

December 15, 2000

TO: Farouk Eltawila, Director DSARE
FROM: Ralph Meyer, Sr. Technical Advisor 
SUBJECT: EMBRITTLEMENT OF Zr-1%Nb CLADDING DURING A LOCA

A week ago I told you of a German paper I just discovered that seemed to question the appropriateness of using the 17% oxidation criterion for M5 cladding. Although the results reported in that paper were obtained for the Russian VVER cladding alloy (E110), that alloy and M5 are both nominally Zr-1%Nb. These results would likewise raise questions about ZIRLO cladding, which also contains niobium (Zr-1%Nb-1%Sn nominal), although the results would not be so directly applicable. The citation and abstract of the paper I found are given below.

J Böhmert, "Embrittlement of ZrNb1 at room temperature after high-temperature oxidation in steam atmosphere," *Kerntechnik* 57 (1992) p. 55-58.

"The ductility of ZrNb1 claddings after exposure to steam between 700 and 1000 C was determined by ring compression tests at room temperature and compared with the ductility behavior of Zircaloy. The ductility of ZrNb1 is quickly reduced by steam exposure. Complete embrittlement of ZrNb1 is reached at a relative equivalent oxide-layer thickness of 5%, in contrast to Zircaloy, where a relative thickness of 17% is necessary to produce complete embrittlement. The different behavior of the two alloys is caused by higher oxygen uptake and a more homogeneous oxygen distribution in ZrNb1 after cooling. Between 950 and 1000 C, there is an additional effect of high hydrogen absorption. Validity of the 17% criterion for ZrNb1 can no longer be taken for granted in view of these experimental findings."

During the past week, I have (a) discussed this paper with our laboratory people at ANL and Penn. State, (b) contacted the author, (c) obtained comments from our colleagues at RRC-Kurchatov Institute, (d) found a more recent Hungarian paper on this subject, and (e) obtained further information from colleagues at IPSN. The main results are as follows.

- The author, Böhmert (Boehmert in later correspondence), has done no further work on this subject since 1990. However, he states that their Czech colleagues at UIP Zbratlav had similar results. I have not yet been able to reach the author of that work.
- A Hungarian paper by Griger et al. on work at AEKI was presented at the Halden meeting in 1999, and those results also support Böhmert's findings. This conclusion is not stated, nor is it obvious, in the Hungarian paper, but you can see it in Figs. 4a and 4b if you know that zero ductility corresponds to a relative deformation value for the rings between 5 and 10% (this information pointed out by Georges Hache).
- Yegorova at RRC-KI says they are familiar with Böhmert's tests and criticized them on two grounds. One was that they used direct electrical heating (not true) and the other was that RRC-KI has recent quench results from RIAR that do not show failure below 18% (not definitive as discussed below). Yegorova was not prepared to comment fully

on Böhmert's work, but the reasons tentatively given will not likely lead to a dismissal of Böhmert's results.

- Hache and Grandjean of IPSN have submitted an abstract for the coming ASTM meeting (June 2001) in which they say they have reviewed available information from Germany, Russia, and Hungary. They conclude that application of NRC's 1973 ECCS rulemaking-hearing methodology leads to a provisional criterion for a total oxidation limit of 6% for Zr-1%Nb cladding. The 6% value corresponds to a temperature of 275°F, Hobson's test temperature, rather than room temperature at which Böhmert actually did his tests.
- Hache said that Framatome and EdF say that M5 behaves differently from Russian E110, but they have not yet provided test results to prove it. Böhmert said that Siemens KWU is concerned with the problem and that he sent his working reports to Siemens some months ago. I believe this indicates that the industry knows about this situation and has not informed us.

The relation between ring-compression tests and quench tests has to be clear before one can understand the significance of ring-compression tests such as Böhmert's. In the 1973 Opinion of the Commission at the conclusion of the ECCS hearings, the Commission stated that their selection of embrittlement criteria results primarily from their belief that "retention of ductility in the Zircaloy is the best guarantee of its remaining intact during the hypothetical LOCA." They went on to say that "...and 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." Hence, the 2200°F and 17% embrittlement criteria of 10 CFR 50.46 were based on Hobson's ring-compression tests, and the quench test results were just confirmatory.

The appropriateness of this conclusion is vividly seen in recent JAERI tests on as-received and pre-hydrided Zircaloy tubes. Tanimoto's results for as-received Zircaloy show the demarcation between failure and no-failure just below 20% oxidation for restrained specimens whereas this demarcation for unrestrained specimens is around 60% oxidation. The same high degree of variability is seen as a function of restraint for pre-hydrided specimens although the oxidation levels are lower and suggest a burnup dependence. Thus it is clear that unrestrained quench-test results cited by RRC-Kurchatov Institute (above) and by Framatome in their submittals to NRC on M5 are not sufficient to demonstrate the ductility intended by 10 CFR 50.46.

The situation for ZIRLO is less clear because it contains tin as well as niobium. One might expect ZIRLO's ductility to lie between that of Zircaloy and M5, but no information on LOCA ductility was provided during the review of ZIRLO in 1991 or during the rule change in 1992 to include ZIRLO in 10 CFR 50.46 -- not even quench tests such as provided for M5.

In summary, the facts seem clear enough to question whether plants with M5 cladding have been analyzed with adequate embrittlement criteria for LOCA assessment. A similar question could be raised about plants with ZIRLO cladding. I think there is enough information here to forward to NRR so they will have an opportunity to let the licensees address these questions.