyzing balloons in 50.46(b)(2) – a problem that might exist because of steam ingress through the burst opening. The perception is that the balloons will always be brittle such that the special procedures are not effective and should be replaced with something yet to be developed through future research.

By way of background, it needs to be pointed out that the original rule, which was derived to ensure ductility, never ensured ductility throughout the balloon. This can be seen clearly from Fig. 3. Because of wall thinning near the burst opening, a significant region of cladding would have a thickness less than average and therefore be brittle, assuming that the balloon was at the limiting oxidation value. The brittle region is large and is shown schematically in Fig. 4.

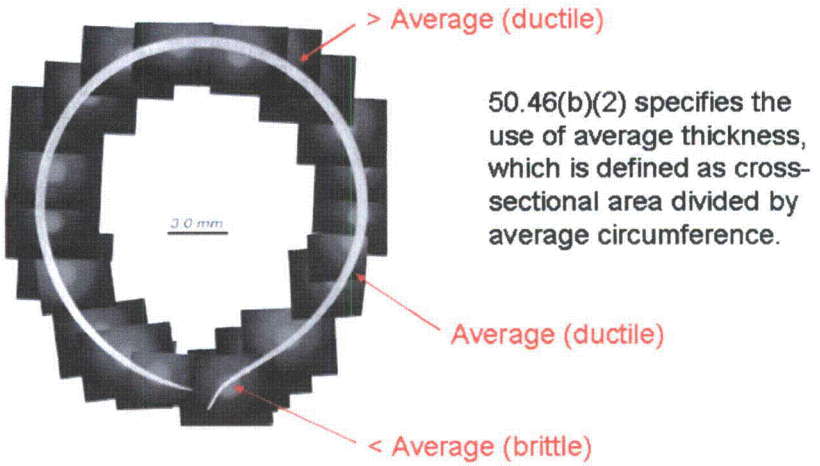


Fig. 3. Wall thinning near the burst opening in unirradiated Zircaloy-2 cladding exposed to simulated LOCA conditions (ANL test OCL#8).

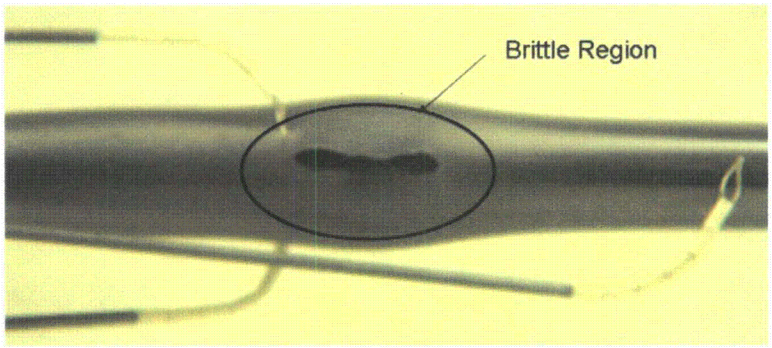


Fig. 4. Schematic of brittle region of the same sample shown in Fig. 3, before sectioning for microscopy.

A question arises because of enhanced hydrogen absorption that was discovered long after the original rule was established in 1973. Two bands of high hydrogen absorption were discovered in balloons in 1980 and confirmed in the current research. An example of these bands is shown in Fig. 5, where it is also seen that the hydrogen concentration at the burst midplane is low.

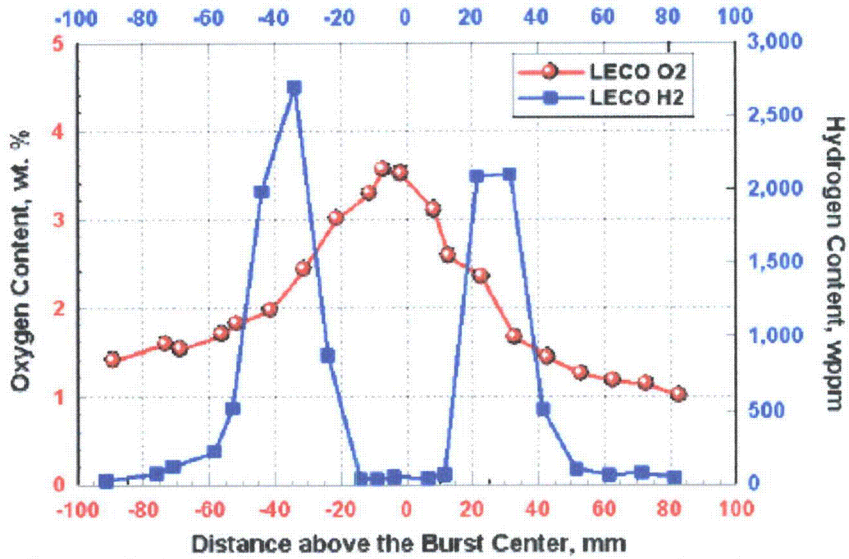


Fig. 5. Hydrogen and oxygen distribution in ballooned region of unirradiated Zircaloy-2 specimen after simulated LOCA test.

Although it has been widely thought that these bands of high hydrogen would cause ductile balloons to become brittle, the balloons were not entirely ductile as just seen and the hydrogen bands may not determine the limiting condition for the balloon (also referred to as the rupture node). Preliminary mechanical tests with as-fabricated rods indicate that the limiting condition is at the burst midplane, where oxygen concentration is high but hydrogen concentration is low (Fig. 6). Thus, the original concept of limiting oxidation to prevent embrittlement in the balloon (i.e., without hydrogen bands) appears to be appropriate for fresh and perhaps medium-burnup fuel rods.

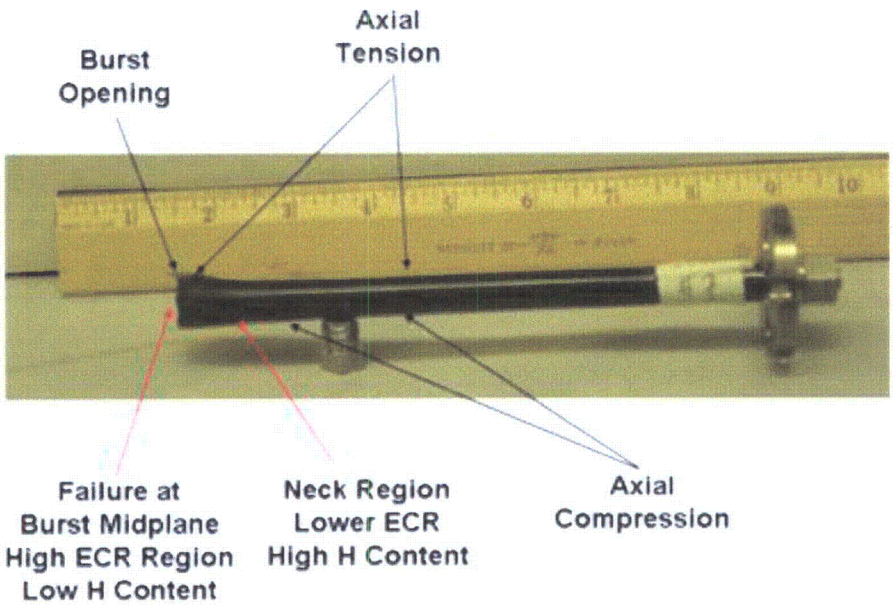


Fig. 6. Fracture of as-fabricated cladding after ballooning and rupture.

High-burnup fuel may behave differently, and this will be determined by testing that is planned for the next two years. For these tests, the hydrogen content from in-reactor corrosion will be present throughout the burst region. However, the midplane location may or may not pick up significant additional hydrogen during a LOCA transient, and preliminary indications are mixed.

If no significant hydrogen absorption occurs at the rupture midplane during the transient, the result will be similar to that of the unirradiated tests. That is, the pre-transient hydrogen will reduce ductility throughout the balloon, but the two hydrogen bands from the transient will have no net effect. No additional consideration would be needed for the balloon node and the original concept of limiting oxidation to prevent embrittlement in the balloon would be appropriate.

If, on the other hand, the hydrogen concentration at the rupture midplane increases during the test, then ductility at that location would be further reduced. Clearly, the oxidation limit for the balloon should not be increased relative to the case where hydrogen absorption during the transient had no effect. Additional consideration of the balloon would be needed in this case, and further testing would hopefully show that the fracture behavior of the balloon could be tolerated as long as the oxidation remained limited. Thus the oxidation limit as currently proposed should be retained and will probably not be altered (although perhaps qualified) by future test results.

Q3. Is 1200°C too high for LOCA embrittlement tests?

A3. Yes, for certain cases, but there are ways to avoid undue penalty, and results at 1200°C are necessary for many important cases.

Most of the testing at ANL has been done with cladding oxidized at a hold temperature of 1200°C following a ramp of 1-2°C/s from 1000°C, and those results were used to obtain

the proposed oxidation limit. Had these samples been oxidized at a lower temperature such as 1100°C, the embrittlement threshold would have been higher because the oxygen solubility in the beta phase is lower at the lower temperature. This effect is illustrated in Fig. 7. The concern is that many fuel rods with the highest hydrogen content, and therefore the lowest oxidation limit, would not be able to reach the ~1200°C temperature limit and therefore would be unduly penalized by using criteria based on this higher temperature. It is argued that such fuel rods would not be able to reach 1200°C because they have high burnup and therefore lower fissile content and lower power levels.

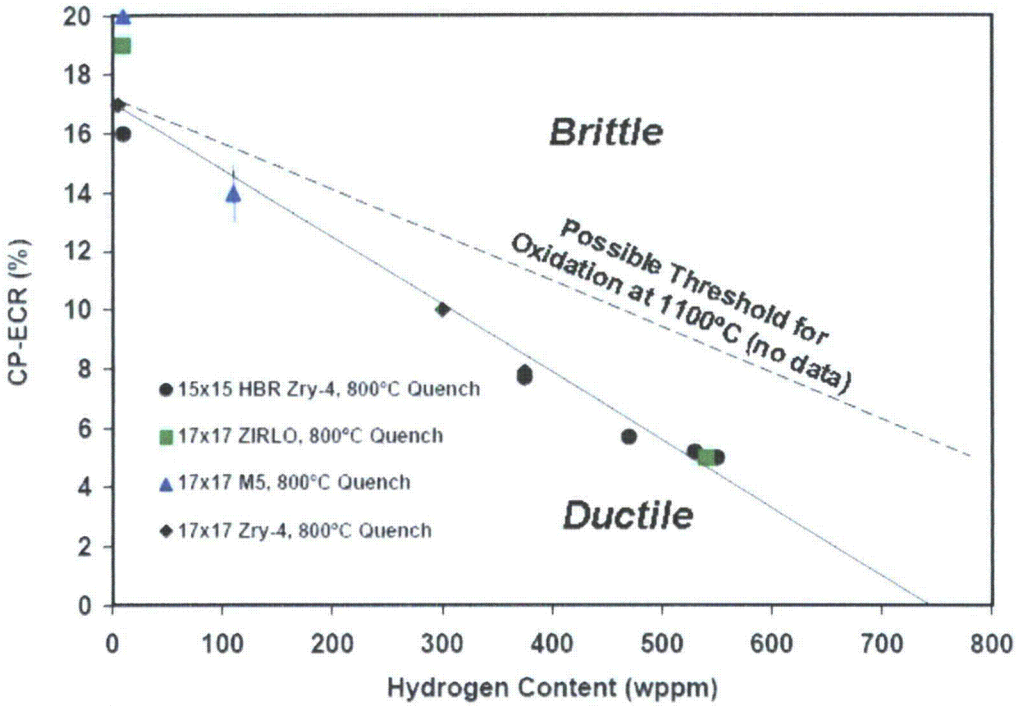


Fig. 7. Projected increase in embrittlement threshold for oxidation at a temperature lower than 1200°C at which the current samples were oxidized.

For a core loading such as described by Fig. 8, all the 3rd cycle fuel would be at very low power levels and therefore would remain well below 1200°C during a LOCA. For these cases, it is possible that a vendor could perform a generic analysis to show that such fuel did not experience rupture. With such results, low power fuel could be removed from further consideration. The proposed rule as currently envisioned would also permit a vendor to do further testing (e.g., oxidation at 1000 or 1100°C using prehydrided unirradiated specimens) to justify a higher limit for fuel with lower peak cladding temperatures. Using one of these options, an undue penalty could be avoided.

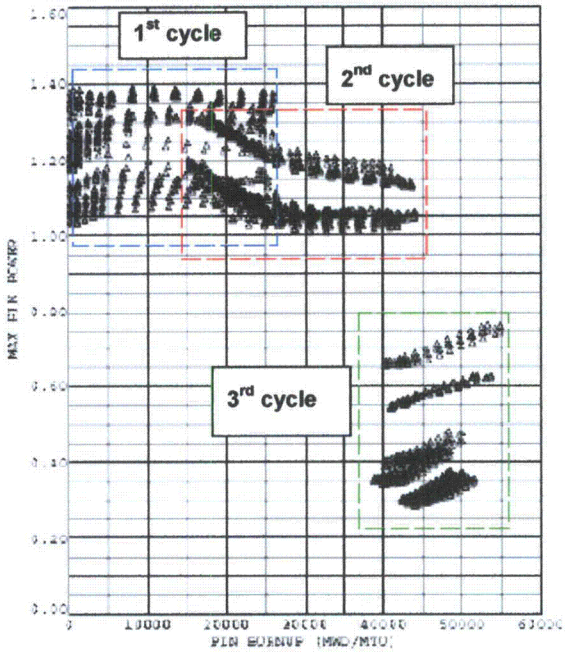


Fig. 8. Power levels for a typical Westinghouse core.

Nevertheless, it is possible to locate 3rd cycle fuel in the core interior where power levels would be high, and there are also plants in which the power level actually peaks in the 2nd cycle as the result of using burnable poisons. These cases are probably more important than the low power cases, and testing at 1200°C is appropriate for them.

It should also be remembered that (1) the corrosion layers and corresponding hydrogen contents of the high-burnup samples tested are reasonable upper-bounds for intermediate-burnup fuel, and (2) samples with 500-600 wppm reached the oxidation limit at <1200°C (e.g., 1160°C for high-burnup ZIRLO) during the heating ramp such that the test results are already somewhat modified for the rods with the largest penalty. Therefore, the test results that have been obtained for oxidation at 1200°C are the results that are most important for assessing fuel behavior during a LOCA.

Q4. Is double-sided oxygen pickup needed in LOCA analysis for non-ballooned axial nodes?

A4. During normal operation, the fuel-to-cladding gap closes and a reaction takes place between UO_2 (and PuO_2) fuel pellets and the zirconium-alloy cladding. Enough oxygen goes into the cladding surface to form a bonding layer that is largely ZrO_2 and sometimes a mixture of ZrO_2, UO_2, and PuO_2. This bonding layer adheres tightly to the cladding and, when fully developed, does not break off as the cladding pulls away from the fuel during a LOCA.

At high burnup, therefore, the cladding is sandwiched between two rich oxygen sources as shown in Fig. 9. When such cladding is heated up during a LOCA, oxygen diffuses into the cladding from the inside diameter (ID) as well as the outside diameter (OD). Because the oxides contain about 26% oxygen by weight and it takes a concentration of less than 1% to embrittle zirconium in the beta phase, there will usually be ample oxygen on both surfaces regardless of the actual oxide thicknesses. Further, since the cladding

is nearly isothermal during a LOCA, the rate of oxygen diffusion into the metal will be the same from the ID and the OD.

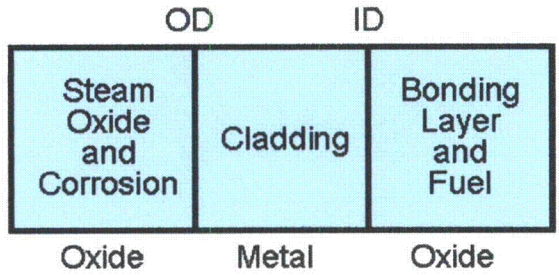


Fig. 9. Illustration of oxygen sources on outer (OD) and inner (ID) surfaces of cladding on high-burnup fuel.

Therefore, double-sided oxygen pickup is needed in LOCA analysis for non-ballooned axial nodes in high-burnup fuel, but not in low-burnup fuel. The problem is that sufficient testing has not been done to determine the manner in which oxygen begins to be picked up on the inside surface of the cladding at intermediate burnup levels.

Conservatively assuming double-sided oxygen pickup at all burnups may not lead to unnecessary penalties, however. See Fig. 10, which would be typical for many LOCA scenarios.

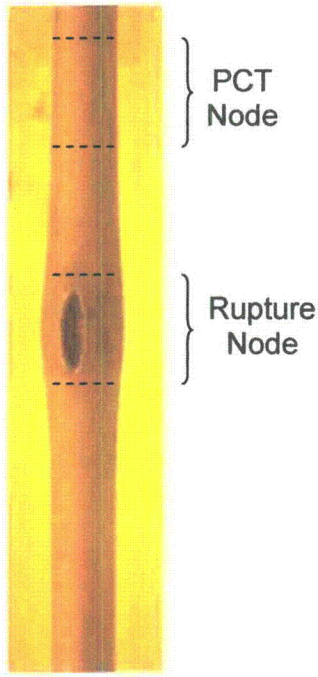


Fig. 10. Ballooned and ruptured region of high-burnup fuel rod

Rupture occurs at a relatively low temperature and significant oxidation does not take place until later. Therefore, every fuel rod that approaches its oxidation limit will already have a ruptured balloon for which two-sided oxidation is calculated according to the

current regulation (no change proposed in this part). If two-sided oxidation in the peak-cladding-temperature (PCT) node were included in the analysis, it would only become limiting if its calculated CP-ECR were higher than that in the balloon. Although the PCT node will often reach a higher temperature than the rupture node, the CP-ECR in the rupture node will be greater for a given temperature because of wall thinning. In the two plant cases we have been able to evaluate, the CP-ECR in the rupture node was greater than the CP-ECR in the PCT node with two-sided oxidation.

In the event that the PCT node is limiting for low-burnup fuel, the wording of the proposed rule would permit a licensee or vendor to obtain sufficient data to determine the burnup at which ID oxygen pickup begins and use one-sided oxygen pickup away from the balloon at lower burnups in the LOCA analysis.

Q5. Is further testing needed for future zirconium-based cladding alloys?

A5. Yes, further testing is needed for embrittlement due to breakaway oxidation, but it may not be needed for high-temperature embrittlement. See Fig. 11.

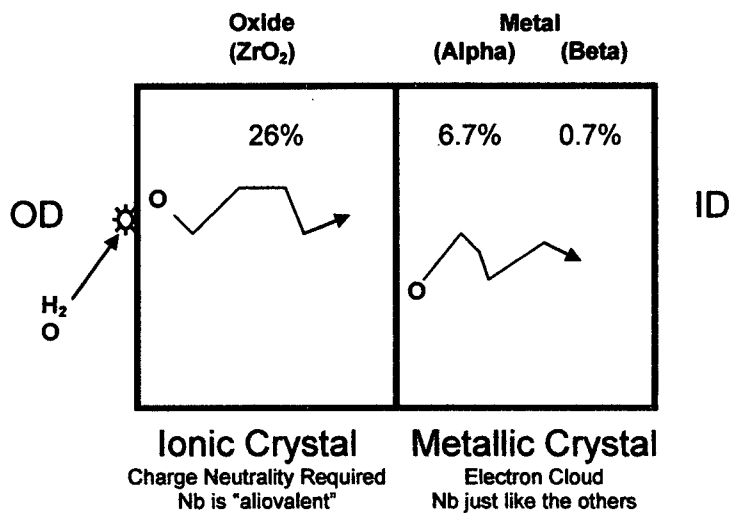


Fig. 11. Schematic of oxygen diffusion from the outside diameter (OD) of a fuel rod.

Diffusion of oxygen in the oxide controls the rate of oxidation whereas diffusion of oxygen in the metal controls the rate of embrittlement. Diffusion in an oxide is strongly affected by impurities, but diffusion of oxygen in a metal is not. Hence, oxidation should be sensitive to major alloying elements, as well as impurities, but high-temperature embrittlement should not. Data in NUREG/CR-6967 support this pattern.

For example, breakaway oxidation has been found to be highly sensitive to many fabrication details, especially surface finish. Stability of the oxide layer is dependent on maintaining compressive stresses and a hypostoichiometric oxide. Certain alloying and trace elements with an ionic charge greater than +4 can cause the oxygen-to-metal ratio to increase and lead to instability. Alloys containing Nb appear to be more sensitive to pickling in HF-containing acids (cleaning solutions) than Zircaloys.

Therefore, periodic testing is needed for breakaway oxidation and is included in the proposed rule. Because our testing experience with current cladding materials used in the U.S. has been positive, this testing could be infrequent (e.g., once per year). Nevertheless, this testing should be performed on cladding that has undergone the entire fabrication process (including end-cap welding) up to the point of insertion into a fuel assembly in order to capture all surface preparations. However, high-temperature embrittlement of future zirconium-based alloys may be the same as those already tested, so a requirement for periodic embrittlement testing is not included in the proposed rule. For confirmation, NRC could require generic high-temperature embrittlement testing of new alloys as part of the review and approval process that is conducted for each new cladding material, if that were desired.

Q6. Is extrapolation of the data to zero oxidation at 800 wppm hydrogen appropriate?

A6. It may not be accurate, but it is reasonable.

Figure 12 shows results with unirradiated samples that have been prehydrided, and the data trend begins to flatten around 450 wppm. If these data were extrapolated, they would not go to zero at 800 wppm. The explanation for this behavior is that the heatup rate used in the tests, which was adjusted to be realistic, was slow enough that the embrittlement CP-ECR does not always occur before reaching 1200°C. Starting around 6% CP-ECR, the peak temperature reached in the test is below 1200°C and decreases as the embrittlement CP-ECR value decreases. At these lower temperatures, the embrittlement threshold increases modestly because of the reduced oxygen solubility in the beta phase.

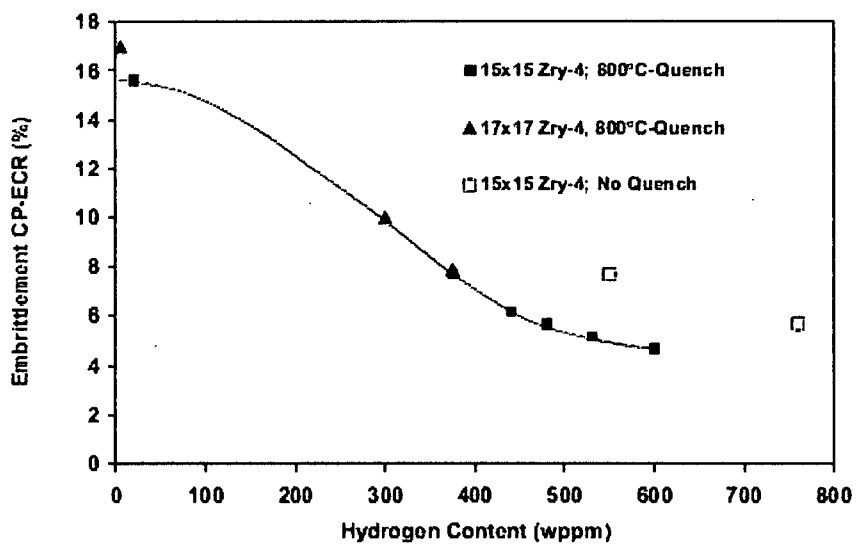


Fig. 12. Embrittlement oxidation level for prehydrided samples heated at ≤1200°C.

The same trend is presumed to exist in the more limited data base with irradiated cladding. Inasmuch as the two data points that determine the right-hand position of the limit line in the proposed rule are at 5% CP-ECR, this 5% value is already somewhat elevated compared to what it would be for a fast ramp to 1200°C and hold at 1200°C. Additional refinement of the shape of the limiting curve was not attempted because the

precision of the data base and the uncertainties in plant analysis do not seem to warrant it.

On the other hand, cladding with 600-800 wppm hydrogen would be nearing its end of life and should be located in low-power core locations (see Q3). It is likely that such fuel would not rupture during a LOCA and could be removed from consideration by generic analysis. If for any reason that portion of the limit curve continued to be a problem for a licensee, a licensee would be free under the proposed rule to provide further data and data assessment to refine that portion of the curve.

Q7. Can prehydrided unirradiated cladding be used in LOCA embrittlement testing as a surrogate for irradiated cladding with the same hydrogen concentration?

A7. Probably, but there are issues with hydrogen distribution that have to be addressed.

Embrittlement testing under LOCA conditions involves high temperatures and phase transformations that are believed to anneal out work hardening and irradiation hardening of irradiated material before significant oxidation occurs. Hydrogen from corrosion is the only parameter that appears to be responsible for the reduction in the embrittlement threshold in high-burnup cladding.

In Fig. 13, embrittlement data for high-burnup Zircaloy-4, ZIRLO and M5 are presented, along with embrittlement data for prehydrided Zircaloy-4, as a function of average hydrogen content. For high-burnup ZIRLO, the post-test hydrogen content is used because it should represent the hydrogen in the metal after corrosion-layer hydrogen is driven off during the LOCA test heating ramp.

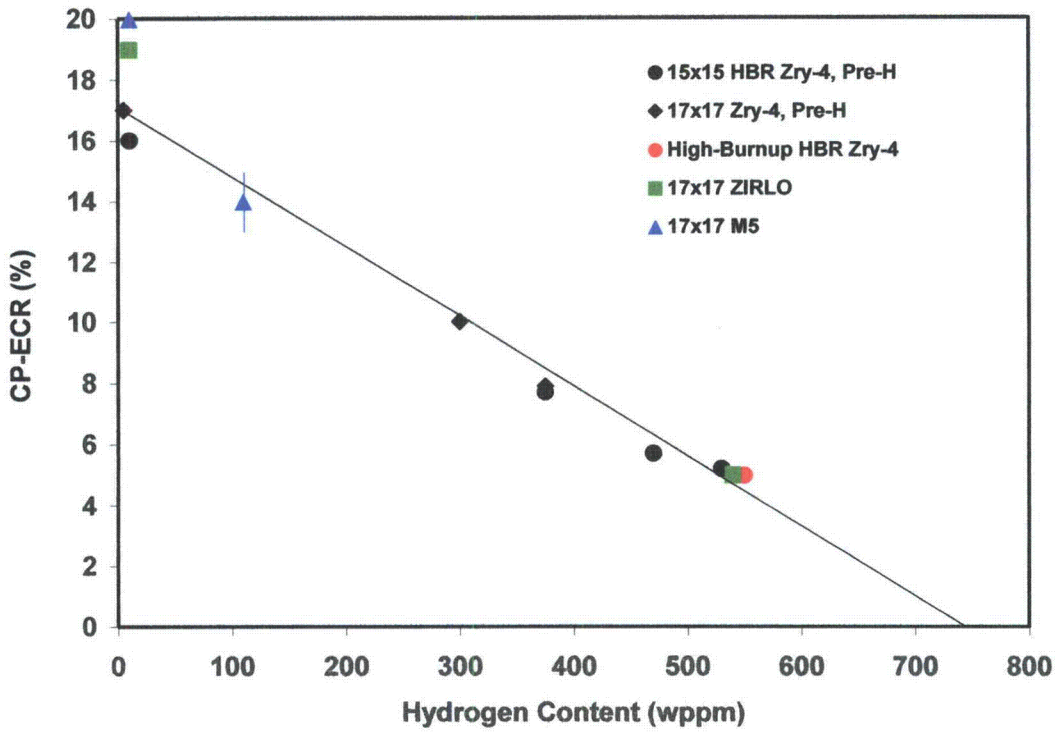
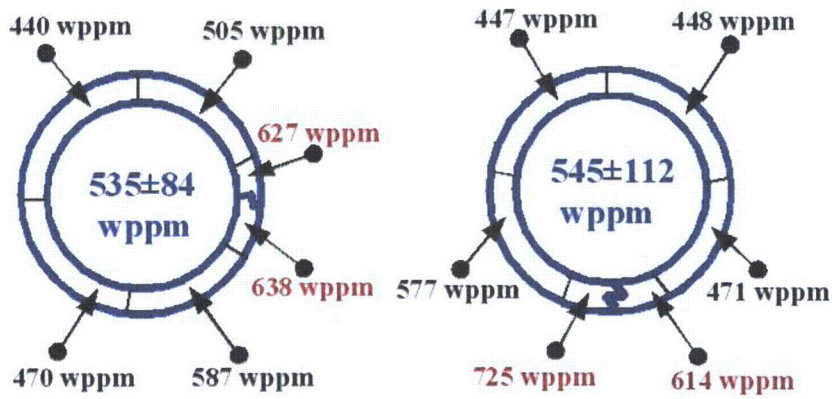


Fig. 13. Embrittlement data for high-burnup Zircaloy-4, ZIRLO and M5 as well as unirradiated prehydrided Zircaloy-4.

The embrittlement data for high-burnup and prehydrided cladding appear to be consistent, and the results suggest that prehydrided cladding should be a reasonable surrogate for high-burnup cladding. However, there are two issues that need to be addressed.

The first issue is the significant difference observed for high-burnup ZIRLO between measured pre-test hydrogen (620±140 wppm) and post-test hydrogen (540±100 wppm). The results strongly suggest that some of the pre-test hydrogen is in the ZIRLO corrosion layer, where it would not affect embrittlement. Given that the current practice in hot cells is to measure hydrogen content with the corrosion layer intact, it is not clear what the target hydrogen content should be in the prehydrided cladding.

The second issue has to do with the large circumferential variation (about ±100 wppm) in hydrogen content for ZIRLO and Zircaloy-4, which leads to local variations in hydrogen content, ranging from about 450-700 wppm for those samples. Figure 14 shows the circumferential variation in hydrogen concentration that was measured following LOCA-type steam oxidation to about 6% CP-ECR and ring-compression testing at 135°C for ZIRLO specimens.



(a) 30% ductility (b) 10% ductility

Fig. 14. Hydrogen variation in two high-burnup ZIRLO rings following oxidation to 6% CP-ECR, slow-cooling, and ring compression testing.

Both rings were sectioned from the same 25-mm-long LOCA test sample. ANL results show that such variation in hydrogen content is confirmed by the circumferential variation in hydride rim thickness observed in metallographic images. These hydrogen variations are real, therefore, and are not the result of experimental uncertainties. The results also indicate that very little diffusion or homogenization of hydrogen occurs in the circumferential direction during the LOCA test transients. For prehydrided cladding with uniform hydrogen concentration, it is not clear if the target concentration should be at the average value, at the maximum value, or at a value in between for high-burnup cladding.

Q8. Can the present results be applied equally to UO_2 and MOX fuel?

A8. Yes. Embrittlement of the cladding, which is the subject of the proposed rule change, is caused by oxygen diffusion into the cladding metal. The oxygen on the outside diameter (OD) of the cladding comes from steam and is therefore independent of the type of fuel pellet inside the cladding.

However, the source of oxygen on the inside diameter (ID) of the cladding away from the rupture is the fuel pellet itself, so this source needs to be considered to determine if UO_2 and MOX fuel pellets might affect ID oxygen ingress differently.

Figure 15 shows the free energy of formation of an oxide for different metals. The larger values near the bottom of the plot represent a greater affinity for oxygen. Notice that U, Pu, and Th, the most common fissile materials, have almost the same affinity for oxygen as Zr such that oxygen would be exchanged freely among them. Many other metals would give up their oxygen to Zr more readily such that all oxide fuel types would provide oxygen to the zirconium-alloy cladding ID.

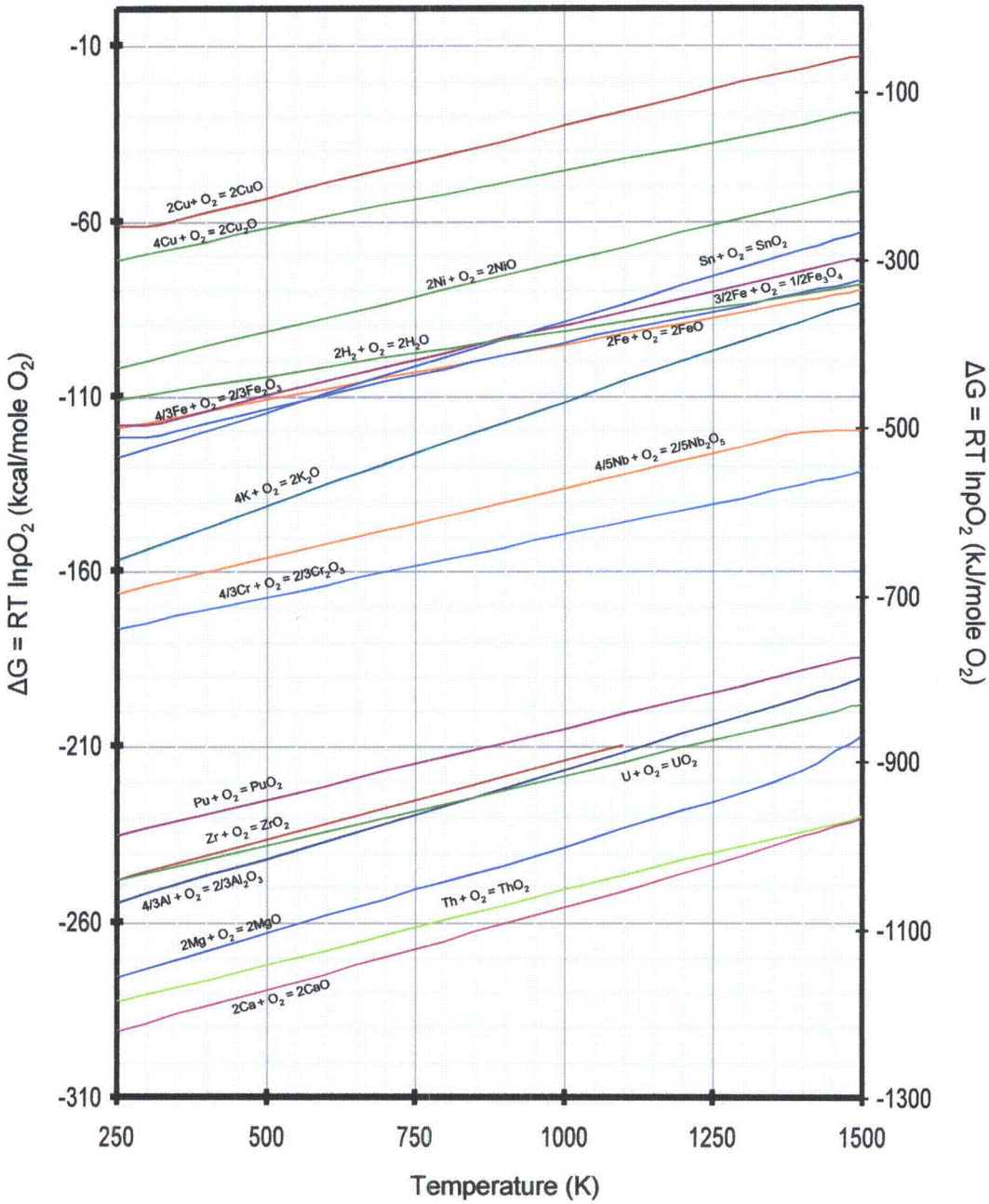


Figure 15. Free energy of formation of oxides of alloy constituents and some impurities.

Generally, the amount of oxygen on the surfaces of the cladding (i.e., the thickness of the OD and ID ZrO_2 layers) is more than sufficient to provide oxygen for diffusion into the metal and cause embrittlement. Data for high-burnup fuel tested under LOCA conditions show exactly the same amount of oxygen diffusing into the metal from the ID and the OD in spite of the fact that the ZrO_2 layers were of different thickness and the oxygen came from different sources. Further, a lot of PuO_2 was present near the pellet surface of this UO_2 fuel because of its high-burnup.

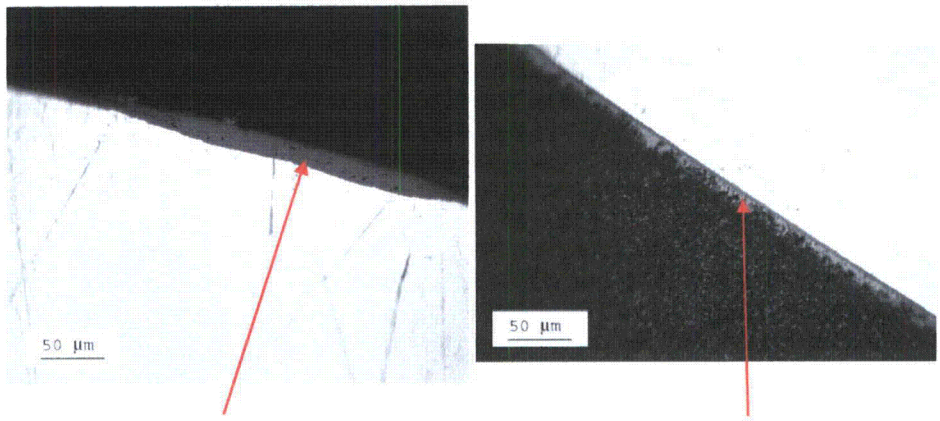
Notice that metal fuel would not provide an oxygen source for ID oxygen pickup and that a metal fuel might form low-melting eutectics with a zirconium-alloy cladding material. Thus the proposed rule would not be appropriate for metal fuels, but it would work for all oxide fuel types. Of course MOX fuel might provide a different heat source for LOCA analysis compared with UO_2 fuel, but that difference would be handled in the evaluation models and not in the embrittlement criteria, which are the subject of the rule change.

Q9. Should CRUD be included in LOCA analysis?

A9. Mineral deposits referred to as CRUD accumulate on top of the corrosion layer, which is an oxide (ZrO_2) on the surface of the fuel rod cladding. CRUD may affect heat transfer, which would be handled in the safety analysis. Thus CRUD may affect cladding temperatures and hence the amount of corrosion and hydrogen absorption during normal operation. However, CRUD will have no direct effect on embrittlement and should not have an effect on breakaway oxidation, which are the subjects of the proposed rule change.

In principle, elements in CRUD could diffuse into the surface oxide and then into the metal where they would behave as impurities that could affect breakaway oxidation and embrittlement. Intimate contact between the CRUD and the OD oxide surface would be required for this transfer so only tenacious CRUD (not fluffy CRUD) needs to be considered. The main elements in tenacious CRUD are typically Fe, Ni, Cu, and Cr. Of these, only Cr might be able to get into the oxide because of the relatively low affinity for oxygen of these metals compared with Zr (see Fig. 15).

If Cr diffused through the oxide and into the metal, it would have no effect on embrittlement because Cr is already a common alloying element in the zirconium-based cladding materials and results show that embrittlement is insensitive to these elements. On the other hand, these alloying elements have some effect on oxidation rates and, therefore, Cr might have an impact on breakaway oxidation. But breakaway oxidation appears to be caused by surface roughness and surface contaminates on the fresh metal rather than anything that develops after the surface becomes oxidized. Figure 16 shows tenacious CRUD that varied azimuthally on a high-burnup BWR fuel specimen. Although the CRUD thickness varied considerably, no unusual oxidation behavior was seen, and this cladding had a very long breakaway-oxidation time.



Thick Corrosion Layer
No Crud Layer

Thin Corrosion Layer
Thick Crud Layer

Fig. 16. Tenacious CRUD of varying thickness on high-burnup BWR cladding.

Q10. Do manufacturing variability and trace elements have a significant effect on LOCA behavior?

A10. Yes for breakaway oxidation, but identifying trace elements that are detrimental to oxide stability is very challenging. The following examples illustrate this conclusion.

During the past 30 years there have been significant improvements in the manufacturing of cladding materials used in U.S. reactors. In general, these improvements have been made to optimize cladding performance under normal reactor operating conditions. However, many of these improvements have also resulted in improved performance under LOCA conditions. For Zircaloy-4, improved performance under LOCA conditions has resulted from the elimination of pickling as a final finishing process as well as the adoption of belt or wheel polishing for the cladding outer surface and grit polishing for the inner surface. During the 1970s and part of the early 1980s, pickling with a mixture of hydrofluoric and nitric acid was used to remove surface impurities and, in some cases, about 25 microns of the outer surface. Such treatment, especially in combination with high surface roughness, resulted in enhanced in-reactor corrosion, nodular corrosion, and reduced time for breakaway oxidation under LOCA conditions.

During the period of 1980-1985, Leistikow & Schanz generated and published a large database on the oxidation behavior of Zircaloy-4 exposed to steam at temperatures from 600-1600°C. It can be assumed that they used 1970s vintage cladding with high surface roughness and pickled surfaces. Based on the ANL breakaway-oxidation criterion of 200-wppm hydrogen pickup, the minimum breakaway time for this 1970s Zircaloy-4 was about 1800 s at 1000°C. In the ANL breakaway oxidation studies, both older, rough-surface (ca 1980s) and modern polished Zircaloy-4 were tested, and the minimum breakaway times at 1000°C were found to be 3800 s and 5000 s, respectively. Although most of the fabrication processes are vendor-proprietary, the increase in minimum breakaway oxidation time from 1800 s to 5000 s is a dramatic example of the effects of manufacturing processes, particularly surface finish, on breakaway oxidation.

Another example involves differences in minimum breakaway oxidation times between two Zr-1%Nb alloys, standard Russian E110 and AREVA M5. These differences are

even more dramatic than what has been observed for vintages of Zircaloy-4. Based on ANL results, minimum breakaway times for standard E110 oxidized at 1000°C were observed to be <600 s, while the breakaway oxidation time reported by AREVA for M5 oxidized at 1000°C was >6000 s. Because the standard E110 tubing supplied to ANL has high surface roughness and was cleaned by pickling, ANL polished some samples to reduce surface roughness and impurities, and ANL machined and polished other samples to remove surface and substrate impurities. These finishing steps resulted in about a factor of two increase in breakaway oxidation time at 1000°C, still well below breakaway times for modern Zircaloy-4 and M5. These finishing steps also produced very stable oxide-layer growth on the E110 at 1100°C and 1200°C.

In parallel, some other E110 samples were pickled in an HF-containing solution of a type reportedly used in the early 1980s for Zry-2. Such surface treatment results in fluoride surface impurities that significantly degraded the oxide layer after short-time exposure to steam at 1000°C. Based on the ANL studies, it appears that E110 has detrimental impurities on the surface (within a micron), in the substrate (within 40 microns), and in the bulk. In addition to surface impurities, high-surface roughness and scratches resulted in local regions of unstable oxide growth and breakaway oxidation. It was also observed that E110 was much more sensitive to surface treatments, such as pickling, than Zircaloy-4.

Regarding impurities in the bulk, the Russian manufacturer, TVEL, studied the steam oxidation behavior at 1100°C for 600 s of seven lots of E110 with varying levels of alloy and impurity levels. They found improved oxide layer integrity, decreased hydrogen pickup, and increased ductility for lots with 25-45 wppm impurities (sum of Ni, Al, Si, Ca, K, F, Cl, Na, and Mg) as compared to lots with 110-135 wppm of these impurities. However, TVEL could not isolate which of these impurities was detrimental.

Perhaps the most compelling data with regard to the effects of bulk impurities comes from the Russian KI/RIAR study comparing the performance of E110 made from a mixture of iodide and electrolytic Zr with the performance of E110 made from French sponge Zr. Based on the presentation of results in NUREG/IA-0211 in terms of measured ECR, it is difficult to discern the dramatic benefits of ingots made from sponge Zr. However, when expressed in terms of test times at 1000°C and 1100°C, as in done in Table 1, it is clear that E110 made from sponge Zr has superior performance.

Table 1. Russian results for E110 made from ingots composed of iodide and electrolytic Zr plus 1 wt.% Nb and for otherwise identical $E110_{G(fr)}$ made from ingots composed of sponge Zr plus 1 wt% Nb

Test #	Material	T, °C	t, s	H, wppm	Ductility, %	
					RT	135°C
44	E110	1000	865	NM	0	NM
93	$E110_{G(fr)}$	1000	5013	12	1	54
41	E110	1100	284	1130	0	NM
90	$E110_{G(fr)}$	1100	933	≤48	17	60

Q11. Could strength be used instead of ductility as a performance indicator during a LOCA?

A11. Thirty five years ago, proposals to base LOCA requirements on strength were discussed at a lengthy ECCS hearing. The Commission decided against strength-based criteria because they did not believe that loads during a LOCA could be known and therefore they were unable to determine what minimum strength to require. Instead, they adopted ductility-based criteria (see p. 1098 of the 1973 Commission opinion).

> *The stress calculations, the measurements of strength and flexibility of oxidized rods, and the thermal shock tests all 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.*

When NRC started the current research project about ten years ago, the goal was to determine the effects of fuel burnup on the existing LOCA criteria. That has been done. About midway through this research, the discussion of strength-versus-ductility came up again. At that time, NRC noted that the basis for the criteria should not be changed, and it was pointed out that load assessment was not included in NRC's research plans.

Of course it would be possible to change the basis for the LOCA criteria. But it would be necessary to define LOCA loads as well as to characterize fuel rod strength. If the U.S. industry wants to go in this direction, a petition can always be made to NRC under 10 CFR 2.802. However, this would be far more difficult than measuring the effect of burnup on ductility, and it would involve a reduction in safety margin that might be controversial.

Q12. Were results from other laboratories considered and are they consistent with ANL's results?

A12. Yes, results from other labs were considered. Results were consistent when test conditions and data interpretation methods were similar. A summary of these results and related interactions with other laboratories follows.

France

AREVA and EdF have sponsored embrittlement work at CEA with as-fabricated and prehydrided Zry-4 and M5. Prior to mid 2005, the post-quench data published by AREVA, EdF and CEA were based on room-temperature (RT) ring-compression, bend, and impact tests. For ring-compression and bend tests, the total (elastic + plastic) displacement was plotted versus measured weight gain or measured ECR. Because PQD must be determined at 135°C in terms of plastic deformation and CP-ECR, no useful PQD results can be determined from these early published data.

In 2005, AREVA formalized a process of data exchange with ANL such that all results went through AREVA. A day-long data exchange with CEA, EdF, and AREVA was held at ANL on May 9, 2005. Many of the details regarding CEA test protocol (e.g., heating rates) came from this meeting. The purpose of the data exchange was to help ANL in planning PQD tests with prehydrided and high-burnup Zry-4. ANL was permitted to use the data for test planning, but could not publish them. Since 2005, CEA has been a regular participant in program review meetings, not to mention frequent e-mail

interactions between Billone at ANL and Brachet at CEA regarding draft materials and issues.

Both ANL and CEA have generated data for prehydrided Zircaloy-4. Although the Zircaloy-4 cladding vendors were different, the data trends are consistent. But a quantitative comparison cannot be made between the data sets because CEA used unrealistic heating and cooling rates which biased their results. The heating ramps from 1000°C to 1200°C (fast for CEA, slow for ANL) and the quench temperatures (1200°C for CEA and 800°C for ANL) were quite different. The rapid temperature rise and the quench at 1200°C tend to reduce ductility at the same oxidation level. For prehydrided Zircaloy-4 samples quenched at 600°C, ANL cooling time (120 s) was much faster than CEA cooling time (2200 s), resulting in lower ductility for the ANL samples.

Later, ANL used the CEA data for prehydrided M5 to plan test times for high-burnup M5. However, CEA did not pursue tests to determine the ductile-to-brittle oxidation level as a function of hydrogen content. For example, the CEA results for prehydrided M5 with 120-wppm H oxidized at 1200°C were low ductility at 11% CP-ECR and no ductility at 18% CP-ECR. All one can conclude from this CEA data set is that prehydrided M5 embrittles between 11% and 18% CP-ECR (ANL measured 14% for high-burnup M5 with 110-wppm H).

As early as 2004, CEA, with AREVA and EdF approval, began sharing PQD data in tabular form, as well as load-displacement curves for each sample tested. The data were divided into two sets: data from which published graphs were generated and unpublished data. The tabular data for published graphs included actual hydrogen content (as high as 740 wppm vs. published target value of 600 wppm for Zry-4), test time, measured weight gain and ECR, CP-ECR, ring-compression test temperature (RT and 135°C), total and plastic strain to failure. The tabular data sets are very useful because they allow for direct comparison in terms of CP-ECR. CEA also provided ANL with some proprietary data in graphical form for 600-wppm Zry-4 oxidized at 1000°C, and CEA has shown ANL additional unpublished PQD data for 300-wppm Zry-4 oxidized at 1200°C and ring-compressed at 135°C. These tests remained at their target temperature long enough that the effects of the unrealistic heat-up and cool-down rates were minimized. As mentioned above, the unpublished data were provided to ANL to help plan test matrices for PQD studies. However, ANL is not allowed to publish these proprietary data or to share them, especially with NRC and EPRI. Nevertheless, the data are known to ANL staff and do not reveal any inconsistencies with ANL data.

Japan

In 2001 it was revealed that the JAEA criterion used to assess LOCA cladding behavior had been changed to a strength-based concept and the testing method used for evaluation involves an axial restraint leading to axial tensile loading during a simulated LOCA. Consequently, JAEA has performed only a limited number of ring compression tests and these have been with as-fabricated and prehydrided cladding. Most of their published data on PQD of oxidized rings are not very useful because the ring-compression tests were conducted at RT and effects of cooling rate and quench temperature were compared based on 20% offset strain.

Nevertheless, close collaboration has been maintained with JAEA regarding the results of LOCA integral tests. The JAEA data are consistent with ANL integral test data with

respect to secondary hydrogen pickup in the balloon region following burst and oxidation. However, differences in sample characteristics (fueled for ANL, defueled for JAEA) and in internal pressure and gas volume (higher for ANL, very low for JAEA) render one-to-one comparisons of results less meaningful.

Russia

The LOCA studies at ANL and Russian KI/RIAR both received funding and direction from NRC, and these studies were conducted in parallel with considerable cooperation and exchange of testing methodology and data. Both ANL and KI/RIAR observed the same early breakaway oxidation on E110 at 1000°C and 1100°C. Nevertheless, the differences in the presentation of data make comparison of graphical results in the final reports difficult. However, when results are expressed in terms of test time or calculated CP-ECR, rather than measured ECR, the results are consistent. The Russian results were published in NUREG/IA-0211 prior to full realization of the need to use CP-ECR as a surrogate (variable transformation) for time and the irrelevance of measured ECR values for embrittlement.

Other

Cognizance of LOCA-relevant research in other countries has been maintained with formal meetings and personal contacts. In May 2004, ANL hosted an OECD LOCA Topical Meeting, which was attended by 60 participants from 16 countries who made 20 presentations. Annual multi-day program review meetings at ANL have had participation from Russia (Kurchatov), France (AREAV, CEA, IRSN, EdF), Japan (JAEA), Germany (AREVA), Norway (Halden), Sweden (Studsvik), Spain (CSN), Finland (VTT), as well as all segments of the U.S. industry including EPRI. Staff from ANL and NRC have visited various laboratories performing LOCA tests, including CEA, JAEA, Halden, UJP (Czech Republic), and others. Comparisons of ANL data on cladding embrittlement with others are usually not valid because of differences in ring-compression test temperatures, differences in heating rates, and differences in cooling rates. ANL has clearly developed more advanced testing techniques, and these techniques need to be standardized for future industry testing.

Q13. Is ring-compression testing non-prototypical and overly conservative?

A13. Ring compression tests are standard ductility screening tests used for materials that transition from ductile to brittle failure. They are not designed to represent any particular load so the question of prototypicality is not relevant. Although testing and data-analysis techniques have vastly improved since 1973, ring-compression test results were used in 1973 to define the 17% oxidation and 2200°F temperature limits to ensure post-quench ductility.

Compressing a ring induces hoop bending stresses that transition from tensile to compressive across the wall of the cladding. Other testing techniques used to determine the transition from ductile to brittle behavior include loading modes that are similar (e.g., axial bending) or more severe (e.g., axial or hoop tension). CEA and KI-RIAR have conducted companion axial bending tests using oxidized cladding samples. They both found that ductility and embrittlement results determined from axial bending tests were comparable to those determined from ring compression tests. As such, it appears that if oxidized cladding embrittles in the hoop direction, it will also embrittle in the axial

direction. This similarity is expected because embrittlement in the prior-beta phase is independent of loading direction.

A14. Does axial fuel relocation need to be addressed in the revised rule?

A14. No, although fuel relocation may need to be addressed in a LOCA analysis.

Axial relocation of fuel particles into the balloon will increase the heat source in the balloon and can be addressed in the evaluation models. While this will increase the temperature in the balloon node and hence increase the calculated oxidation there, fuel relocation will not affect the limiting oxidation value that has been derived directly from measurements.

Fuel loss, which is an extreme case of fuel relocation, has been observed in fuel with a burnup of 92 GWd/t. This is significantly above the maximum burnup that is currently permitted (62 GWd/t) in the U.S. Although such fuel loss may be tantamount to loss of coolable geometry, it also will not affect the limiting oxidation value associated with cladding embrittlement.

Q15. Are more data needed on cooling-rate effects before rulemaking begins?

A15. No. Figure 17 shows the major research results for two cooling rates. The faster cooling rate, labeled 800°C Quench, involved cooling to 800°C before quenching and was designed to be bounding for cooling rates that are expected in LOCA analysis. The limiting oxidation value in the proposed rule modification is based on this faster rate and should thus be adequate for all analyzed sequences. The slower cooling rate involved the same cooling to 800°C followed by natural cooling with the furnace turned off.

Should a licensee desire to refine its LOCA analysis and use an oxidation limit for a slower cooling rate, the proposed rule would permit the licensee to use these "No Quench" data or to generate additional data to justify a higher limit.

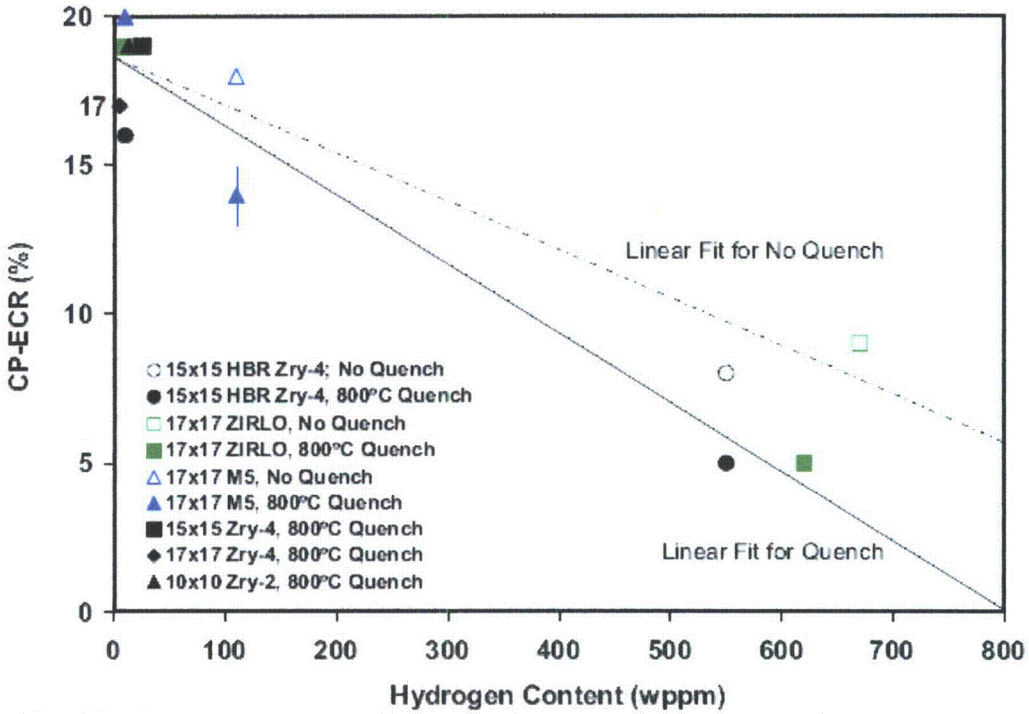


Fig. 17. Embrittlement threshold expressed as an oxidation level (CP-ECR) vs. pre-test hydrogen content for as-fabricated cladding alloys and high-burnup Zircaloy-4, ZIRLO, and M5 cladding, which were oxidized at ≤1200°C and either quenched at 800°C or cooled without quench.

Q16. Are more data needed on ballooning and flow blockage before rulemaking begins?

A16. No. Ballooning strain and the amount of flow blockage are needed in LOCA analysis, but they do not affect the limiting oxidation value, which is the subject of the current rulemaking effort. Some ballooning strain and flow blockage values being used by licensees may be out of date, but those values are not prescribed by NRC. NRC plans to do further testing to assess the effect of burnup on ballooning and blockage in order to assist the staff in its review of licensing evaluation models.

Rulemaking Comments

From: Ralph Meyer [rmeyer@anl.gov]
Sent: Monday, October 05, 2009 2:42 PM
To: Rulemaking Comments
Cc: Scott, Harold; billone@anl.gov
Subject: Comments on ANPR (RIN 3150-AH42)
Attachments: Response to 2008 FR Comments.pdf

Rulemaking Comments:

Please accept the attached comments on the Advance Notice of Proposed Rulemaking of August 13, 2009 (RIN 3150-AH42).

Ralph Meyer

Received: from mail1.nrc.gov (148.184.176.41) by TWMS01.nrc.gov
(148.184.200.145) with Microsoft SMTP Server id 8.1.393.1; Mon, 5 Oct 2009
14:42:17 -0400
X-Ironport-ID: mail1
X-SBRS: 5.3
X-MID: 6882338
X-IronPort-AV: E=Sophos;i="4.44,507,1249272000";
d="pdf'?scan'208,217";a="6882338"
Received: from ne-exch.ne.anl.gov ([146.139.96.22]) by mail1.nrc.gov with
ESMTP; 05 Oct 2009 14:42:15 -0400
Received: from smtp.anlmail.anl.gov ([146.137.222.81]) by NE-EXCH.ne.anl.gov
with Microsoft SMTPSVC(6.0.3790.3959); Mon, 5 Oct 2009 13:42:15 -0500
Received: from MeyerLaptop ([69.2.176.156]) by smtp.anlmail.anl.gov over TLS
secured channel with Microsoft SMTPSVC(6.0.3790.3959); Mon, 5 Oct 2009
13:41:58 -0500
From: Ralph Meyer <rmeyer@anl.gov>
To: <Rulemaking.Comments@nrc.gov>
CC: "'Scott, Harold'" <Harold.Scott@nrc.gov>, <billone@anl.gov>
Subject: Comments on ANPR (RIN 3150-AH42)
Date: Mon, 5 Oct 2009 14:42:07 -0400
Message-ID: <001001ca45eb$8b1cc4f0$a1564ed0$@gov>
MIME-Version: 1.0
Content-Type: multipart/mixed;
boundary="----=_NextPart_000_0011_01CA45CA.040B24F0"
X-Mailer: Microsoft Office Outlook 12.0
Thread-Index: AcpF64qQejqGa80nSH6b3Asfjtkg7g==
Content-Language: en-us
Return-Path: rmeyer@anl.gov
X-OriginalArrivalTime: 05 Oct 2009 18:41:58.0994 (UTC) FILETIME=[85534B20:01CA45EB]