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From: Harold Scott
To: Ralph Meyer
Date: 2/6/06 1:43PM
Subject: December memo to T Martin

A-29

December 21, 2005

MEMORANDUM TO: Thomas O. Martin, Director
Division of Safety Systems
Office of Nuclear Reactor Regulation

FROM: Farouk Eltawila, Director */RA/*
Division of Systems Analysis and Regulatory Effectiveness
Office of Nuclear Regulatory Research

SUBJECT: EXECUTIVE SUMMARY OF DEFICIENCIES IN ACCEPTANCE CRITERIA FOR LOSS-OF-COOLANT ACCIDENTS

In our research to develop a performance-based option for the acceptance criteria in 50.46(b), we found a number of circumstances under which the current criteria might not provide the protection we expect (i.e., the 2200°F temperature limit and the 17% oxidation limit might not prevent cladding embrittlement). Although these situations are mentioned in passing in a draft Research Information Letter (RIL) that was recently reviewed by your staff, their significance might not be apparent, so we are providing this executive summary for easy reference. Because of the way licensees are using this regulation, however, there is no indication of problems with the safety of current operating reactors as is discussed below. Further, these situations are accommodated in the proposed criteria that are described in the draft RIL.

1. Peak Cladding Temperature

During 1972-1973, Hobson found that a 17% oxidation limit was not able to prevent cladding embrittlement for LOCA temperatures above 2200°F (1204°C). Therefore, when 50.46 was established in late 1973, peak cladding temperature was limited to 2200°F such that the pair of limits (17% and 2200°F, the embrittlement criteria) would ensure cladding ductility. Current research also shows that the oxidation level corresponding to the onset of embrittlement starts decreasing rapidly as the LOCA temperature approaches 2200°F. Therefore, we see no reason to alter this limit. All current LOCA analyses have been done using this limit, and the proposed criteria we described in the draft RIL retain this limit, so no issues are raised for peak cladding temperature.

2. Embrittlement of M5 Cladding

Figure 1 shows the embrittlement of M5 cladding compared with Zircaloy cladding at a relatively low temperature that might be typical of a small-break LOCA. Embrittlement (i.e., zero ductility) occurs at 2-3% offset strain, so the trend in Fig. 1 shows that M5 cladding embrittles around 14% oxidation, well below the 17% limit in 50.46(b).* Nevertheless, oxidation is so slow in M5 compared with Zircaloy that it takes just as long for M5 to reach 14% oxidation as it takes for Zircaloy to reach ~20% oxidation. Therefore, if you use a Zircaloy oxidation kinetics equation (e.g., Cathcart-Pawel or Baker-Just) to calculate the oxidation level – rather than using a true measured value – M5 cladding will exhibit embrittlement at 17% or higher just as Zircaloy does.

*Offset strain and equivalent cladding reacted (ECR) are well defined measures of deformation during a ring-compression test and of cladding oxidation, respectively.

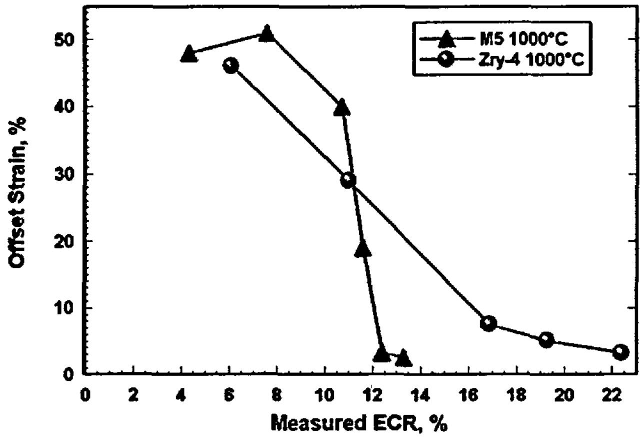


Fig. 1. Deformation versus oxidation at 1000°C for unirradiated M5 and Zircaloy cladding.

Based on statements from Framatome staff, we believe that licensees have been using the Baker-Just or Cathcart-Pawel (CP) correlation for oxidation calculations for M5 cladding up to the present time, and the 17% limit is satisfactory for those analyses. The rule in its current form permits the use of best-estimate models, and if licensees chose to use a best-estimate oxidation model for fuel with M5 cladding, the 17% limit would not provide the protection that is needed. In the proposed criteria described in the draft RIL, the Cathcart-Pawel correlation is required for all cladding alloys to accommodate this effect.

3. Rough Surfaces of Older Cladding

During the investigation of the difference in LOCA behavior between the French M5 cladding and the Russian E110 cladding, we discovered that one of the major factors was surface finish. Recent Western-made cladding that we have seen in the laboratory has had polished surfaces whereas the Russian cladding we tested had unpolished and sometimes anodized (etched) surfaces. Some older U.S.-made Zircaloy cladding also had unpolished and etched surfaces. The effect of surface finish can be seen in Fig. 2.

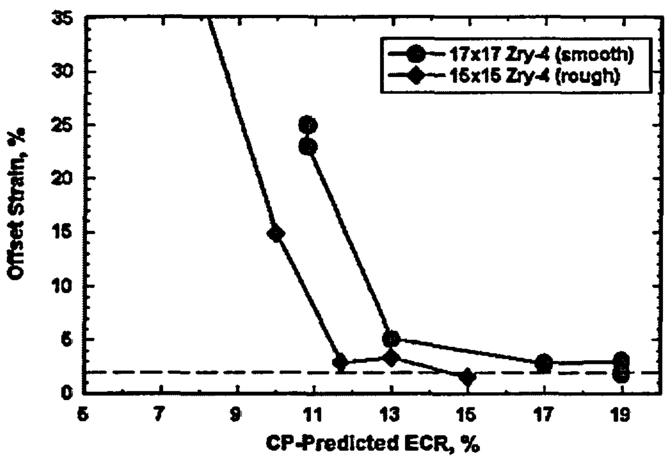
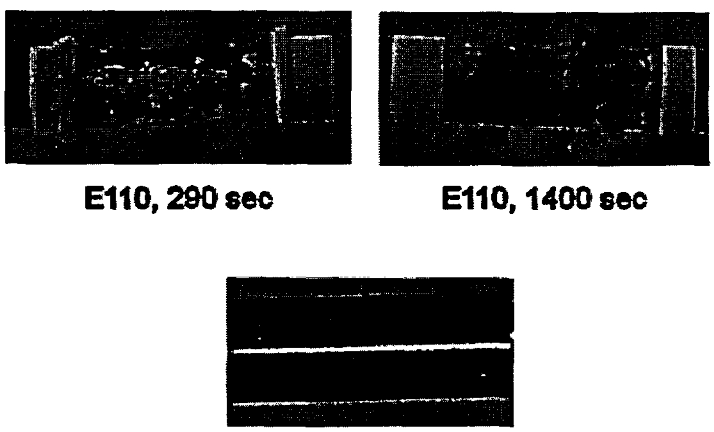


Fig. 2. Deformation versus oxidation at 1200°C for unirradiated Zircaloy (CP-Predicted same as Measured at this temperature).

The cladding with rough surfaces embrittles around 12-13% oxidation whereas the cladding with the smooth surface embrittles around 17% oxidation. When Hobson did the original work on embrittlement using specimens with rough surfaces, he did not measure oxidation, but rather calculated it with the Baker-Just correlation. Of course, the Baker-Just correlation is now known to be quite conservative, and at 1200°C the Baker-Just correlation will predict 17% oxidation for conditions that will produce only 13% measured oxidation. Because the Baker-Just correlation was required for all licensing calculations prior to 1989 when the best-estimate rule was adopted, it is likely that LOCA analyses for the older cladding with rough surfaces were adequate. In the proposed criteria that were described in the draft RIL, we have included a performance-based test that would account for any variations such as those caused by surface condition.

4. Breakaway Oxidation

The old standard Zircaloy cladding alloy contained tin and some minor alloying elements whereas the newer M5 alloy and the Russian E110 alloy contain niobium instead of tin (ZIRLO contains some tin and some niobium). Zirconium and tin both have a valence of +4 and they mix well together. Niobium has a valence of +5 and a very limited solubility in zirconium. We think that this difference makes M5 and E110 more susceptible to breakaway oxidation than Zircaloy, but all zirconium alloys have this susceptibility. There are other important factors as well, and we found that M5 is much less susceptible than E110. Figure 3 shows examples of these two alloys after oxidation at different times at 1000°C.



E110, 290 sec **E110, 1400 sec**

M5, 2400 sec

Fig. 3. The appearance of breakaway oxidation on E110 cladding and normal oxidation on M5 cladding after oxidation at 1000°C.

In addition to surface finish, the other factor that we found to be important is the source of zirconium metal used for alloy preparation. Very pure electrolytic zirconium, as used by the Russians, produces a more susceptible alloy. On the other hand, relatively impure sponge zirconium, as used in the West, appears to contain beneficial impurities (+3 valence) that suppress the breakaway process. For the Western alloys used in U.S. plants, preliminary tests show that the time required for the breakaway process to begin is longer than expected LOCA times (~1800 sec). This has been demonstrated already for unirradiated cladding and will be verified soon in tests that are planned on irradiated cladding. A performance-based time limit to preclude breakaway is included in the proposed LOCA criteria described in the draft RIL.

5. Burnup and Corrosion

Corrosion (oxidation) takes place during the normal burnup process. The existence of this corrosion, and the associated hydrogen that has been absorbed, reduces the amount of LOCA oxidation that will produce embrittlement. This can be seen in Fig. 4, where embrittlement is seen to occur around 7% oxidation during a LOCA transient.

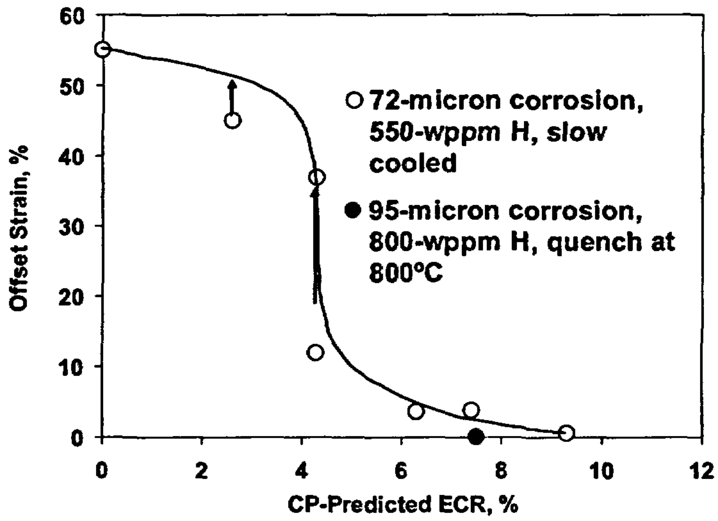


Fig. 4. Deformation versus oxidation at 1200°C for irradiated 15x15 Zircaloy (rough surface). CP-predicted oxidation is the same as measured oxidation at this temperature.

Although corrosion at operating temperatures and oxidation at LOCA temperatures have some different characteristics, the sum of these two oxidation levels appears to give a good indication of when embrittlement will occur. The data shown in Fig. 4 are for high-burnup fuel from H. B. Robinson with older Zircaloy cladding with a rough surface. The ~80 microns of corrosion thickness converts to an oxidation level of about 6%. When this is added to the 7% seen in this figure, the resultant total oxidation of 13% is approximately correct for this cladding type (see Fig. 2). We expect to get a similar result when planned tests are performed on irradiated ZIRLO and M5 cladding.

According to Information Notice 98-29 and the clarifying letter from NRC to NEI of March 31, 1999, "total oxidation" as stated in 50.46 is expected to mean pre-LOCA oxidation plus transient oxidation. Because current plant analyses should conform to this interpretation, the burnup effect should be accommodated for operating reactors. This addition of pre-LOCA oxidation and transient oxidation is included in the proposed criteria that were described in the draft RIL.

6. Localized Embrittlement in the Balloons

Requirements are given in 50.46(b) for applying the oxidation limit to the ballooned region of fuel rods. Around 1980, it was discovered in two laboratories that this limit did not ensure ductility everywhere along the ballooned region, and we have confirmed this finding in our recent research. Hydrogen, which is formed from oxidation on the inside surface of the balloon, is not swept away and is absorbed at a high rate into the cladding metal. This results in local bands of embrittlement as indicated in Fig. 5.

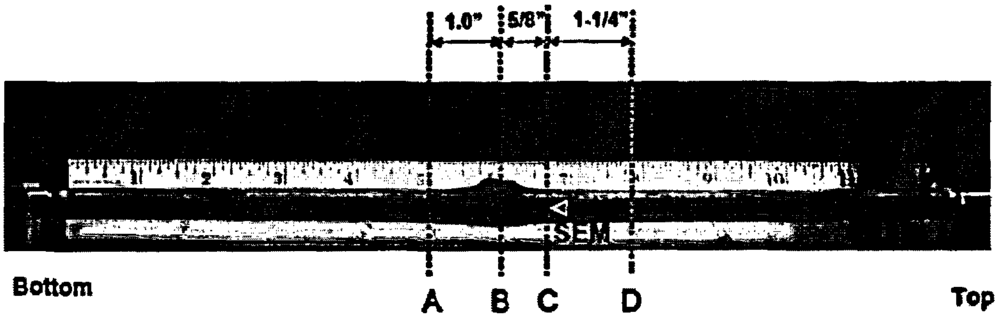


Fig. 5. High-burnup Zircaloy-clad BWR fuel rod after ballooning, rupture, oxidation, and quenching in a simulated LOCA transient.

This figure shows a low-corrosion high-burnup BWR rod that was subjected to a simulated LOCA transient with a peak cladding temperature of 1200°C. Locations A, B, and C show where the specimen broke during post-test handling and they correspond to the embrittled bands (D simply marks the location where a metallography specimen was taken). Such bands of embrittlement are also seen in tests with unirradiated tubes of all the alloys we have tested, confirming that this is neither an alloy effect nor a burnup effect.

Although we now recognize that the oxidation limit in 50.46(b) does not provide ductility everywhere in the balloon, as was intended, there are extenuating circumstances. One is that these bands seem to result in clean breaks without any shattering of the balloon wall. Such clean breaks in combination with the random axial location of the balloon (current understanding) would result in a very constrained geometry such that little fuel material could be lost from the break areas. Another is that the bands of embrittlement will be confined to just a few inches of the total length of a fuel rod (12-14 feet).

At the present time, there is nothing in current plant analyses or in proposed LOCA criteria to specifically deal with this phenomenon. We hope to understand the behavior of these embrittled regions better from further integral tests that are planned for fuel rods with Zircaloy, M5, and ZIRLO cladding during FY06-08.

In summary, we have discovered, or re-discovered, five phenomena that can adversely affect the embrittlement criteria in 50.46. Four of these are related to burnup and alloy effects that have been the subject of our research and are explicitly accounted for in proposed criteria that were described in the draft RIL. Further, as long as licensees continue to (a) use Zircaloy, ZIRLO, and M5 cladding made from sponge zirconium and with polished surfaces, (b) do their LOCA analyses with the Cathcart-Pawel or Baker-Just oxidation correlations, and (c) subtract pre-transient oxidation from the 17% limit, we believe that these four phenomena will be adequately accounted for. The remainder of testing that is planned on heavily corroded Zircaloy and on irradiated ZIRLO and M5 cladding is expected to confirm this understanding

The fifth phenomenon (localized embrittlement in the balloons) is neither burnup nor alloy dependent and is not addressed by the revised criteria we have been discussing. Nevertheless, we do not think this phenomenon creates a safety issue for the reasons mentioned above, and we plan to get more information on this subject during the balance of the LOCA work that is being done at ANL.

The fifth phenomenon (localized embrittlement in the balloons) is neither burnup nor alloy dependent and is not addressed by the revised criteria we have been discussing. Nevertheless, we do not think this phenomenon creates a safety issue for the reasons mentioned above, and we plan to get more information on this subject during the balance of the LOCA work that is being done at ANL.

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