Response To NRC Staff Review of NEI/EPRI Reports On Creep and Fracture Toughness of Zircaloy SNF Cladding

NRC/NEI Meeting

April 18, 2001

Main - no

SCC Mechanism

- Role of Chemistry - No Review Comments
  - Implication: Staff Agrees That SCC Cannot Occur Without Presence of Corrosive Species?
  - Lack of Stress Environment is an Added Support
- Role of Stress and Time-At-Stress
  - ***Issue 1***: Without Debating the Issue of Crack Initiation by Other Means, the Report Addresses Pre-Existing Cracks Using FM Analysis Applying Compounded Conservatism
  - ***Issue 2***: Prevention of Crack Initiation Not Crack Propagation is the Issue. If a Crack is Initiated by SCC, Failure is Almost Assured
  - ***Issue 3:*** Again, Crack Initiation is the Issue, and Once a Crack is Initiated, How it Propagates Becomes Irrelevant
  - ***Issue 4:*** This is Reflected in the Data and is Discussed in the Report. See Reference to Cox's Work (41)

SCC Mechanism

- Role of Stress and Time-At-Stress (cont.)
  - ***Issue 5***: This is Compounding the Effects of Pre-Cracking. The Referenced Figure Clearly Shows 200 MPa Threshold Stress for Pre-Cracked Specimens.
  - ***Conclusions:***
    - **(1) The Issues Raised in the Review Are Shown to Have Already Been Adequately Covered in the Report with Supporting Literature**
    - **(2) The Staff Review Ignores the Fact That the Corrosive Environment Needed to Initiate SCC is Not Present In Spent Fuel**
    - **(3) The Staff Concludes that the Report's Summary 2.6(b) Has Not Been adequately Supported, yet It Curiously States that "*literature information indicate that SCC...is unlikely....*" If the Staff already know that SCC is unlikely, why take issue with the Report's literature-based arguments? Also, we are curious to know the literature being referred to by the Staff!**

SCC Mechanism

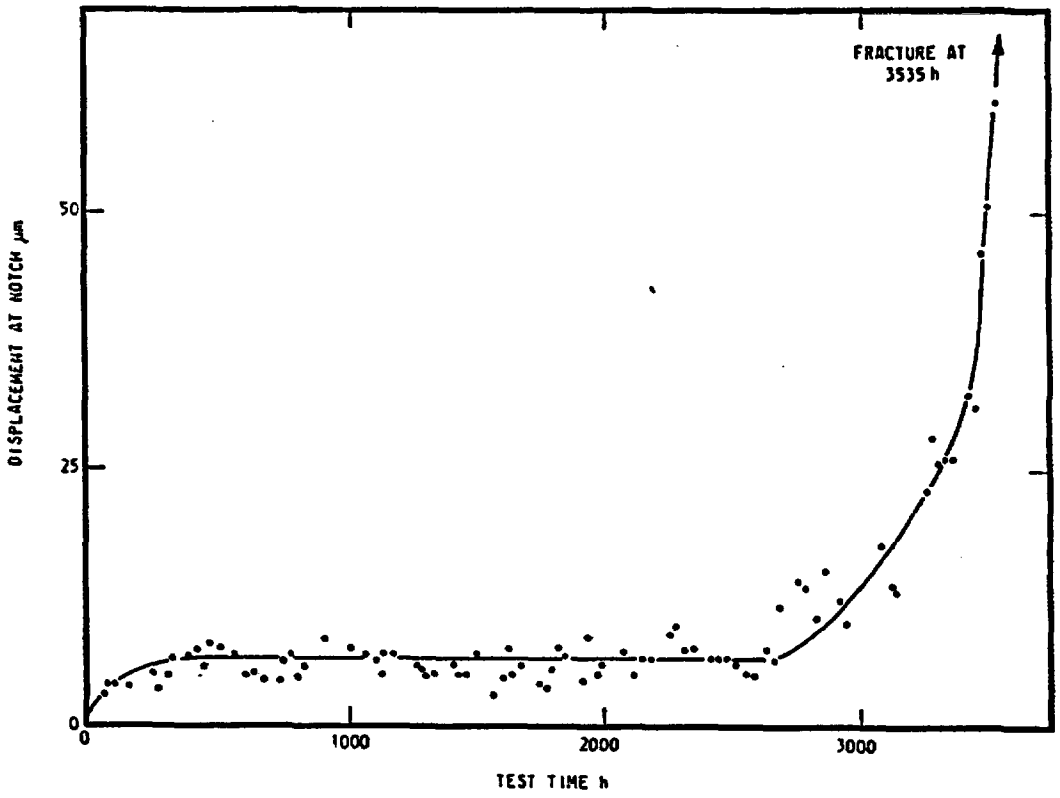
- Effect of Irradiation/High Burnup
  - ***Issue 1:*** Reference to Dissolution of Hydrides as a Condition of Dry Storage Claimed in the Report is Incorrect. The Report Simply Refers to Cox's Experiment
  - ***Issue 2:*** There is no Circular Calculations Here. The Calculations are as Follows: $K_I = 1.12\ \sigma\ (\pi a)^{1/2}$, where $\sigma = 150$ MPa, $a = 73\mu m$. This Gives $K_I = 2.54$ MPa $m^{1/2}$ as Stated in the Report
  - ***Conclusions:***
    - (1) The Above Stated "Issues" are Shown to be Non-Issues
    - (2) Consequently, The Staff Conclusions Regarding 2.6 (c) are not Valid
    - (3) Conclusion (3) From the Previous Viewgraph Applies Here Also.

DHC Mechanism

- Evolution of DHC Mechanism
  - ***Issue 1:*** Figure 7 Indeed shows the Intermittent Crack Advance. Each dot in the Figure Represents a Sonic Signal. This is the Fundamental Behavior of DHC
  - ***Issue 2:*** (a) The K_{IC} - CSED is a Correlation Similar to Many Other Correlations Found in Fracture Mechanics, e.g. K_{IC} - CVN Correlation of Barsum and Rolfe, and Paris's Tearing Modulus,... It is no more and no less valid than any Empirical Correlation.

    (b) Data Scatter is a Fact-of-Life. That is Why we Rely on Correlations as Engineering Tools.

    (c) Not Only There Is Not Sufficient Fracture Toughness Data for High Burnup Cladding, There is no Accepted Standard for the Fracture Testing of Cladding Geometry. This is the Main Motivation for Developing the K_{IC} - CSED Correlation.



Displacement-time curve for round notched bar of Zircaloy-2 containing 100 ppm hydrogen and tested at 350 K with K_I of 15 MPa $\sqrt{m}$.

Figure 3-3. Intermittent DHC Crack Evolution (Ref. 48)

DHC Mechanism

- Re-Orientation of Hydrides
  - ***Issue 1:*** This May Shorten Failure Time, But the Key Is to Preclude DHC Altogether, Which is the Theme of the Report
  - ***Issue 2:*** DHC Evolves by The Hydrogen-in-Solution Diffusing Up the Stress Gradient to the Crack Tip Which is in a State of Triaxial (At Least Biaxial) Tension
  - ***Issue 3:*** This Repeats What is Already Discussed at Length in the Report. The Report States That Hydrides Re-Orientation Can Occur and It Describes the Conditions Under Which It Occurs in Dry Storage. It Simply Says that the Experimental Evidence is Against Massive Re-orientation.
  - ***Issue 4:*** DHC Can Occur at Any Hydrogen Level. The Report Makes No Claim Regarding Hydrogen Concentration and Temperature. It Simply Quotes Experimental Results. How Can Such Information be Raised as an ISSUE?

DHC Mechanism

- ***Conclusions:***
  - (1) The Report Clearly States that the Thermal History in Dry Storage is Troublesome with Respect to DHC. It Clearly Points Out Under What Conditions DHC Can Occur.
  - (2) The Report Made No Claims That Hydride Re-Orientation Could Not Occur.
  - (3) The Staff Review Totally Ignored the Role of the Stress in DHC, and Made no Reference to the Report's Claim That the Main Line of Defense Against DHC is K_{IH} for Stage-I DHC and K_I.for Stage-II DHC. The 73 μm Crack Size Determined From SCC Consideration and The Maximum Hydride Length of 100 μm were Used in the Discussion of the Role of The Stress on DHC, Not The K_{IC} - CSED Correlation. In Storage, the Stress Intensity is a Factor of 2 Below the DHC Initiation Threshold and a Factor of 4 Below the DHC Crack Propagation Threshold.

Creep Data Of Zircaloy Cladding

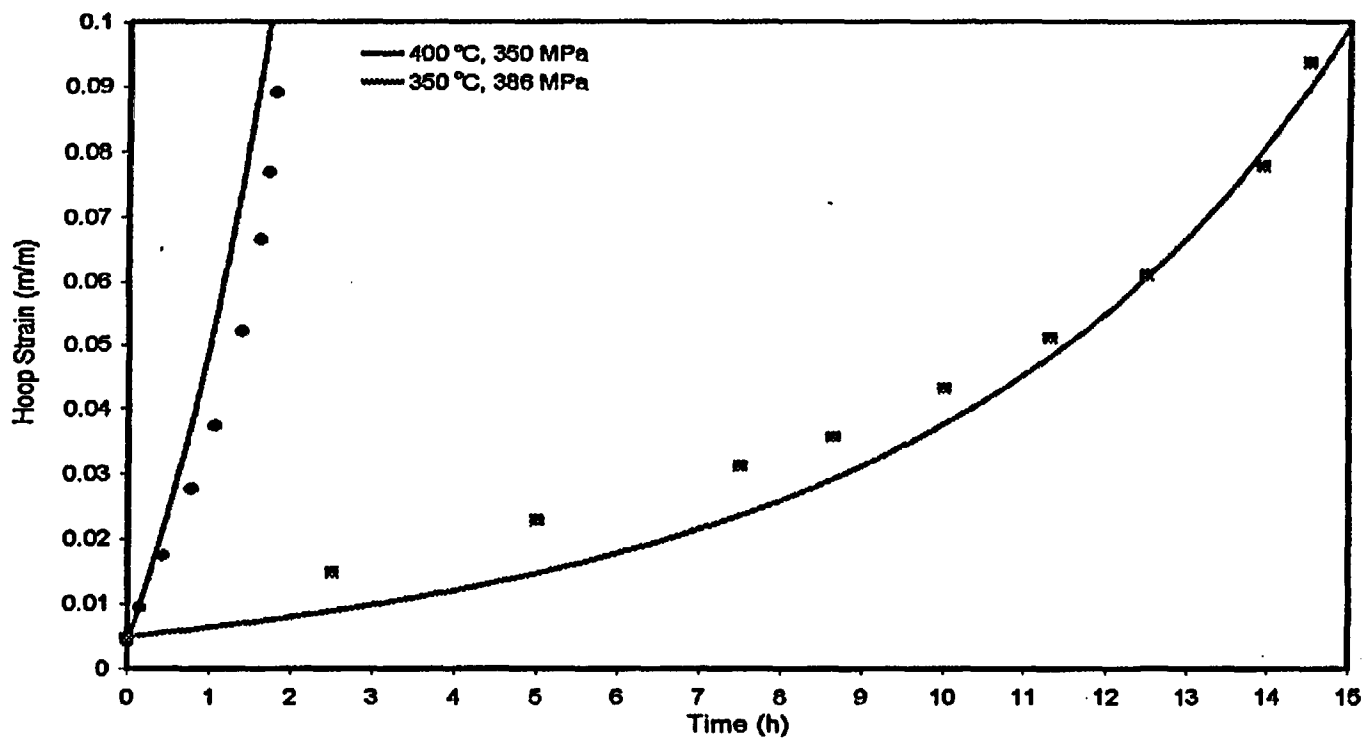
- Goll et al Data
  - ***Issue 1:*** This Test was Intended to Check the Possible Occurrence of DHC As a Result of Hydride Precipitation Radially Following an Accelerated Creep Test.
  - ***Issue 2:*** The Relevance of This Comment to the Issue at Hand Needs to be Checked Further.
  - ***Issue 3:*** The Goal is to Seek Reasonable Assurance Rather Than Full Assurance Which is an Impossibility

Creep Data Of Zircaloy Cladding

- Acceptance Criterion - Creep Based Limit
  - ***Issue 1:*** This Requires High Stresses, in the Instantaneous Plasticity Regime. Are There Any Data for DCCG at Stress Levels Below, say, 30% of Yield Stress?
  - ***Issue 2:*** The 1.7% is a Measurement Not a Material Limit of Any Kind. Einziger's Data of 7% Without Failure Supports The 2% Limit
  - ***Issue 3:*** This is the Whole Misunderstanding. The Strain Limits quoted are In the Instantaneous Plasticity Regime Where the Stress is at Ultimate Tensile Strength. Elementary Mechanics Dictate That Plastic Instability Cannot Occur at Stress Levels Below The Ultimate Strength. This is Clearly Shown by Recent French Creep Rupture Tests as Shown in the Following Figures.
  - ***Issue 4:*** Strain Rate Has a Well Known Effect on Mechanical Response. NRC's MATPRO Data Base Shows This Clearly

Analysis of Creep Rupture Tests

FALCON Prediction of CEA/EDF Uniaxial and Biaxial Creep Rupture



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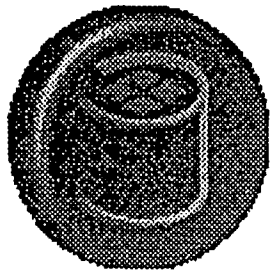
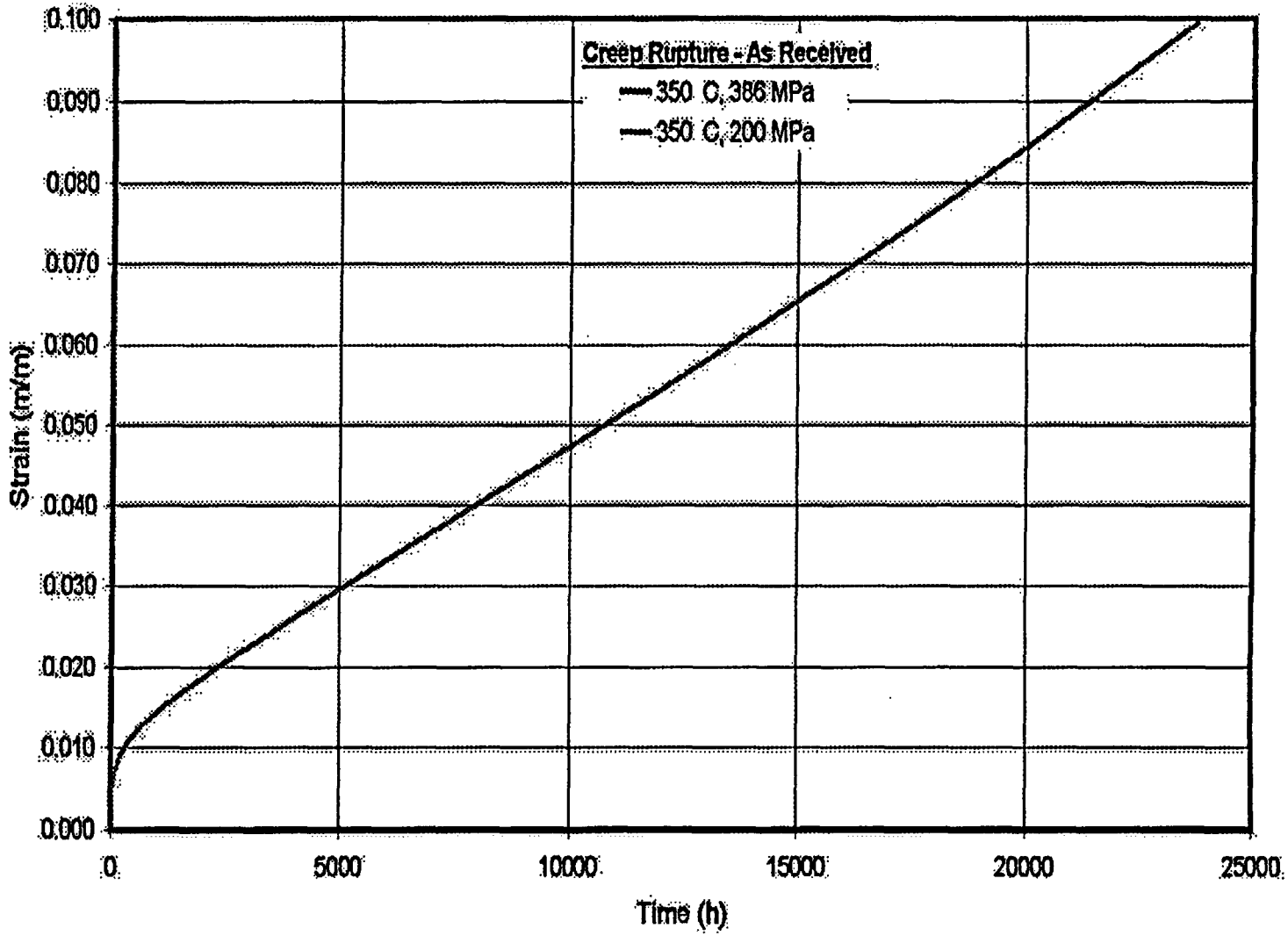
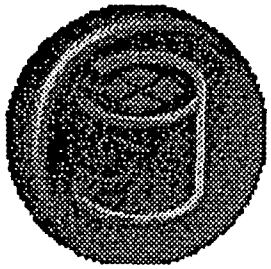
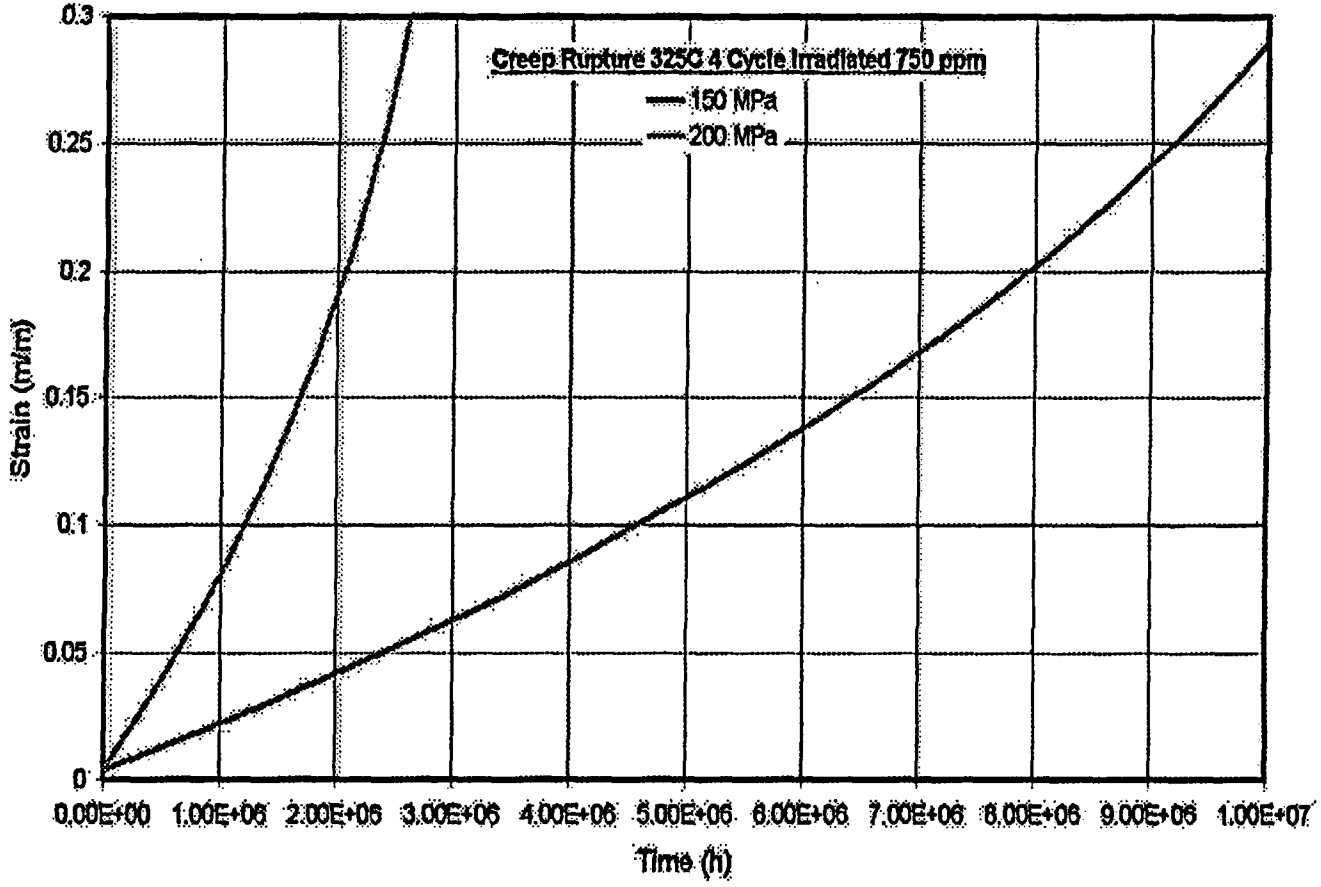




Creep Response for 200 MPa Initial stress Showing no sign of Tertiary Behavior at10% Strain, Which is the Rupture Strain for the 386-MPa Case





FALCON Analysis Simulating Creep Rupture Behavior In Dry Storage

Creep Rupture - Relation to Acceptance Criterion

- Tertiary Creep is Initiated When the Stress Reaches the Yield Strength
- ***Strain-Range Partitioning***
  - Strains Accumulated While the Stress is still in the Elastic Regime are Far Less Damaging Than Strains During Tertiary Creep
  - Concept can be Demonstrated by Conducting Post-Creep Tension Tests At Various Creep Strain Levels
  - The 2% Limit can be Validated by Such Tests

Conclusions From Creep Rupture Tests

- Uniform & Failure Strains Measured in Accelerated Creep Tests are <u>Incorrect</u> Indicators of Cladding Creep Capacity
  - Tests are Initiated in Tertiary Creep
  - Cladding Fails at its Ultimate Strength
  - In Conventional Creep Tests, Tertiary Creep is Approached From Below, and Takes Decades for Stress to Reach Yield

$$\sigma_{yield} = \sigma_{\theta 0} / \left(2e^{-\varepsilon_{\theta\theta}} - 1\right)$$

Creep Data Of Zircaloy Cladding

- Acceptance Criterion - Creep Based Limit
  - ***Issue 5:*** The Goll Data is From Creep Rupture Tests Initiated at or Near the Yield Strength. Therefore the Measured Strains Belong to the Instantaneous Plasticity Regime. Again, the Stress Should Be Invoked Here. See Figures
  - ***Issue 6:*** Nothing Can be Worse Than the Data from the EPRI Hot Cell Program. Garde's Measurements Show 3% Plastic Strain in the Ligament Beneath the Hydride Lens.
  - ***Issue 7:*** All Fracture Mechanics Calculations are Based on Idealized Flaws. This is the Accepted Practice.
  - ***Conclusions:*** We Believe The Issues Can Be Resolved Using Well Known and Accepted Mechanics Principles. The Above Explanations are Aimed in That Direction

Creep Data Of Zircaloy Cladding

- Pin-Hole Equivalent Failur Mode
  - ***Issue 1:*** One Pin-hole is Enough to Depressurize the Rod. The Probability of Having a String of Pin-holes Occurring at Exactly the Same Time is Nearly Zero.
  - ***Issue 2:*** It Seems That a Dynamic Event is Implied Here. How?
  - ***Issue 3:*** This was Covered in a Previous Comment
- ***Conclusions:***

  (1) A Strain Limit of 1% is Not Sufficient In Itself as a Criterion. It Was Based on Plastic Instability Considerations, Therefore Requiring Definition of the Stress State.

Creep Data Of Zircaloy Cladding

- ***Conclusions:***

  (1) An Acceptance Criterion as Restrictive as the 1% Strain Limit is Not Sufficient In And By Itself. It Was Based on Plastic Instability Considerations, Therefore, Requiring Definition of the Stress State.

  (2) The Staff Reviewers Dismissed, Without Mention, the Analysis of Strain Localization and the Interpretation of The Measurements on Which the Strain Limit is Based?

  (3) The Larson-Miller Parameter is a Stress-Time-To-Failure Criterion, Which is Totally Empirical. Is it Being Proposed as A Replacement to a Strain-Based Criterion?

Fracture Toughness Data

- CSED Approach - Issues Related To Cladding
  - ***Issue 1:*** Good Question, The Report Should Have Shown It on the Figure. It is the Radius of the Plastic Zone Surrounding the Crack Tip Assuming Elastic/Perfectly Plastic Material.
  - ***Issue 2:*** Equation (8) is Correct. The Total Elongation in the First Term is the Elastic+Plastic
  - ***Issue 3:*** Elastic/Perfectly Shape of the Stress-Strain Curve is a Good Approximation for Highly Irradiated Zircaloy
  - ***Issue 4:*** We Agree! Stress-Strain Curves By Themselves Do Not Provide "Fracture Mechanics" Information. However, The Area Under the Curve Represents the Energy Capacity of the Material, and as Part of a Correlation, They are Shown to Give Fracture Information.

Fracture Toughness Data

- CSED Approach - Issues Related To Cladding (cont.)
  - ***Issue 5:*** Equation 9 is Correct. It is Obtained From Equation 8 by Direct Substitution of Parameters
  - ***Issue 6:*** The Strain and Stress Measures in the CSED are the True Strain and True Stress in the Uniaxial Case, and are the True Tensorial Quantities in the Multiaxial Case.
  - ***Issue 7:*** Equation 13 Has Balanced Units If we Remember that the 3.5 Factor Came From ρ, r and E, Where ρ Has the Units of m, E Has Units of MPa and r is unitless.

$$\sigma \rightarrow \sigma_y \ , \ \rho \rightarrow \rho_y \ , \ K_I \rightarrow K_{IC}$$

Equation (6) can then be rewritten as follows:

$$K_{IC} = \sigma_y \sqrt{2\pi\rho_y} \tag{7}$$

Now we introduce the first of two postulates, namely, to assume instead that the point of instability occurs, not when $\sigma = \sigma_y$, but rather when the elastic strain energy density associated with the LEFM singularity solution reaches the critical value U_c in the neighborhood of the crack tip. As is usual in fracture mechanics, justification for this alternative assumption must come from data. (Note that for elastic-perfectly-plastic material, the above two instability assumptions are equivalent, because for such material the failure condition is only a function of stress regardless of the magnitude of the strain, whereas for strain hardening materials, the stress is not sufficient by itself to describe a limiting condition). Equating the critical strain energy density of the LEFM material to the critical strain energy density of the actual material, we obtain:

$$U_c = \sigma^2/(2E) = \sigma_y \varepsilon_{TE} - \sigma_y \varepsilon_y/2, \tag{8}$$

where σ_y, E, ε_{TE}, ε_y are, respectively, the yield stress, the elastic modulus, the total elongation (elastic + plastic), and the yield strain. The left-hand side of Eq. (8) reflects the presence of crack-tip singularity and the resulting pseudo-elastic stress σ, whereas the right-hand side describes material with distributed damage reflected in the total elongation and the other measured properties. Substituting the quantity $r = \varepsilon_{TE}/\varepsilon_y$, which is a measure of the material's ductility in Eq. (8), we obtain:

$$\sigma_y^2 = 2EU_c/(2r-1) \tag{9}$$

Substituting Eq. (9) in Eq. (6) gives:

$$K_{IC}^2 = 4\pi EU_c \rho_y/(2r-1) \tag{10}$$

For a highly confined crack tip, which is the plane-strain condition imposed on fracture toughness testing, ρ_y is of the order of 10 microns. However, under plane-stress condition, and under conditions of ductile fracture, ρ_y can be an order of magnitude larger. The parameters ρ_y and r play the same physical role in characterizing the level of ductility of the material, and they both increase proportionately with increasing ductility. This suggests a second postulate, namely, that the changes in ρ_y and r occur in such a way that the quantity

$$\rho_y/(2r-1) = 10\times10^{-6} \tag{11}$$

plays the role of a material constant. For example, at an r-value of unity, indicative of totally brittle material, ρ_y has the minimum value of 10 microns, whereas for ductile fracture where r can grow to a value of 10 or greater, the plastic zone size can be of the order of 150-200 microns. Rolfe and Barsum [18] estimated ρ_y to be about 50 times smaller than the plane-strain specimen

thickness B defined in Eq. (2). Using a K_{IC} estimate of 20 MPa$\sqrt{m}$ presented earlier for cladding with relatively high hydrogen content, and 700 MPa as a typical high-burnup yield strength, we calculate an estimate of 40 μm for ρ_y. This gives r = 2.5 as the ductility ratio and a total elongation (elastic + plastic) of about 1.8%, which is in the conservative range of the data for high burnup cladding with relatively high hydrogen content. Upon substituting $\rho_y = 10 \times 10^{-6}$ m in eq. (10), we obtain the following simple correlation:

$$K_{IC} = 0.01121\sqrt{E}(U_c)^{1/2} \tag{12}$$

Substituting a typical high burnup value for E of 96,000 MPa in Eq. (12), we obtain:

$$K_{IC} = 3.5\sqrt{U_c} \tag{13}$$

as an approximate upper bound for highly irradiated Zircaloy. In application, the appropriate value of E should be used if available. Otherwise, Eq. (13) is recommended. Comparison of Eqs. (12) and (13) to data for aluminum alloys is shown in Figure 2, which shows excellent agreement. Clearly, there are much more data that can be added to Figure 2.

As discussed in detail above, expressions (12) and (13) make use of two physically-based postulates. The first, which is expressed in Eq. (6), led to the development of Eq. (10). The second postulate, expression (11), is somewhat intuitive, but has its basis in the fact that crack tip local behavior is not independent of the materials macro-mechanical ductility. Both of these postulates lead to a semi-empirical correlation that can be validated by data, and in this sense, they are consistent with the general premise of the Fracture Mechanics Field that is largely empirical. The simplicity of this correlation should not detract from its acceptance, unless it can be shown that it statistically disagrees with fracture toughness data developed in the traditional way. Potential users of this correlation are encouraged to present supporting or counter examples before they adopt or reject its use.

It should be emphasized that the fracture toughness correlation presented above is not intended to replace an ASTM-qualified fracture toughness test for cladding tubes under typical high-burnup conditions. Since such ASTM-qualified tests have not yet been developed, and in view of impending dry storage decisions, the above fracture toughness correlation offers an acceptable alternative. The application of the developed correlation will lead to a conservative assessment of the safety margin, or lack thereof, against cladding rupture in dry storage. As the above-described development would show, this correlation is no more empirical and no less reliable than fracture-toughness-based criteria in other safety-related industries such as the aircraft and pressure vessel industries.

Fracture Toughness Data

- CSED Approach - Issues Related To Cladding (cont.)
  - ***Issue 8:*** There is No Limitation on How The CSED is Quantified. The Uniaxial Test is all that is Available in Most Cases. However, the Material Ductility is Dependent on the Biaxiality or Triaxiality of the Stres State. A Biaxiality Factor is Applied to the Uniaxial Data When it is Combined with Biaxial Tests To Quantify CSED. However, One Should Keep In Mind that Energy is a Scalar, and Multiaxial Tests Reduce to a Scalar Quantity When Integrated.
  - ***Issue 9:*** Aluminum and Titanium Alloys Have Very Similar Stress-Strain Curves to Zircaloy. They Differ From Ferretic Steels. Austinetic Steels Exhibit Significant Hardening Unlike Irradiated Zircaloy. The CSED-K_{IC} Correlation Has Been Developed Implicitly for Irradiated Zircaloy-Like Materials

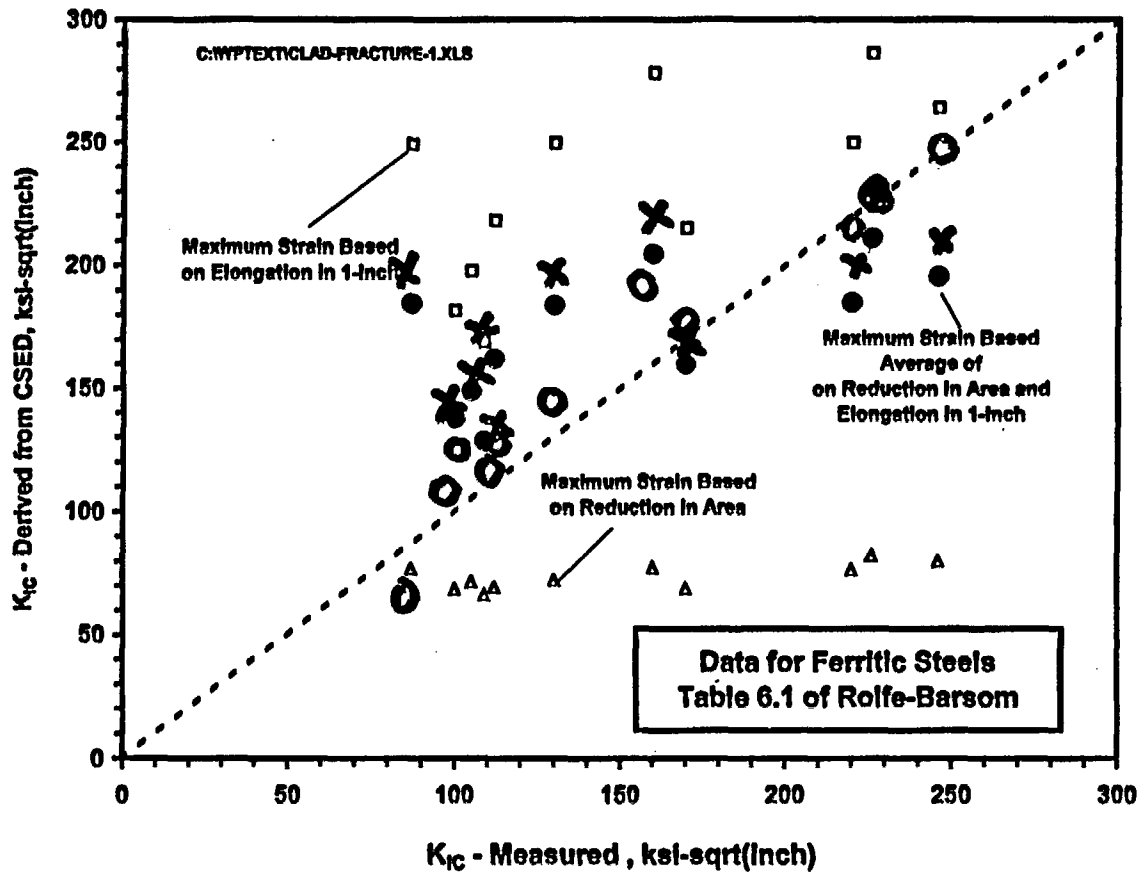
Fracture Toughness Data

- CSED Approach - Issues Related To Cladding (cont.)
  - ***Issue 10:*** We Obtained Different Values From Those Shown In the Figure. Our Values are Shown on the Figure in x, and the Values From the Rolfe-Barsum Correlation in Circles. As Can Be Seen, Despite the Fact that the Correlation was not Meant for Steels, the Agreement appears to be Not Bad
  - ***Issue 11:*** The Scatter is Typical For High Burnup Cladding, But It Can Be Explained. However, The Best-Fit Curve Works Remarkably Well.
  - ***Issue 12:*** Good Observation

CSED Approach – Issues Related to Cladding

Draft

10. When CSED approach was applied to ferritic steels, predicted and measured toughness values differed by a factor of 2.



Fracture Toughness Data

- Summary of Staff Analysis
  1. We Do Not Agree With This Assessment
  2. The Report Shows a Couple of Examples Where the Correlation Gave Excellent Agreement. Certainly More is Needed and Attempts Will be Made to Seek More Comparisons.
  3. We Agree Totally, and We Welcome This Type of Positive Feedback.