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NRC Staff Review of NEI/EPRI Reports On Creep And Fracture Toughness Of Zircaloy SNF Cladding

UNITED STATES NUCLEAR REGULATORY COMMISSION

NRC/NEI Meeting

April 18, 2001

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Scope of Discussion

For zircaloy cladding, under spent fuel dry storage conditions:

1. Limit For Creep Strain
2. Estimation of Fracture Toughness

References:

1. ***EPRI Report, "Creep As The Limiting Mechanism For Spent Fuel Dry Storage."***
2. ***Goll et. al. "Short-Time Creep and Rupture Tests On High Burnup Fuel Rod Cladding."***
3. ***EPRI Report, "Fracture Toughness Data For Zirconium Alloys – Application to Spent Fuel Cladding In Dry Storage."***

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Scope Of The EPRI Creep Report

Spent nuclear fuel (SNF) zircaloy cladding failure under dry storage conditions due to:

1. **Stress Corrosion Cracking**
2. **Delayed Hydride Cracking**
3. **Creep**

EPRI Report, "Creep As The Limiting Mechanism For Spent Fuel Dry Storage."

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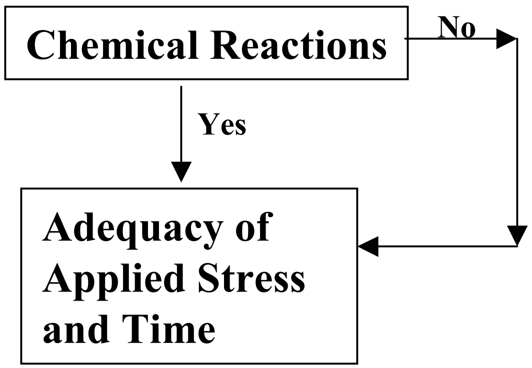
Major conclusions of this report are as follows:

For dry storage of spent fuel,

- **Stress corrosion cracking (SCC) is not a factor.**
- **The stress intensity factor values are such that delayed hydride cracking (DHC) is not a factor.**
- **Existing data support a newly proposed criterion of 2% allowable creep strain without failure.**

Stress Corrosion Cracking (SCC)

During storage, no release of fission products, and no effects of iodine, or Cd-Cs



<u>Threshold stress, MPa for SCC</u>

Material	Unirradiated	Irradiated
Zr-2	360	180
Zr-4	300	200

Assume, oxide thickness of 100 µm
Assume license pressure of 18.6 MPa
Then, calculated hoop stress < 150 MPa

Analysis With Pre-Existing Cracks

From literature, K_{ISCC} = 3.4 – 5.5 MPa√m

Using, a crack size of 160 µm, cladding hoop stress of 200 MPa, and using fracture mechanics relation, K_I = 5.0 MPa√m.
Can not remain sub-critical in the reactor.

Assuming that flaws of approximately 70 µm can, however, preexist, a K_I of only 2.54 MPa√m for a stress of 200 MPa is possible.

Thus SCC under dry storage can be ruled out.

Stress Corrosion Cracking (SCC)

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Role Of Stress And Time-At-Stress

Claims:

1. Crack initiation requires intergranular cracking – not possible under dry storage conditions. Thus, the onset of second stage for rapid crack propagation (transgranular failure) is unlikely.
2. Higher strain rates (due to rapid strain) lead to (non-ductile) transgranular failure. Under dry storage conditions of low stress and long failure times, this is not likely.

Issues:

1. Crack initiation via. void nucleation, coalescence, and growth is potentially feasible due to creep under dry storage conditions.
2. Localized high stress rate as crack grows with elevated stress intensity needs greater consideration because of its potential adverse influence on creep life.

Stress Corrosion Cracking (SCC)

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Role Of Stress And Time-At-Stress

Issues:

3. **Local inhomogeneities such as hydrides can result in mixed-mode crack growth behavior – one need not have just transgranular crack propagation for catastrophic failure**
4. **The stress-dependent minimum time for SCC failure by transgranular mode could be influenced by the presence of hydrides and surface flaws.**
5. **An estimate for threshold stress for SCC can be calculated as approximately 130 MPa by conservatively including pre-cracking (From Figure 4). This value is less than the cladding hoop stress of 150 MPa, which is reported in the EPRI report.**

Thus, summary 2.6 (b) of the report has not been adequately supported; however, other literature information indicate that SCC of zircaloy cladding is unlikely under dry fuel storage conditions.

Stress Corrosion Cracking (SCC)

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Effect of Irradiation/High Burnup

Claim:

1. **Hydrides dissolve at the start of SCC testing. Results do not indicate any adverse effect of the presence of hydrogen on the susceptibility to SCC.**

Issues:

1. **Even when hydrides dissolve, the associated cracks are left behind. Thus, the influence of these cracks when they potentially adjoin and become SCC sites needs to be examined.**
2. **In presenting the argument that pre-existing cracks remain sub-critical, it appears that a "circular calculation" might have been performed. Under the stipulated conditions, calculations show that a stress intensity factor of 3.77 MPa√m can be obtained, which exceeds the K_{ISCC} of 3.4 MPa√m. The paper reports a lower value of 2.54 MPa√m for stipulated conditions by using circular calculation.**

Thus, summary 2.6 (c) of the report has not been adequately supported; however, other literature information indicate that SCC of zircaloy cladding is unlikely under dry fuel storage conditions.

Delayed Hydride Cracking (DHC)

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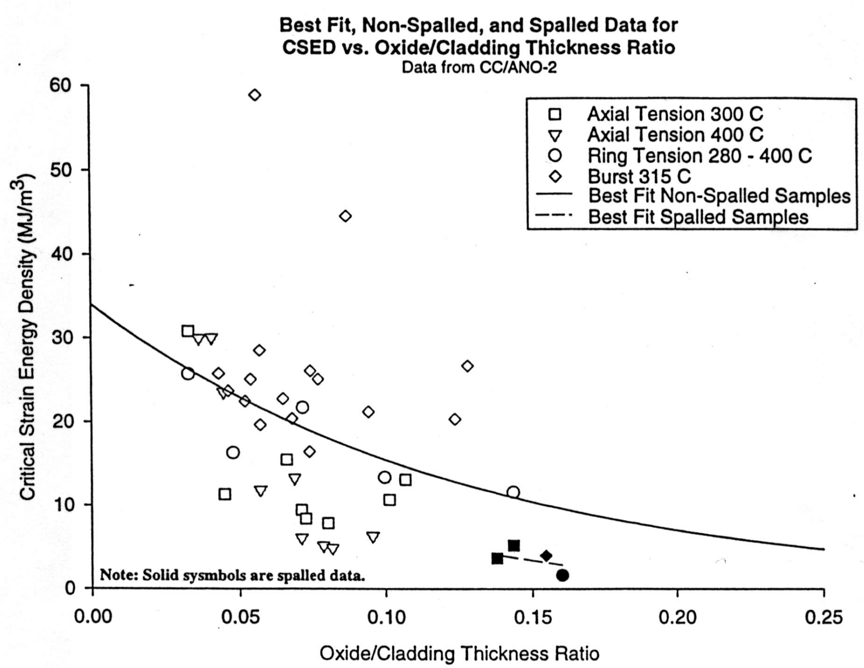
1. **Hydrogen – Presence and Adequacy**
2. **Temperature, and Temperature History**
3. **Role of Stress**

Effect of Oxidation
Effect of Hydrides



Assume crack of approx. 100 µm.
Stress of 150 MPa.
Then, $K_I = 3.0$ MPa√m.
$K_{IH} = 5$ MPa√m
$K_{ISCC} = 3.4 – 5.5$ MPa√m.

DHC is not a factor under dry storage conditions

Delayed Hydride Cracking (DHC)

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Evolution of the DHC Mechanism

Claims:

1. **Hydride crack growth mechanism, consisting of intermittent crack advances can be discerned from Figure 7.**
2. **Critical stress intensity factor values required for analysis of DHC can be estimated from the proposed critical strain energy density (CSED) model. DHC is not a factor under dry storage conditions.**

Issues:

1. **The mechanism for crack growth can not be discerned from Figure 7, which depicts the crack displacement-test time profile. The Figure only shows plastic instability above 2800 hrs.**
2. **The critical strain energy density (CSED) relationship has not been verified for zircaloy cladding to the same extent as for aluminum. Considerable scatter of data for zircaloy is observed. Thus, the K_{Ic} estimation for zircaloy cladding from the CSED formulation may not be valid. Additionally, sufficient fracture toughness data for zircaloy cladding, representative of highly oxidized (high burnup) state, is not yet available for analysis.**

Delayed Hydride Cracking (DHC)

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Re-Orientation of Hydrides

Claim:

1. **Hydride reorientation presents no additional concerns and would not impose undue restrictions on vacuum drying.**

Issues:

1. **The possibility of a chain of hydride precipitates, rather than an isolated one of approximately 100 µm, could lead To DHC. This possibility has not been analyzed.**
2. **The effect of the biaxial nature of the stress state on the crack tip in "attracting" hydrides is not clear.**
3. **For hydride reorientation, the claim that stresses > 138 MPa and several thermal cycles are needed may not be supportable. For example, literature data indicate that hydride reorientation occurs between 68 and 200 MPa, depending on texture and temperature, and is typically 120 to 150 MPa for cladding similar to commercial fuel.**
4. **The EPRI reported data has an upper bound cycling temperature of 250 °C and a hydrogen level of only 50 ppm. Both of these conditions (temperature and hydrogen level) are below those expected for dry storage of high burnup rods.**

Thus, Summary 3.7 of the report has not been adequately supported.

Creep Data Of Zircaloy Cladding

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1. For burn-up 16-18 GWD/MTU, at 482 to 571 °C, for up to 7600 hr. of creep, at stresses of 40 to 75 MPa, uniform strain of 1.7% to 7.0% was observed. (Einzinger et. al.)

2. At 300 and 370 °C, for stress levels of 320 to 630 MPa, observed failure strains were ≥ 2.5% to ≅ 6%. (Goll et. al.)

3. At 350-400 °C and 350-386 MPa stress, observed creep ductility was 9-12%. Hydrogen was in the range of 215 – 1040 ppm.(Bouffioux and Rupa)

These results justify a proposed 2% creep strain limit criterion.

Creep Data Of Zircaloy Cladding

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Goll et. al. Data

Claims:

1. **Data provide support for a proposed 2% creep limit criterion**
2. **Ductile zirconium matrix immediately blunts cracks emanating from radially-oriented hydrides**

Issues:

1. **The follow up creep test after accelerated creep tests was done for 5 days 100 MPa stress and at 150 °C. These conditions are insufficient to test the ductility of zirconium matrix. In the absence of supportive microscopy evidence, crack blunting by ductile matrix is only a conjecture.**
2. **Test specimens have an oxide thickness of 25 µm that represents about 175 ppm of hydrogen. This oxide thickness has little relevance to high-burn up SNF.**
3. **Hydride orientation and cracking are statistical processes. Observation of non-failure in limited number of experiments can not be adequately equated to full assurance of non-failure under dry storage conditions.**

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Creep Data Of Zircaloy Cladding

Acceptance Criterion – Creep-Based Limit

Claims:

1. Absence of voids in zircaloy cladding leads to diffusion-controlled cavity growth (DCCG) being not a limiting failure mechanism for dry storage.
2. The proposed 2% criterion is valid for all cladding types regardless of oxide thickness or spalling of the oxide.

Issues:

1. Sufficient literature data show that voids are initiated due to plastic deformation of zircaloys. Potentially, adjacent voids can coalescence and grow during creep to critical size for fracture under favorable stress orientation, stress, and temperature.
2. The cited reference by Einzinger et. al. indicates uniform strain range from 1.7% To 7% for low burnup fuel. Thus, the observed lower strain limit is less than the proposed strain limit.

Creep Data Of Zircaloy Cladding

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Acceptance Criterion – Creep-Based Limit

Issues:

3. **Consistency is needed when using *uniform* and *total* creep strain values. The cited Garde et. al. data were for a total strain of 0.58% and an uniform strain of only 0.05%. Due to potential stress magnification in the localized "necked" region by a factor of 2 or greater as a result of plastic instability, uniform strain is more appropriate for establishing a strain limit for licensing.**
4. **The cited Fuketa et. al. data Indicate significant scatter in the two sets of stress rates. The Data show that the strain capacity drops for both stress rates with increasing hydrogen levels. The difference vanishes when hydrogen levels exceed 450 ppm.**

Creep Data Of Zircaloy Cladding

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Acceptance Criterion – Creep-Based Limit

Issues:

5. The cited Goll et. al. data show uniform strains ≥ 2.5% for hydrogen levels up to 600 ppm without spalling. However, commercial high burnup fuel cladding can have hydrogen levels > 600 ppm with spalling.
6. Many of the cited data are for materials not typical for high burnup. Either the cladding is unirradiated or the cladding is uniformly hydrided. Literature show that when the hydride density is large, the local ductility decreases to near zero.
7. Calculations of critical stress intensity factor, K_{Ic}, to support postulations do not contain information on flaw characteristics; Thus, the variability and sensitivity in such calculations can not be determined.

Thus, Conclusion 9 of the report has not been adequately supported.

Creep Data Of Zircaloy Cladding

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Pin-Hole Equivalent Failure Mode

Claims:

1. Using the concept of "leak-before-failure", a pin hole with a depth almost equal to cladding thickness (600 µm) will require a stress level of 170 MPa for fracture – higher than cladding stress, which is < 150 MPa.
2. The calculations are valid for all cladding conditions regardless of oxide thickness or the state of the oxide, coherent or spalled.

Issues:

1. The calculations do not address the potential for the presence of a series of pinholes and the margin of safety in the stress level that would be required for reasonable assurance of safety.
2. The analysis does not address whether the velocity of the pressure wave is greater than the velocity of the crack.
3. Reliable fracture toughness data for high burnup cladding containing spalled oxide are lacking to support the calculations.

Thus, conclusion 10 of the report has not been adequately supported.

Summary Of Staff Analysis

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From an analysis of both EPRI report and other literature, the staff concludes that an increase of the current 1% creep strain limit for high burnup zircaloy cladding is not justified for dry storage of commercial spent nuclear fuel.

It could be worthwhile to evaluate creep data for zircaloy cladding from parametric considerations such as, for example, Larson-Miller or other type, so that probabilistic failure estimates can be made under dry storage of spent nuclear fuel. Such parametric treatments of creep data inherently include the mechanisms for creep.

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NRC Staff Review of NEI/EPRI Report On Fracture Toughness Data For Zirconium Alloys – Application To Spent Fuel Cladding In Dry Storage

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Scope of Discussion

Reference:

1. *EPRI Report, "Fracture Toughness Data for Zirconium Alloys – Application to Spent Fuel Cladding in Dry Storage", January 2001.*

EPRI's evaluation of fracture toughness data of zirconium alloys to estimate values applicable to spent fuel conditions

1. **The basis for estimate – Critical Strain Energy Density (CSED)**
2. **Applicability to Zirconium alloy cladding**

Fracture Mechanics Concept

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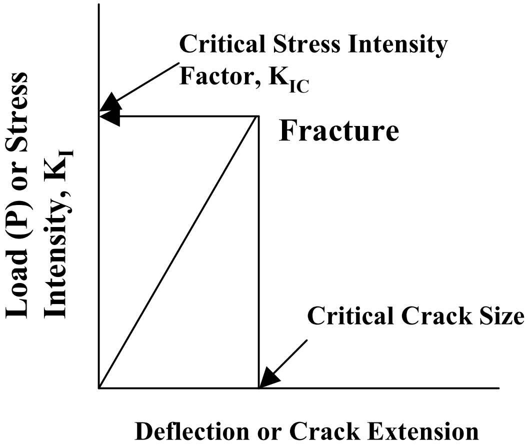
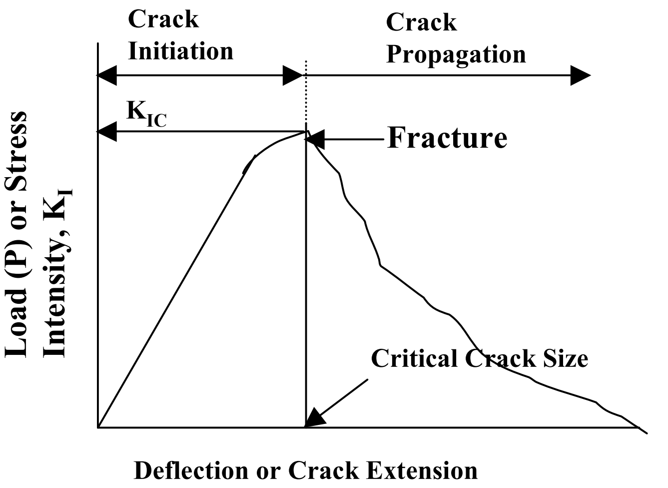
Linear Elastic Fracture Mechanics (LEFM)



Elastic-Plastic Fracture Mechanics



As Load (P) increases, stress (σ) increases
As Load (P) increases, stress intensity factor (K_I) increases
At fracture, the stress equals fracture stress (strength, σ_f)
At fracture, the stress intensity factor becomes critical stress intensity factor, K_{Ic}, fracture toughness.

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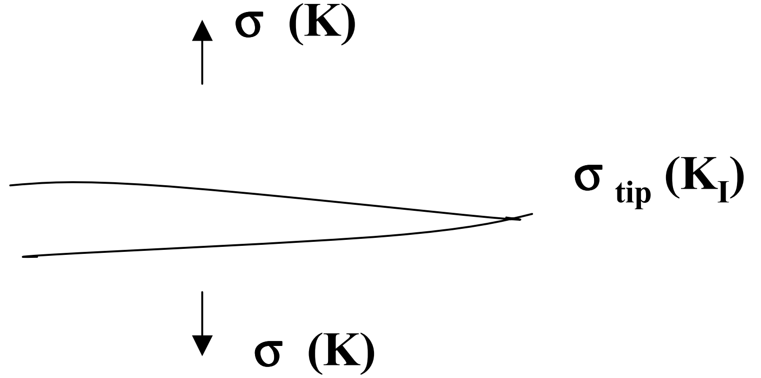
Fracture Mechanics Concept

Stress Magnification At Crack Tip: $$\sigma_{tip} = \frac{\sigma}{\sqrt{\rho}}, \qquad (1)$$

Stress Intensity At Crack Tip: $$K_I = \frac{1}{Y}\sigma\sqrt{c}, \qquad (2)$$

At 'fracture', $$\sigma \rightarrow \sigma_y, \quad c \rightarrow c_{crit}, \quad K_I \rightarrow K_{Ic} \qquad (3)$$



Critical Strain Energy Density (CSED)

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CSED is defined as the integral of the product of stresses and strains obtained in a test.

$$U_c = \sum_{0}^{\varepsilon_{ij}} \sigma_{ij} d\varepsilon_{ij} \quad \text{(Eq.5 of EPRI Report)}$$

By equating the CSED of the LEFM material to the CSED of the actual material,

$$U_c = \frac{\sigma^2}{2E} = \sigma_y \varepsilon_{TE} - \frac{1}{2}\sigma_y \varepsilon_y$$

(Eq. 8 of EPRI Report)

Critical Strain Energy Density (CSED)

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EPRI Paper relates the CSED to K_{Ic} by:

$$K_{Ic}^2 = \frac{4\pi E U_c \rho_y}{2r-1},$$ **(Eq. 10 of the report)**

where, ρ_y is the 'position' (plastic zone size) of the order of 150 – 200 μm.

And, finally, for 'highly irradiated zircaloy':

$$K_{Ic} = 3.5\sqrt{U_c}$$ **(Eq. 13 of the report)**

U_c represents the area under the stress-strain curve in an uniaxial test.

CSED Approach – Issues Related to Cladding

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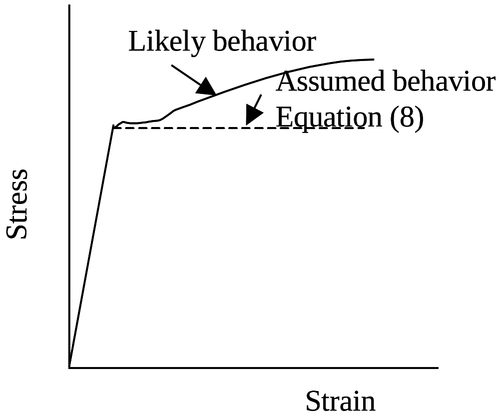
1. In the equation defining K_{Ic}, the definition of ρ is not clear. What is the 'position' of the crack? Is it crack size? Is it plastic zone size? Is it crack size plus the plastic zone size?

2. In equation (8), why is the stored elastic energy subtracted from the total strain energy, considering that the strain energy consists of energy for yielding, energy for crack nucleation, and energy for crack propagation?

3. Why is the yield stress being multiplied by the total energy contribution? For example, typically, stress does not remain constant with strain after yielding.



CSED Approach – Issues Related to Cladding

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4. Fracture energy determinations involve analyzing load-deflection (load-crack length) behavior of well-defined cracks under specific loading conditions. These provide usable data for crack initiation and crack growth. Analysis and integration of "typical" stress-strain data, under uniaxial tests do not provide such information.

5. The equation (9) of the report seems to be incorrect, as it takes contributions from the two equals for the total strain energy in equation (9).

6. What measure of strain should be used for determining CSED? Reduction in area (that will include Poisson effects) or tensile elongation?

CSED Approach – Issues Related to Cladding

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7. The total strain energy, U_c, in equation (13) should be divided by crack length (or, crack area?); otherwise, an imbalance in units exists. However, questions still persist with respect to the adjustable constant, 3.5, which depends on the factor 'r' and its interpretation.

8. CSED approach estimates the fracture toughness of a material on the basis of uniaxial stress-strain curves. Crack initiation and crack propagation under other loading situations, for example creep, might involve complex fracture mechanisms, thus rendering the application of CSED less meaningful.

CSED Approach – Issues Related to Cladding

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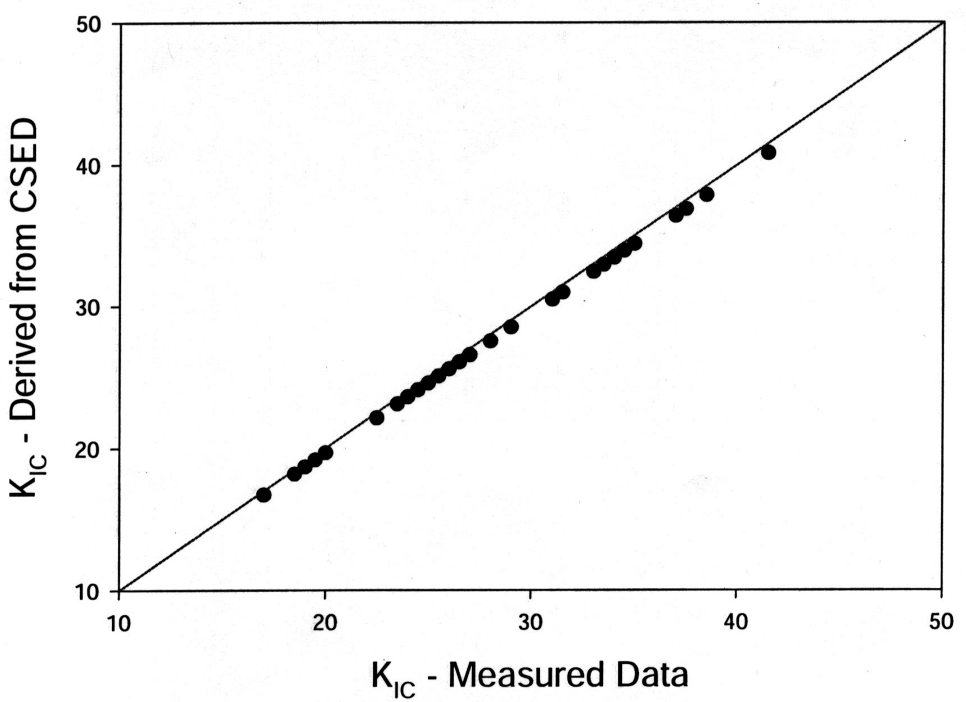
9. Very good correlation between K_{Ic} and U_c (CSED) has been shown for aluminum alloys. Does one obtain such good correlation for other materials, especially for materials of fuel cladding?



CSED Approach – Issues Related to Cladding

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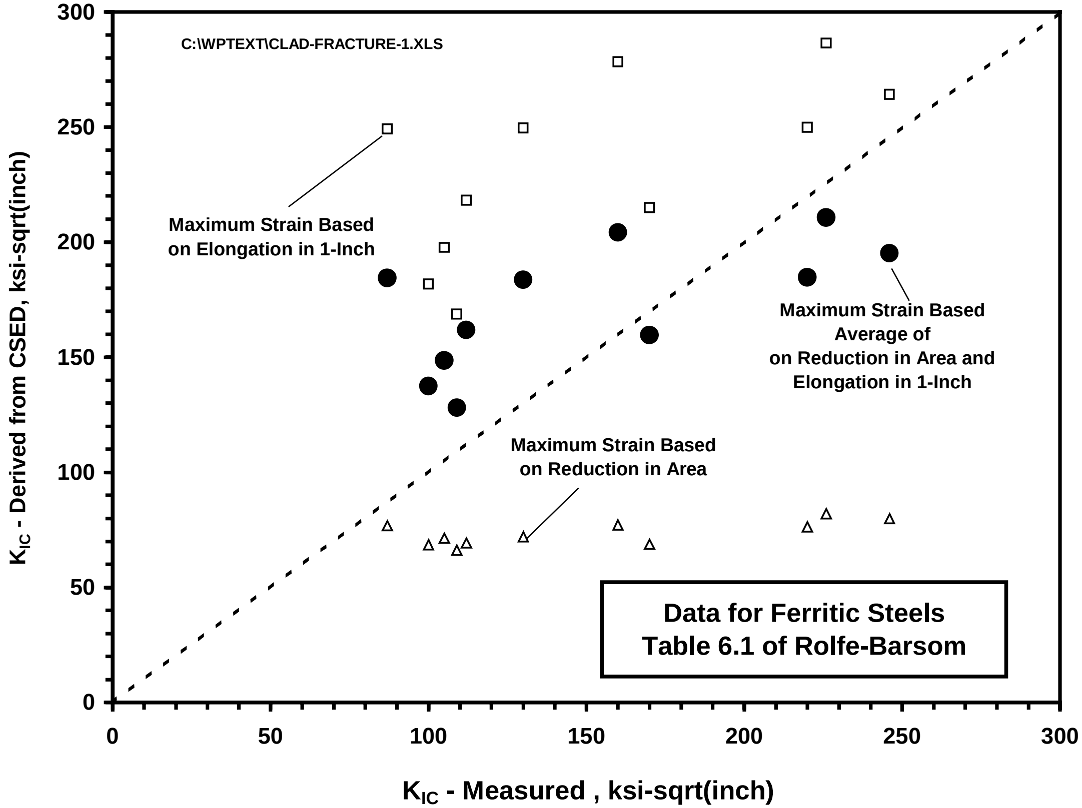
10. When CSED approach was applied to ferritic steels, predicted and measured toughness values differed by a factor of 2.



CSED Approach – Issues Related to Cladding

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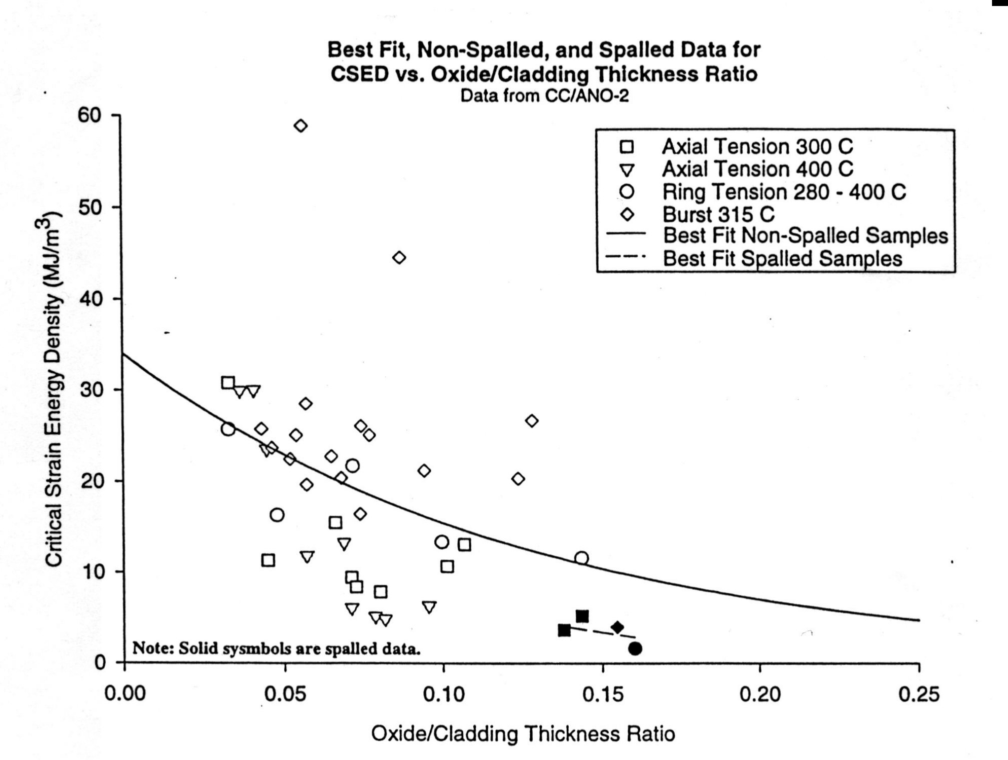
11. An examination of Figure 3 of EPRI report, which shows the relationship between CSED and oxide thickness for irradiated and hydrided fuel cladding shows wide scatter in the data.



CSED Approach – Issues Related to Cladding

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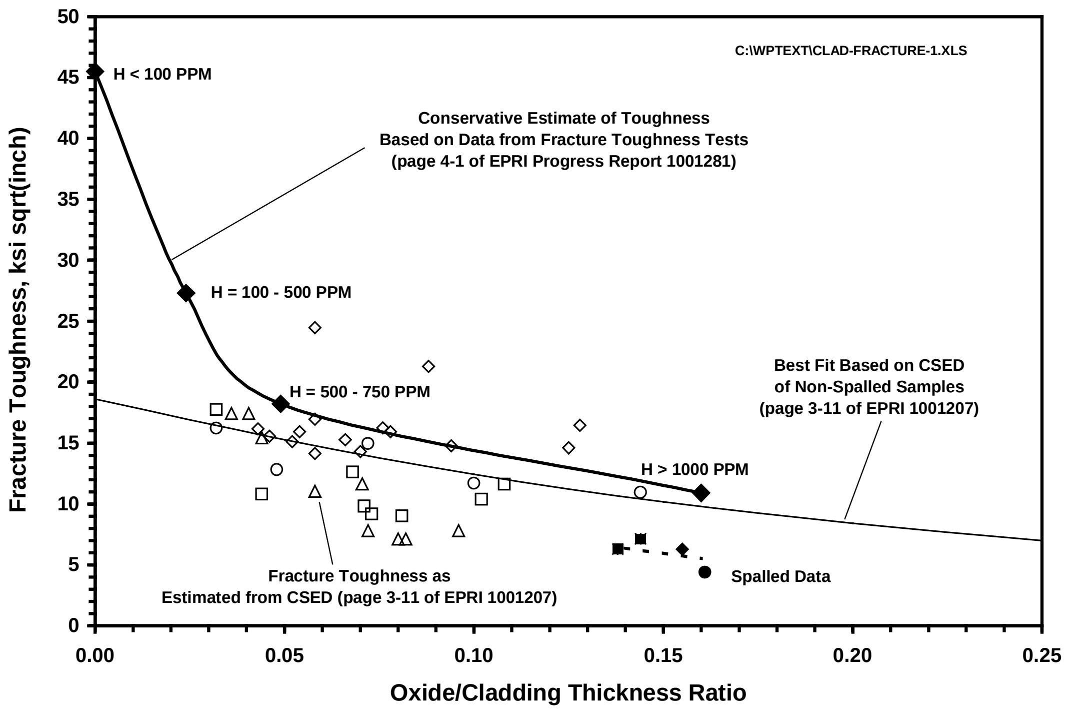
12. Re-casting the data provides some useful information, although large uncertainties still pertain. In general, CSED estimates seem more conservative.



Summary Of Staff Analysis

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1. The analysis of the EPRI report seems to indicate some fundamental flaws in the derivation of critical strain energy density (CSED) and its relationship to fracture toughenss.
2. Applicability and validity to typical cladding materials, particularly zircaloy is not available.
3. Because of its simplicity and potential supplement to fracture toughenss testing, correlations of CSED with fracture toughness could permit structural integrity evaluations of irradiated and hydrided cladding to be performed in situations where only limited data are available; however, adequate calibration and validation of the CESD methods are needed.