

PUBLIC SUBMISSION

As of: August 20, 2014
Received: August 20, 2014
Status: Pending_Post
Tracking No. 1jy-8dwj-jg6i
Comments Due: August 21, 2014
Submission Type: Web

Docket: NRC-2012-0041
 Issuance and Availability of Draft Regulatory Guide

Comment On: NRC-2012-0041-0002
 Draft Regulatory Guide DG-1261 - Conducting Periodic Testing for Breakaway Oxidation Behavior

Document: NRC-2012-0041-DRAFT-0008
 Comment on FR Doc # N/A

Submitter Information

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3/24/2014
79FR 16106

General Comment

Please see comments in the attached file.

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Attachments

EPRI_50 46c_Tech_comments

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RULES AND DIRECTIVES
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SUNSI Review Complete

Template = ADM - 013

E-RIDS= ADM-03

Add= *m. Flanagan (mef)*

August 20, 2014

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

Subject: RIN 3150-AH42, "Performance-Based Emergency Core Cooling Systems Cladding
Acceptance Criteria"

DG-1261, Conducting Periodic Testing for Breakaway Oxidation Behavior

DG-1262, Testing for Post Quench Ductility

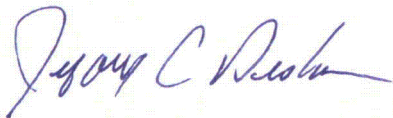
DG-1263, Establishing Analytical Limits for Zirconium-based Alloy Cladding

The Electric Power Research Institute (EPRI) is pleased to provide comments on the draft regulatory guides as published in the Federal Register on March 24, 2014.

EPRI was an active participant in the NRC-sponsored loss of coolant accident (LOCA) research program at the Argonne National Laboratory and also conducted extensive independent and complementary research in the area. Most of the feedback provided in this submittal is based on direct research experience, particularly issues identified in an EPRI coordinated international LOCA round robin test program. Key test parameters of the round robin effort were taken from the NRC draft regulatory guides.

Our comments on the draft regulatory guides are contained in Attachment A. Should you have questions related to these comments, please contact Dr. Ken Yueh at 704-595-2613 or kyueh@epri.com.

Sincerely,



Jeffrey C. Deshon
Senior Program Manager
Fuel Reliability Program

c: Mr. Neil Wilmshurst
Ms. Christine King
Dr. Erik Mader
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Attachment A

Comment 1

Reference	DG-1261, CONDUCTING PERIODIC TESTING FOR BREAKAWAY OXIDATION BEHAVIOR , (ADAMS Accession No. ML12284A324)
Statement	Page 6: "To confirm that slight composition changes or manufacturing changes have not inadvertently altered the cladding's susceptibility to breakaway oxidation, 10 CFR 50.46c calls for periodic measurement of the onset of breakaway oxidation."
Comment	Fuel cladding fabricated from non electrolytic process source materials show consistent and stable time to breakaway oxidation. These observations do not support a concern that slight changes in the composition and manufacturing process could impact the incubation time to breakaway oxidation. Therefore, periodic testing could be removed from 50.46c and could be captured in the manufacturing qualification process.
Rationale	<p>The bulk of the breakaway oxidation test database, which encompasses fuel cladding manufactured by different vendors/processes and spans a wide range of chemical compositions, do not show short breakaway incubation time behavior. Therefore, aside from a single occurrence of short breakaway oxidation (described below), the bulk of data do not support a need for periodic testing.</p> <p>The only instance of observed significant reduction in the incubation time to breakaway oxidation is the Russian E110 cladding manufactured from electrolytic sourced material. Subsequent testing of the alloy fabricated from non electrolytic source material, using identical tubing fabrication process, showed expected incubation time to breakaway oxidation [1]. Detailed chemical analyses of the electrolytic and non-electrolytic cladding showed significantly higher fluorine content in the electrolytic processed cladding. This is expected since fluorine containing compounds are widely used in the Russian electrolytic process as a processing material. The effect of fluorine on reducing the breakaway incubation time from surface etching is well documented in the Argonne National Laboratory (ANL) research program [2].</p>

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Comment 2

Reference	DG-1261, CONDUCTING PERIODIC TESTING FOR BREAKAWAY OXIDATION BEHAVIOR , (ADAMS Accession No. ML12284A324)
Statement	Page A-11: Weight-Gain Benchmarks: "After thermal benchmarking, samples should be tested without TCs welded onto the sample to determine the weight gain. These tests should be conducted at 800 °C and 1,000 °C. The test times should be less than those that result in breakaway oxidation. For 1,000 °C, an isothermal test time of Appendix A to DG-1261, Page A-12 2,000 s is recommended. For Zircaloy-2 (Zry-2), Zry-4, and ZIRLO™ alloys oxidized at 1,000 °C for $\leq 2,000$ s, the measured weight gain (normalized to the surface area exposed to steam) was in good agreement with the Cathcart-Pawel (CP) correlation predictions (Ref. 3). If the measured weight gain differs from the CP-predicted weight gain by $\geq 10\%$, then data-generating testing should not be initiated until the discrepancy is resolved."
Comment	The Cathcart-Pawel (CP) correlation, as the name suggests, is an empirical correlation developed based on testing of Zircaloy-4 and has an applicable temperature range [3]. The use of the correlation as proposed is at the edge or outside the valid range of applicability. One way to address this issue would be to use a vendor generated alloy-specific database for thermal benchmark as a generic requirement. This could then be applied to existing and future alloys.
Rationale	The CP correlation calculated weight gain is known to deviate (over predict) at lower temperatures. The measured weight gain is also alloy-dependent. Experimental data generated by the ANL [2] and the industry coordinated LOCA round robin [4] showed the measured weight gain could deviate more than 10% from the CP calculated under the condition outlined in the draft regulatory guide.

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Comment 3

Reference	DG-1262, TESTING FOR POSTQUENCH DUCTILITY , (ADAMS Accession No. ML12284A325)
Statement	DG-1262, Page 6: "...support the development of analytical limits at peak oxidation temperatures less than 2,200 °F (1,204 °C)."
Comment	The test procedure should be applicable above 2200°F. One way to ensure adequacy of the procedure is to specify a check to ensure no steam starvation at higher temperatures in lieu of the temperature limitation.
Rationale	The document describes procedures used to determine the post quench ductility. The 2200°F is only an input parameter. The procedure should be valid for a higher temperature as long as there is sufficient steam flow. The bases for this statement derives from testing conducted under a wide range of steam flow conditions in the post-quench ductility round robin test program [4]. The different steam flow rate conditions utilized by the participating laboratories produced consistent results, indicating that significant margin to steam starvation still exists. Higher temperature test results using similar test procedures were presented at the 1973 rulemaking [5].

Attachment A

Comment 4

Reference	DG-1263, Establishing Analytical Limits for Zirconium-Based Alloy Cladding, USNRC Draft Regulatory Guide, (ADAMS Accession No. ML12284A323)																																																
Statement	Page 8: “Irradiated cladding material can be used to demonstrate that a cladding alloy’s embrittlement behavior is accurately characterized by using prehydrided material. To demonstrate this, an acceptable approach would be to determine the ductile-to-brittle transition for irradiated material with hydrogen contents within 50 wppm of the anticipated maximum hydrogen content and within 50 wppm of half of the anticipated maximum hydrogen content.”																																																
Comment	<p>Use of test specimens with hydrogen pre-charge can provide a suitable surrogate for irradiated materials, obviating the need for the hydrogen concentration to be within 50 ppm of irradiated data.</p> <p>Moreover, if testing of irradiated cladding for significant compositional changes is elected, testing to compare hydrogen pre-charge data can be reduced to one irradiation exposure near end-of-life condition.</p>																																																
Rationale	<p>The NRC test results plotted below showed no difference in behavior between irradiated and non-irradiated materials for multiple alloys made by different fuel vendors. The test data support the use of hydrogen pre-charge as a surrogate for irradiation.</p> <div><p>Legend:</p><ul style="list-style-type: none">■ Zirc-4; 800°C Quench, Non-Irradiated× Zirc-2; 800°C Quench, Non-Irradiated+ ZIRLO - 800°C Quench, Non-Irradiated× M5 - 800°C Quench, Non-Irradiated◆ M5 - 800°C Quench, Irradiated● ZIRLO - 800°C Quench, Irradiated■ Zirc-4 - 800°C Quench, Irradiated— Pol. Fit<table border="1"><caption>Approximate data points from the graph</caption><thead><tr><th>Hydrogen Content (wppm)</th><th>Embrittlement CP-ECR (%)</th><th>Material/Condition</th></tr></thead><tbody><tr><td>0</td><td>20</td><td>Zirc-2; 800°C Quench, Non-Irradiated</td></tr><tr><td>10</td><td>18</td><td>Zirc-4; 800°C Quench, Non-Irradiated</td></tr><tr><td>20</td><td>16</td><td>Zirc-4; 800°C Quench, Non-Irradiated</td></tr><tr><td>110</td><td>15</td><td>M5 - 800°C Quench, Irradiated</td></tr><tr><td>200</td><td>11</td><td>ZIRLO - 800°C Quench, Irradiated</td></tr><tr><td>250</td><td>10</td><td>M5 - 800°C Quench, Irradiated</td></tr><tr><td>300</td><td>9</td><td>Zirc-4 - 800°C Quench, Irradiated</td></tr><tr><td>300</td><td>10</td><td>Zirc-4; 800°C Quench, Non-Irradiated</td></tr><tr><td>380</td><td>8</td><td>Zirc-4; 800°C Quench, Non-Irradiated</td></tr><tr><td>450</td><td>6</td><td>Zirc-4; 800°C Quench, Non-Irradiated</td></tr><tr><td>480</td><td>6</td><td>Zirc-4; 800°C Quench, Non-Irradiated</td></tr><tr><td>550</td><td>5</td><td>Zirc-4; 800°C Quench, Non-Irradiated</td></tr><tr><td>550</td><td>5</td><td>Zirc-4 - 800°C Quench, Irradiated</td></tr><tr><td>600</td><td>4</td><td>Zirc-4 - 800°C Quench, Irradiated</td></tr><tr><td>600</td><td>5</td><td>Zirc-4; 800°C Quench, Non-Irradiated</td></tr></tbody></table></div> <p>Numerous mechanical tests have shown irradiation damages are annealed out at LOCA temperatures (> 700°C).</p> <p>Irradiation will have a small impact on the chemical makeup of zirconium based alloys, which are typically greater than 97% zirconium. A very small fraction of the alloying elements may transmute into other elements. Testing of different irradiated alloys in the ANL program showed transmutation of the common alloying elements in commercial service today to have no effect on the cladding ductility degradation and therefore additional testing of irradiated cladding of variations of these alloys is not needed. If the alloy composition changes significantly, such as the addition of new alloying elements, the transmutation effect could still be captured by the addition of expected transmutation products (non radioactive isotope) into the alloy melt for evaluation.</p>	Hydrogen Content (wppm)	Embrittlement CP-ECR (%)	Material/Condition	0	20	Zirc-2; 800°C Quench, Non-Irradiated	10	18	Zirc-4; 800°C Quench, Non-Irradiated	20	16	Zirc-4; 800°C Quench, Non-Irradiated	110	15	M5 - 800°C Quench, Irradiated	200	11	ZIRLO - 800°C Quench, Irradiated	250	10	M5 - 800°C Quench, Irradiated	300	9	Zirc-4 - 800°C Quench, Irradiated	300	10	Zirc-4; 800°C Quench, Non-Irradiated	380	8	Zirc-4; 800°C Quench, Non-Irradiated	450	6	Zirc-4; 800°C Quench, Non-Irradiated	480	6	Zirc-4; 800°C Quench, Non-Irradiated	550	5	Zirc-4; 800°C Quench, Non-Irradiated	550	5	Zirc-4 - 800°C Quench, Irradiated	600	4	Zirc-4 - 800°C Quench, Irradiated	600	5	Zirc-4; 800°C Quench, Non-Irradiated
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Attachment A

	<p>If demonstration via irradiated test material is desired, it is not necessary to test cladding at multiple points in life since irradiation effect in this regard is cumulative; in other words, it is easier to detect potential irradiation effects near end of life. In practice, especially under the current requirement of operating lead test assemblies (LTAs) under “non-limiting” locations, it may not be possible to produce irradiated test samples that could bound the operation of the fuel design.</p>
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Comment 5

Reference	DG-1263, Establishing Analytical Limits for Zirconium-Based Alloy Cladding, USNRC Draft Regulatory Guide, (ADAMS Accession No. ML12284A323)
Statement	Page 5: "This analytical limit is acceptable for the zirconium-alloy cladding materials tested in the NRC's LOCA research program, which were Zry-2, Zry-4, ZIRLO™, and M5™. This analytical limit is based on the data obtained in the NRC's LOCA research program."
Comment	The analytical limit should be valid for other zirconium-based alloys. If the composition of a new zirconium-based material falls within the combined experience base of existing alloys tested in the ANL program, the analytical limit should be applicable. The actual boundary could be defined by the solubility of alloying elements in the beta phase.
Rationale	<p>The NRC research program has concluded that the cladding ductility degradation is caused by oxygen diffusion into the prior beta phase. The ANL test data showed the oxygen diffusion is not affected by the alloying elements currently in commercial service. This is consistent with the physical understanding that the diffusion rate of a species in a material is controlled by the material's matrix composition. For zirconium-based alloys, the matrix is greater than 97% zirconium. The solubility of the common alloying elements in the zirconium matrix is high and could not form an intermediate compound that could short circuit the diffusion process.</p> <p>Research Information Letter (RIL) 0801, "Technical Basis for Revision of Embrittlement Criteria in 10 CFR 50.46," concluded: "The trends seen here should apply to all present and future zirconium-alloy cladding materials because no dependence on specific alloy composition has been found."</p>

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Comment 6

Reference	DG-1263, Establishing Analytical Limits for Zirconium-Based Alloy Cladding, USNRC Draft Regulatory Guide, (ADAMS Accession No. ML12284A323)
Statement	Page 11: "Because NUREG/IA-0211, "Experimental Study of Embrittlement of Zr-1%Nb VVER Cladding under LOCA-Relevant Conditions," issued March 2005 (Ref. 4), did not present hydrogen-accumulation data for temperatures between 650 °C and 800 °C, there is no basis for not including time spent at temperatures >650 °C in establishing the analytical limit for transient time."
Comment	Existing data suggest that accounting for times below 800°C is not warranted and the lower temperature requirement could be removed from the regulatory guide.
Rationale	<p>Results presented by Leistikow and Schanz [6,7] indicate two minima times to breakaway oxidation at around 800°C and 1000°C. The results also indicate the incubation time to breakaway is longer at 800°C than 1000°C and increases below 800°C. It is between these two temperatures that the margin to breakaway time is smallest. With the breakaway time established at 1000°C, extending the range to temperatures below 800°C does not offer additional protection.</p> <p>This is also supported by breakaway oxidation testing conducted as part of a round robin effort, which showed that the breakaway time at 800°C is greater than 7000 seconds, compared to 5000 seconds at 1000°C [4]. The round robin effort scope limited the exposure time to a maximum of 7000 seconds for both 800°C and 1000°C tests, but one laboratory carried the experiment to 9,000 seconds without detecting breakaway oxidation for the 800°C test. In another experiment, test samples were pre-oxidized to 20 µm of oxide at 800°C. The pre-oxidized samples were exposed for 18,000 seconds without detecting breakaway oxidation [8].</p>

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Comment 7

Reference	DG-1263, Establishing Analytical Limits for Zirconium-Based Alloy Cladding, USNRC Draft Regulatory Guide, (ADAMS Accession No. ML12284A323)
Statement	<p>Page 12: Accounting for Uncertainty and Variability in Hydrogen Content</p> <p>“The uncertainty in the model can be characterized and quantified by supporting the model with post-irradiation examination that includes values for multiple burnup levels, encompasses all applicable operating conditions and reactor coolant chemistry, and quantifies axial, radial and circumferential variability.</p> <p>To apply the analytical limit in Figure 2 to an individual fuel rod (or fuel rod grouping), the allowable CP-ECR should be based on predicted peak circumferential average hydrogen content for the individual rod (or fuel rod grouping).”</p>
Comment	<p>Existing data support the use of the nominal hydrogen concentration at an axial position along the length of the fuel rod as a sufficient approach for determining the analytical limit for cladding embrittlement. With circumferential and radial hydride uncertainty inherently captured in the ANL test data, it is sufficient to use the axial hydrogen variability for determining the analytical limit for cladding embrittlement based on local hydrogen concentration.</p>
Rationale	<p>The proposed regulation requires the quantification of the uncertainty in the hydrogen uptake model. Post-test examination of ANL ring compressing test samples showed significant circumferential hydrogen concentration variation, thus the circumferential uncertainty is implicitly captured in the ANL test. Therefore inclusion of additional uncertainty to bound the variability would introduce unwarranted conservatism in the application of the analytical limit curve. Also, post-irradiation data are typically selected to cover limiting operating conditions.</p>

Attachment A

Comment 8

Reference	DG-1263, Establishing Analytical Limits for Zirconium-Based Alloy Cladding, USNRC Draft Regulatory Guide, (ADAMS Accession No. ML12284A323)
Statement	Appendix B, "Overview of Acceptable Test Matrices," to this regulatory guide provides a high-level overview of an acceptable test matrix. The test matrix overview is intended to provide a clear picture of the range of material and test conditions that could be used to establish an alloy-specific limit other than the analytical limit in Figure 2. It is intended to complement the test matrix guidance in DG-1262.
Comment	<p>The acceptable test matrix posits conditions and sample characteristics that may not be technically possible or adaptive to individual situations. Incorporating flexibility into the test matrix could be accomplished in various ways, including:</p> <ol style="list-style-type: none">1. At each hydrogen test level, allow a window of hydrogen concentrations to be tested. This will require some interpolation between test samples of slightly different hydrogen concentration. For example interpolation between test samples (a) ductile at X-10% ppm, and (b) brittle at X+10% ppm.2. Allow testing at different hydrogen concentrations and use a best-fit method to determine the ductile-to-brittle transition.
Rationale	<p>The test matrix assumes test samples can be precisely pre-charged to the desired target hydrogen levels. This level of precision/accuracy is not achievable with current hydrogen charging techniques. The inability to precisely pre-charge test samples with hydrogen makes it very difficult to follow the prescribed test procedure. Some flexibility is needed to determine the analytical limit.</p> <p>The "acceptable" test matrix prescribes testing at several hydrogen concentrations which may not be applicable for alloys with low hydrogen pickup, such as M5TM.</p>

References

- 1 Perez-Fero et al, "High Temperature Behavior of E110G and E110 Cladding in Various Mixtures of Steam and Air", Proceedings of WRFPM 2014, Sendai, Japan
- 2 Billone et al, "Cladding Embrittlement During Postulated Loss-of-Coolant Accidents", NUREG/CR6967
- 3 NUREG 17, Zirconium Metal-Water Oxidation Kinetics IV. Reaction Rate Studies
- 4 Yueh et al, "Loss of Coolant Accident Testing Round Robin", Proceeding of the 2013 LWR Fuel Performance Meeting, Charlotte, NC, 2013
- 5 NRC Docket RM-50, April 16, 1973
- 6 S. Leistikow and G. Schanz, "The Oxidation Behavior of Zircaloy-4 in Steam between 600 and 1600°C," *Werkstoffe und Korrosion* 36 (1985) 105-116.
- 7 S. Leistikow and G. Schanz, "Oxidation Kinetics and Related Phenomena of Zircaloy-4 Fuel Cladding Exposed to High Temperature Steam and Hydrogen-Steam Mixtures under PWR Accident Conditions," *Nucl. Eng. and Des.* 103 (1987) 65-84.
- 8 Yueh et al, "Changes in Cladding Properties under LOCA Conditions", Proceeding of the 2013 LWR Fuel Performance Meeting, Charlotte, NC, 2013.