Creep Anisotropy and Transitions in Zircaloy Cladding

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Applications to:

- Zry manufacturers
- In-reactor clad dimensional predictions
- Reliability of spent fuel during dry storage & transportation

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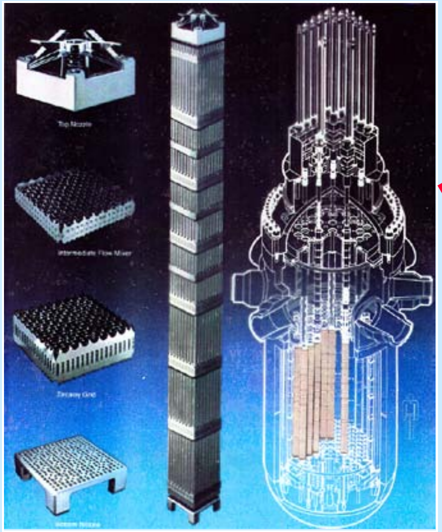
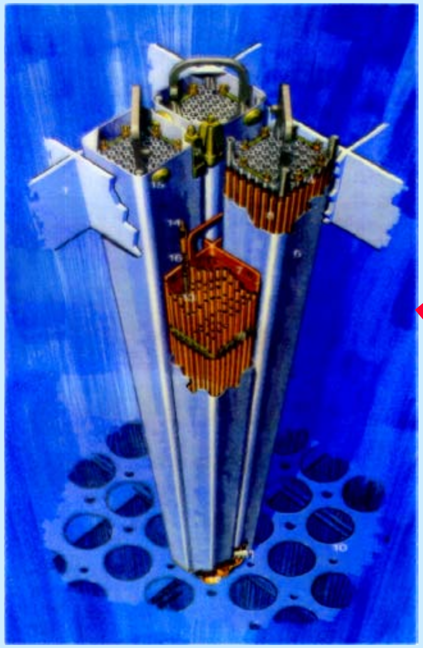
Research supported by NSF and DOE/NEER

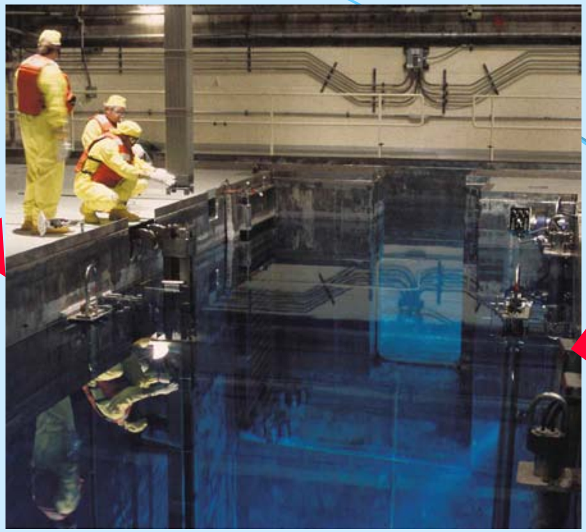
Objective

PWR

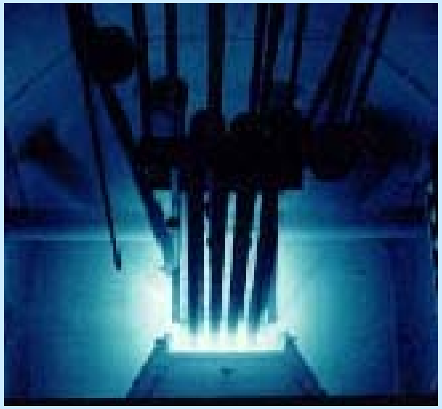




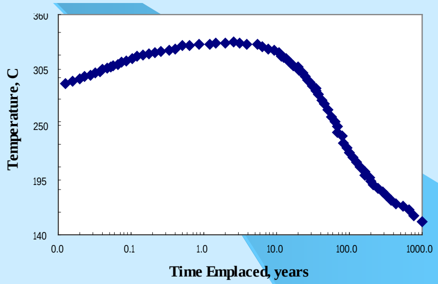
BWR



Storage Pool
at Reactor Site



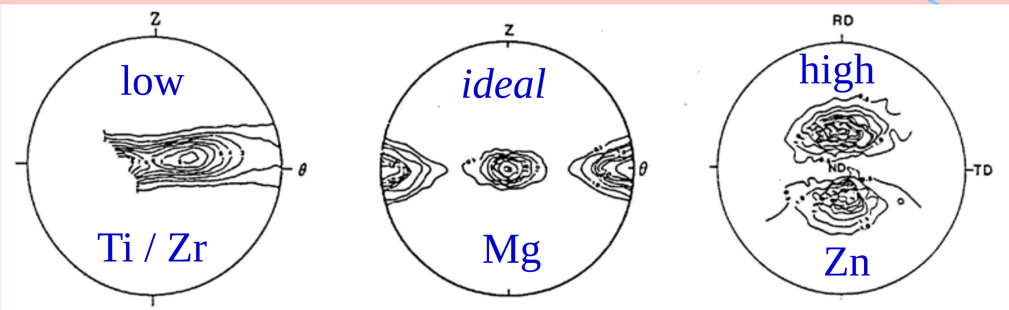
Dry Storage at Site

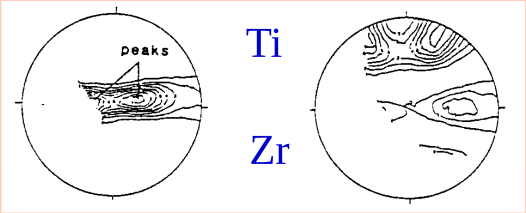


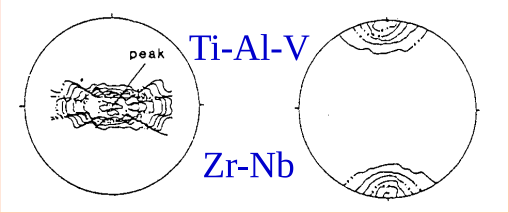
Eventual Disposal

Creep failure during dry storage

hcp metals
effects of c/a-ratio and alloying







X-Ray Pole Figures
Basal, Prism, Pyramidal

↓

CODF

Lower-Bound
Power-Law Creep

Slip Mode
Basal, Prism, Pyramidal

↓

Bulk Polycrystalline
Behavior

in-service prediction | **formability**

Spent fuel reliability

Started as an NSF grant research in 1983

NCSU NUCLEAR MATERIALS

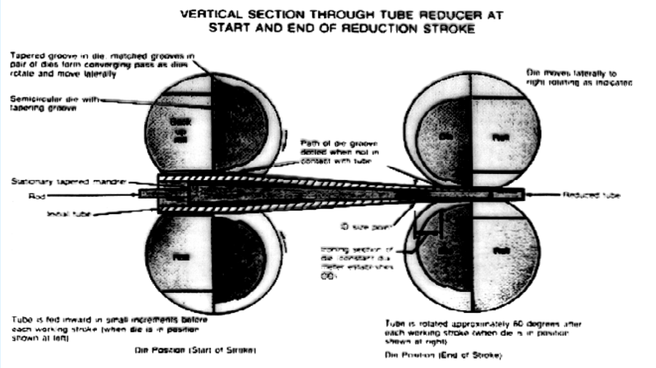
Zircaloys

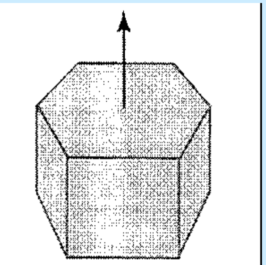
hcp structure with limited slip systems ⇒texture

	Sn	Fe	Cr	Ni
Zry-2	1.2 – 1.7	0.07 – 0.20	0.05 – 0.15	0.03 – 0.08
Zry-4	1.2 – 1.7	0.18 – 0.24	0.07 – 0.13	---
Zry-Nb	1.2 – 1.7	0.18 – 0.24	0.07 – 0.13	1.0 (Nb)

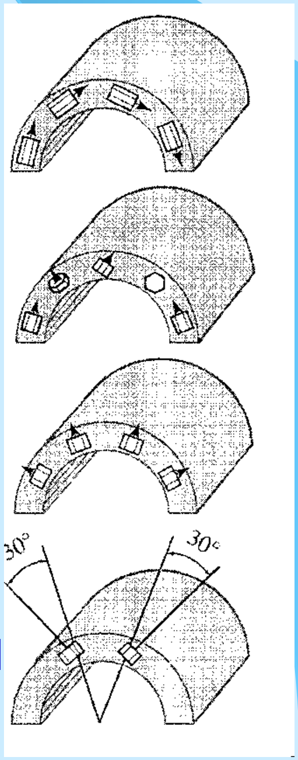
Texture Effects

- **Radiation Growth (Fuel Rods and Channels)**
- **Hydride Orientation**
- **Corrosion and Stress Corrosion Cracking**
- **Deformation and Creep**
  --- anisotropy & formability



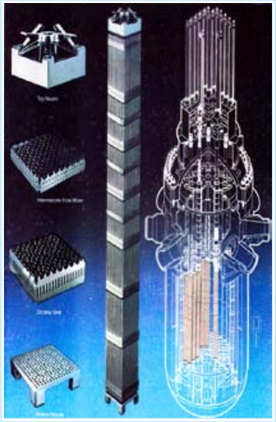


hcp unit cell



MATERIALS

TUBING PLATE TUBE-SHELL



TEXTURE → **ODF - PLASTICITY MODEL**

MECHANICAL ANISOTROPY

FORMABILITY

BIAXIAL CREEP

SCC

Lower-Bound
& Power-Law
Upper-Bound

Bishop-Hill
Rigid-Plastic

IN-REACTOR PERFORMANCE
Cladding, Grids and Channels

TUBE & GRID FABRICATION
from TREX and Sheet

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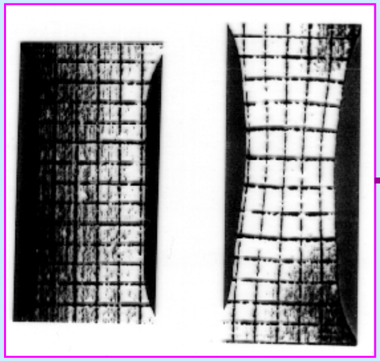
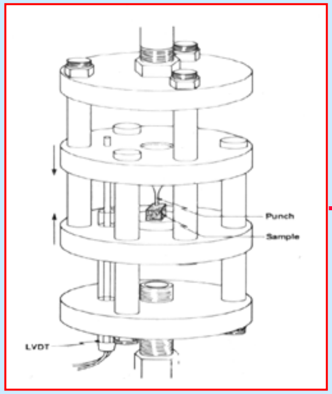
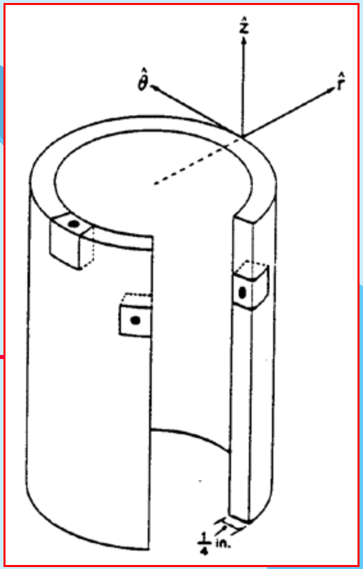
ZIRCALOY MATERIALS & TEST METHODS

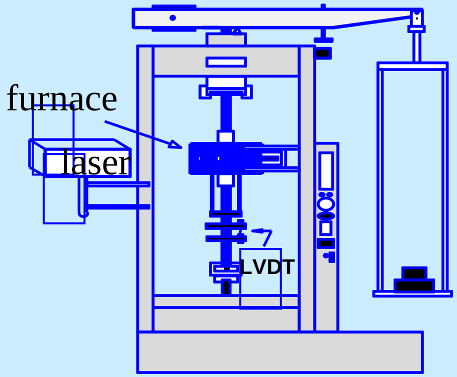


ZIRCALOY - 4 Sheet
0.46 mm [~ 18 mils] thick
Tensile Tests
Gridded Specimens
[31.75mm x 6.35mm]



ZIRCALOY - 2 TREX
100 mm OD x 10 mm wall
Impression Tests
[6.35mm x 6.35mm x 6.35mm]





CLADDING
ZIRCALOY -2 & ZIRCALOY -4
[standard BWR & PWR]
Biaxial Creep
[Creep Locus]

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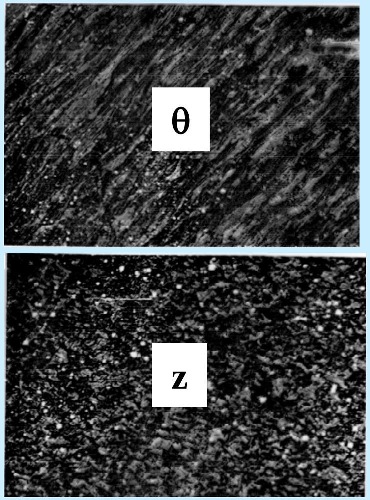
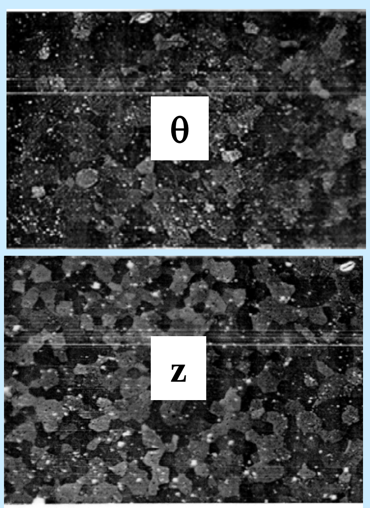
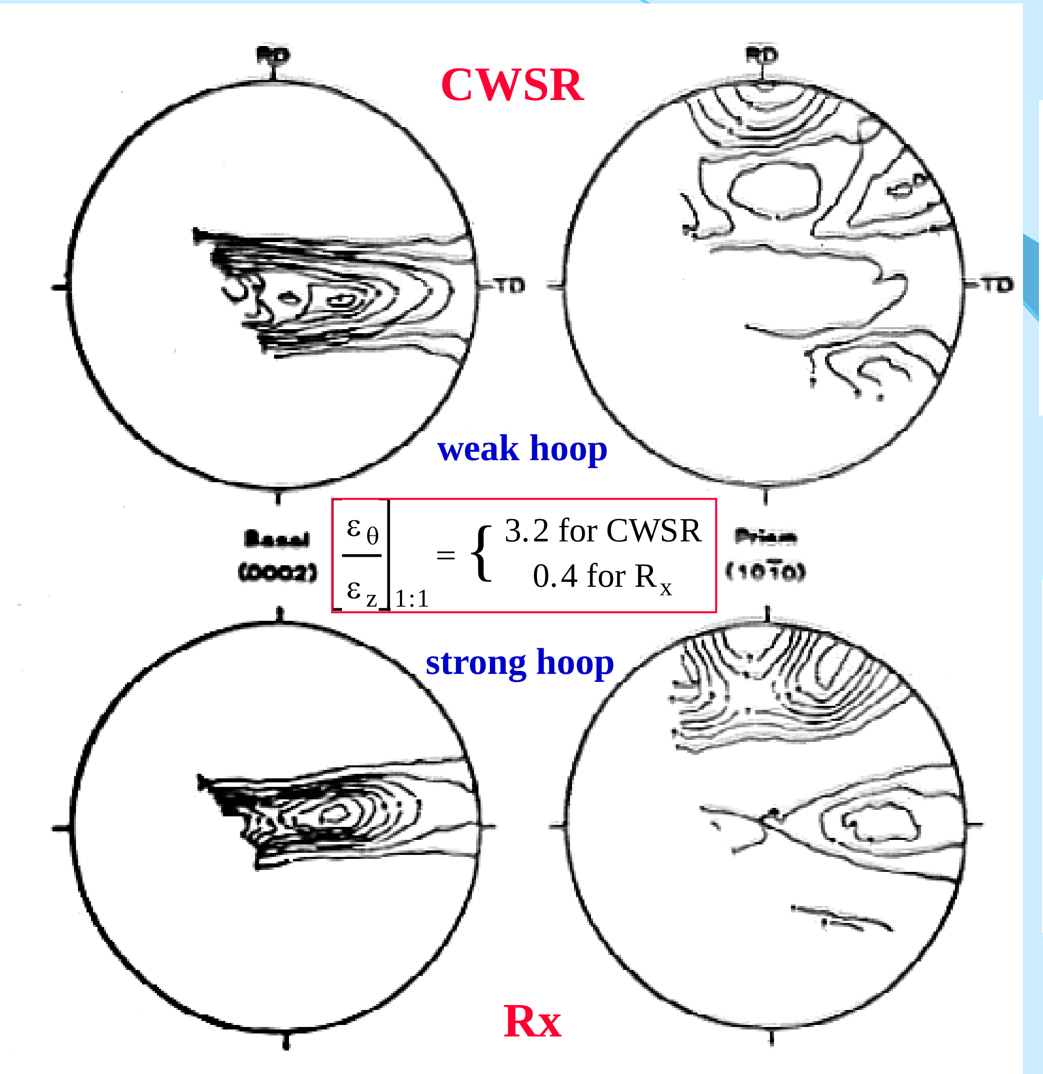
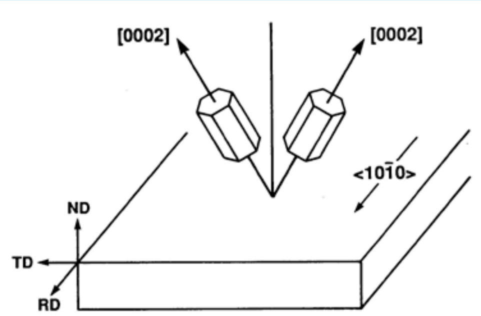
CWSR vs Rx Zircaloy



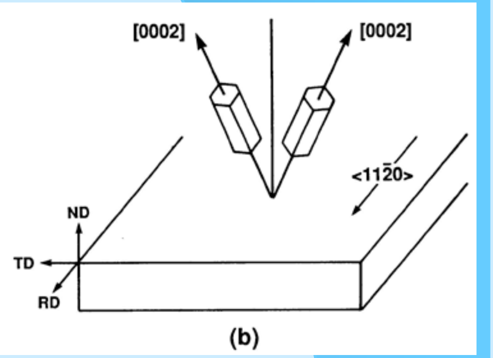
D~10μm



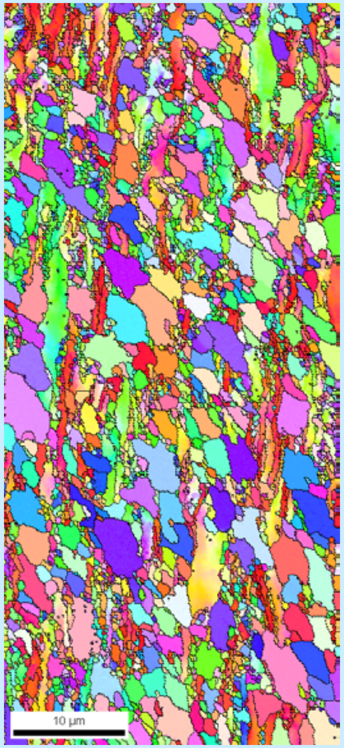


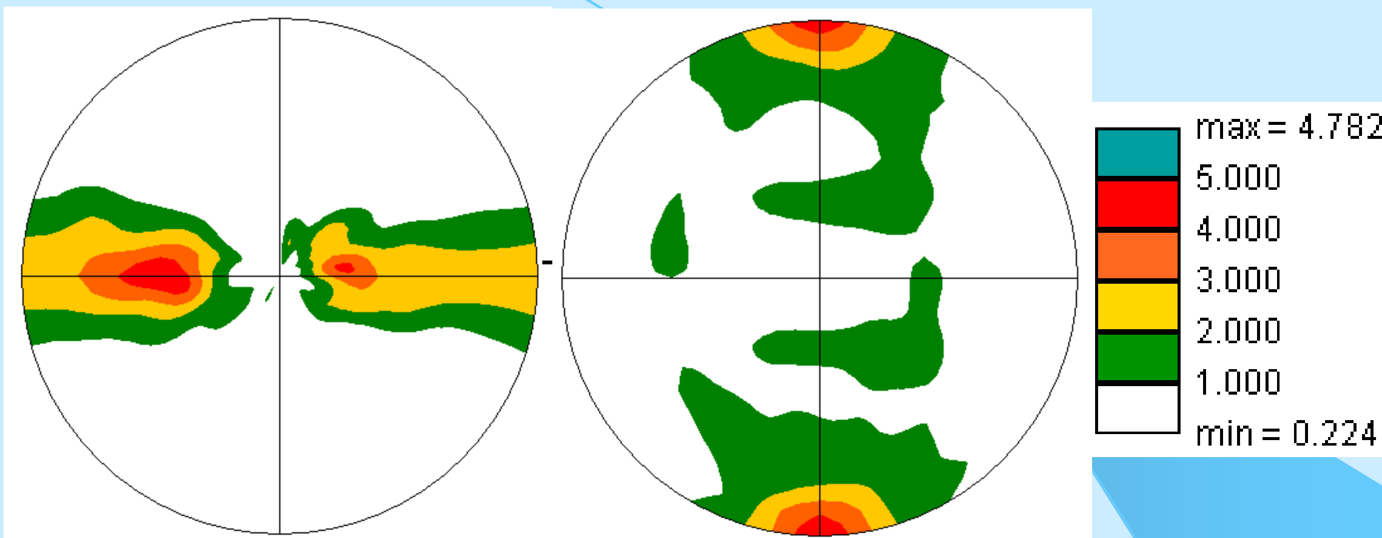


Hoop **stronger** at RT



Texture using OIM





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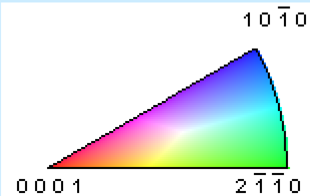

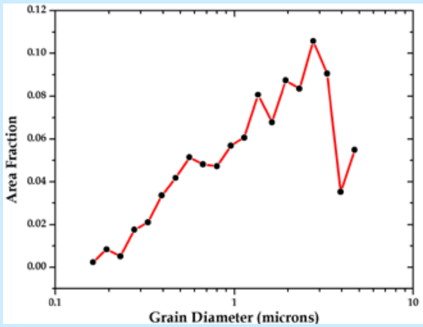

CWSR Zircaloy cladding

CODF-Slip Model

$$w(\theta,\psi,\phi)=\sum_{l=0}^{\infty}\sum_{m=-l}^{l}\sum_{n=-l}^{l}W_{lmn}Z_{lmn}(\cos\theta)e^{(-im\psi)}e^{(-in\phi)}$$

$$Q_{lm}^{i}=2\pi\left(\frac{2}{2l+1}\right)^{\frac{1}{2}}\sum_{n=-l}^{l}W_{lmn}P_{l}^{n}(\cos\Theta_{i})e^{(in\Phi_{i})}$$

$$Q_{lm}^{i}=\frac{1}{2\pi}\int_{0}^{2\pi}\int_{-1}^{1}q^{i}(\chi,\eta)P_{l}^{m}(\cos\chi)e^{(imn)}d(\cos\chi)d\eta$$

$$q^{i}(\chi,\eta)=\frac{I^{i}(\chi,\eta)}{\int_{0}^{2\pi}\int_{-1}^{1}I(\chi,\eta)d(\cos\chi)d\eta}$$

$$\langle\rho(\theta,\psi,\phi)\rangle=\int_{0}^{2\pi}\int_{0}^{2\pi}\int_{-1}^{1}\rho(\theta,\psi,\phi)w(\theta,\psi,\phi)d(\cos\chi)d\psi d\phi$$

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CODF-Slip Model

$$\sigma_g^2 = \frac{R(\sigma_r - \sigma_\theta)^2 + RP(\sigma_\theta - \sigma_z)^2 + P(\sigma_z - \sigma_r)^2}{P(R+1)}$$

$$\begin{pmatrix} \dot{\varepsilon}_r \\ \dot{\varepsilon}_\theta \\ \dot{\varepsilon}_z \end{pmatrix} = \frac{\dot{\varepsilon}_g}{P(R+1)\sigma_g} \begin{pmatrix} (R+P) & -R & -P \\ -R & R(P+1) & -RP \\ -P & -RP & P(R+1) \end{pmatrix} \begin{pmatrix} \sigma_r \\ \sigma_\theta \\ \sigma_z \end{pmatrix}$$

$$\dot{\gamma}^{(k)} = A\left[\frac{\tau^{(k)}}{\tau_0^{(k)}}\right]^n$$

$$\dot{\varepsilon}_{ij}^c = \sum_{(k)} \mu_{ij}^{(k)} \dot{\gamma}^{(k)}$$

$$\tau^{(k)} = \sigma_{ij}^c \mu_{ij}^{(k)}$$

$$\sigma_{ij}^{ub} = \int_\theta \int_\psi \int_\phi w(\theta, \psi, \phi) \sigma_{ij}^c \, d(\cos\theta) \, d\psi \, d\phi$$

$$\dot{\varepsilon}_{ij}^{lb} = \int_\theta \int_\psi \int_\phi w(\theta, \psi, \phi) \dot{\varepsilon}_{ij}^c \, d(\cos\theta) \, d\psi \, d\phi$$

Experimental Techniques

Burst Tests

- Sustained gas pressurization to 15 ksi
- Temperatures from ambient to 600 C
- Post burst specimen profile using Laser Extensometer
- Time to rupture, uniform hoop strain
- Approximate steady-state strain-rate
- Larson-Miller Parameter
- Hoop Strain-Rate vs Stress & Temperature

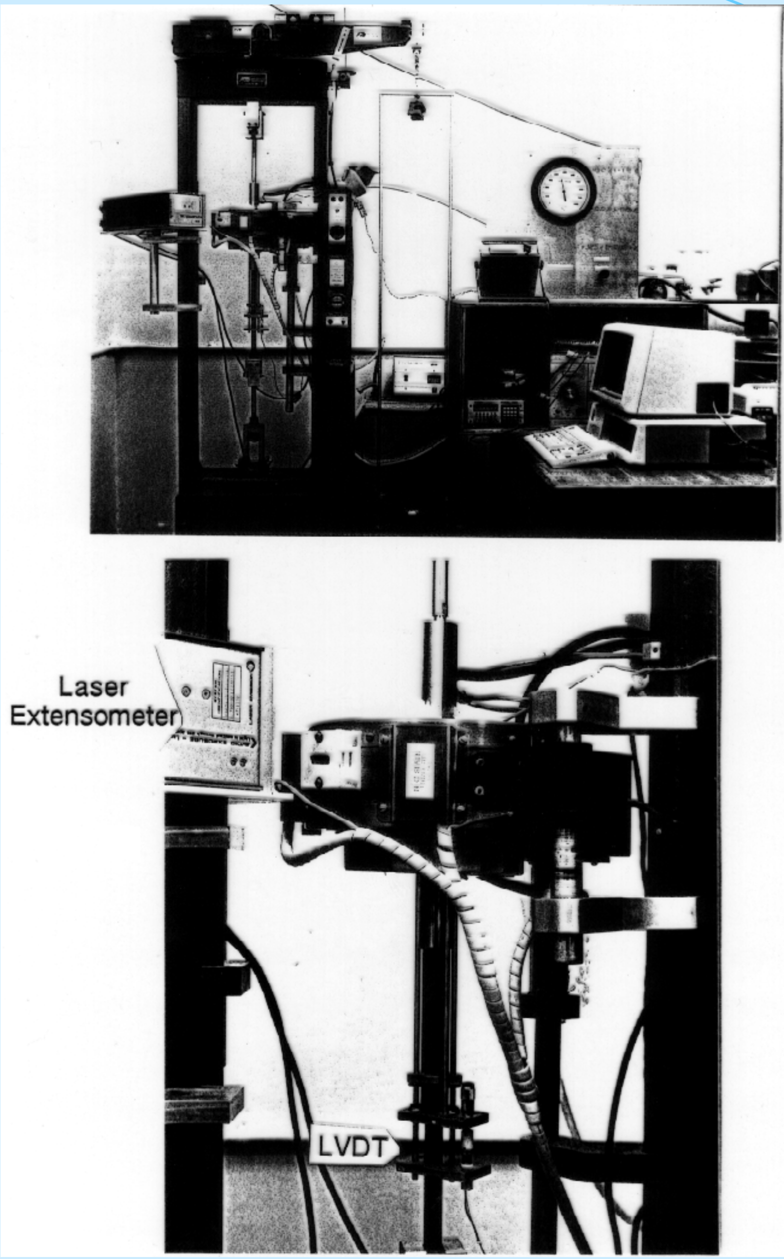
Creep Tests

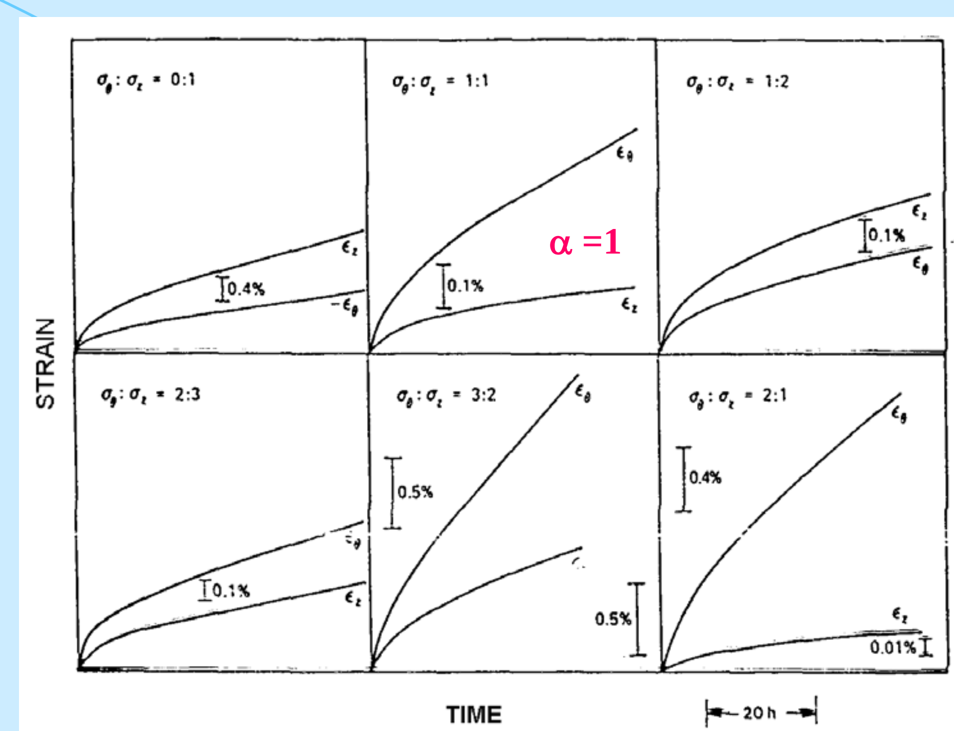
- Biaxial creep tests using internally pressurized tubing
- Stress state (σ_θ/σ_z) varying from 0 (uniaxial) to 2 (internal pressure)
- Monitor hoop and axial strains using Laser and LVDT extensometers vs time
- Hoop and axial strain-rate vs stress
- Primary and steady-state creep
- Transitions In creep mechanisms
- Strain transients due to stress changes

➢ **Biaxial Creep – anisotropy**

➢ **Transitions in Creep Mechanisms**

Biaxial Creep





Biaxial Creep

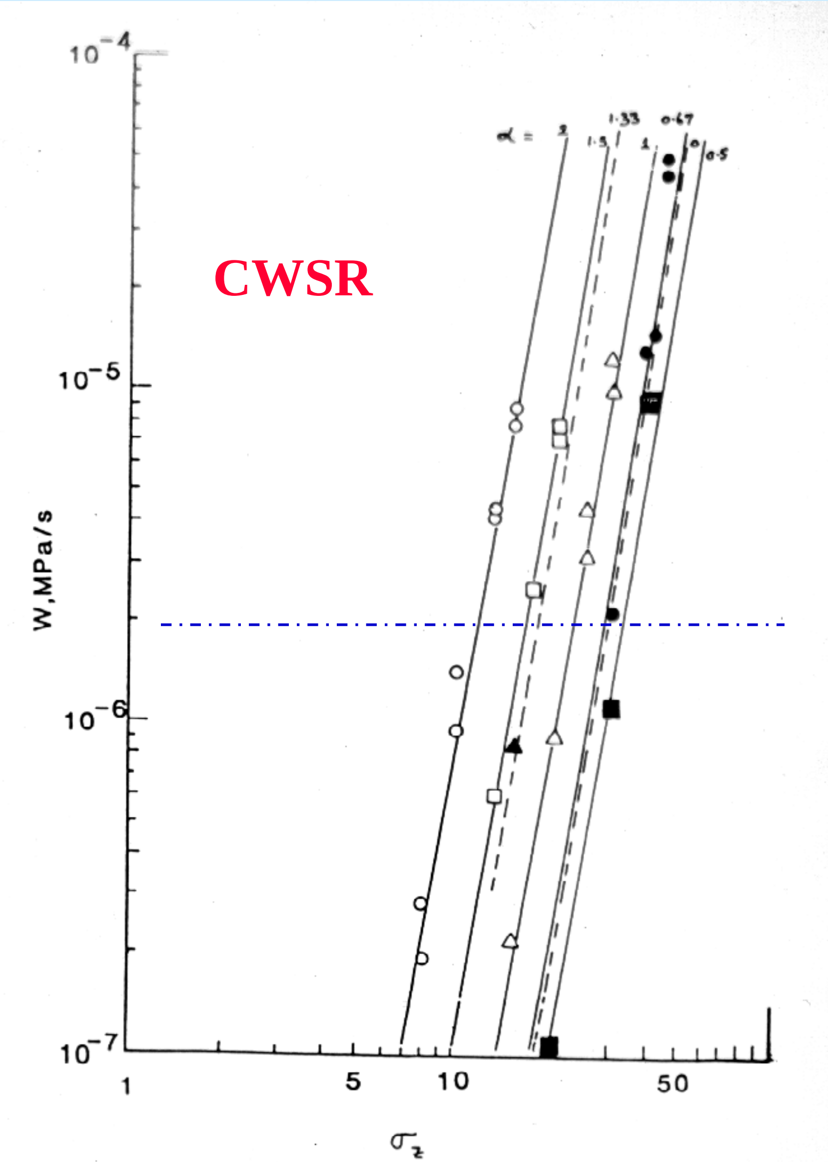
ε_θ, ε_z vs σ_θ, σ_z

σ_θ vs σ_z at constant W

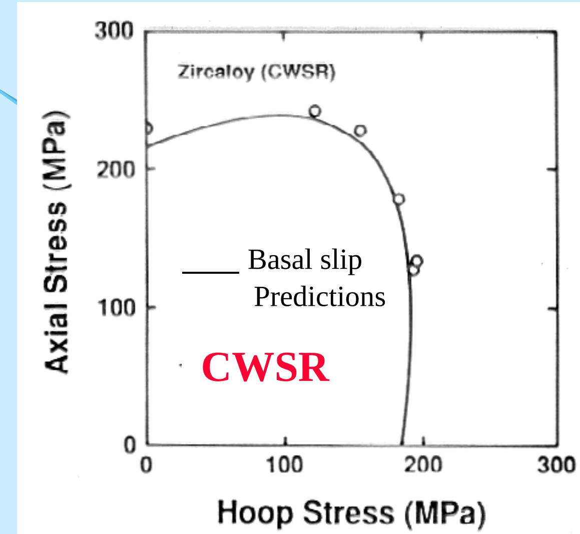
W= $\dot{\varepsilon}_\theta \sigma_\theta + \dot{\varepsilon}_z \sigma_z$ $\Leftarrow$ **Creep Locus**

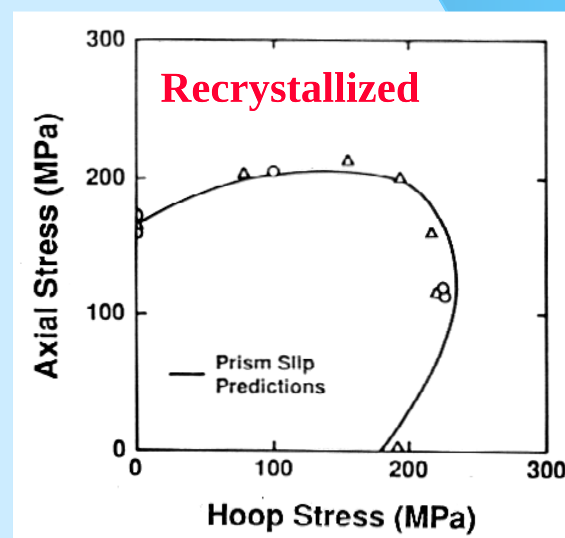
W=energy dissipation rate

Creep Loci



σθ vs σz at constant W





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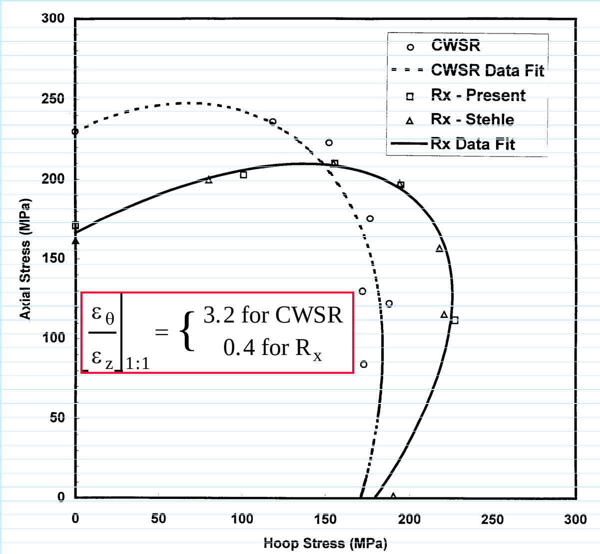
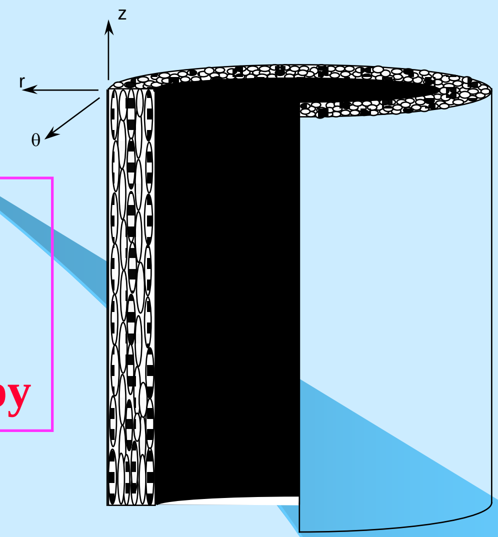
Zircaloys – Expt vs CODF-slip Model

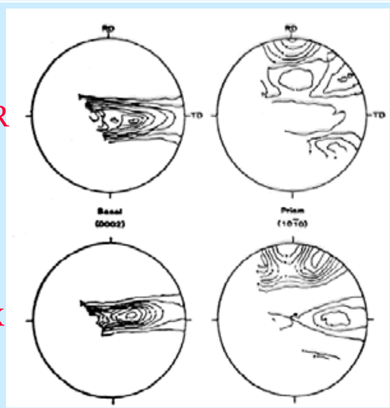


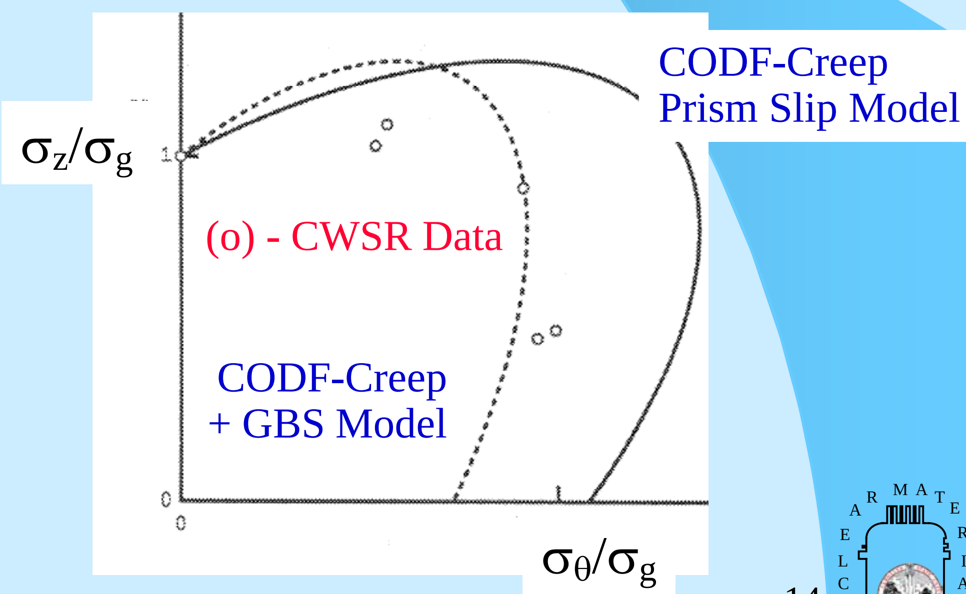
cold-work → texture & grain-shape anisotropy



CWSR

Rx



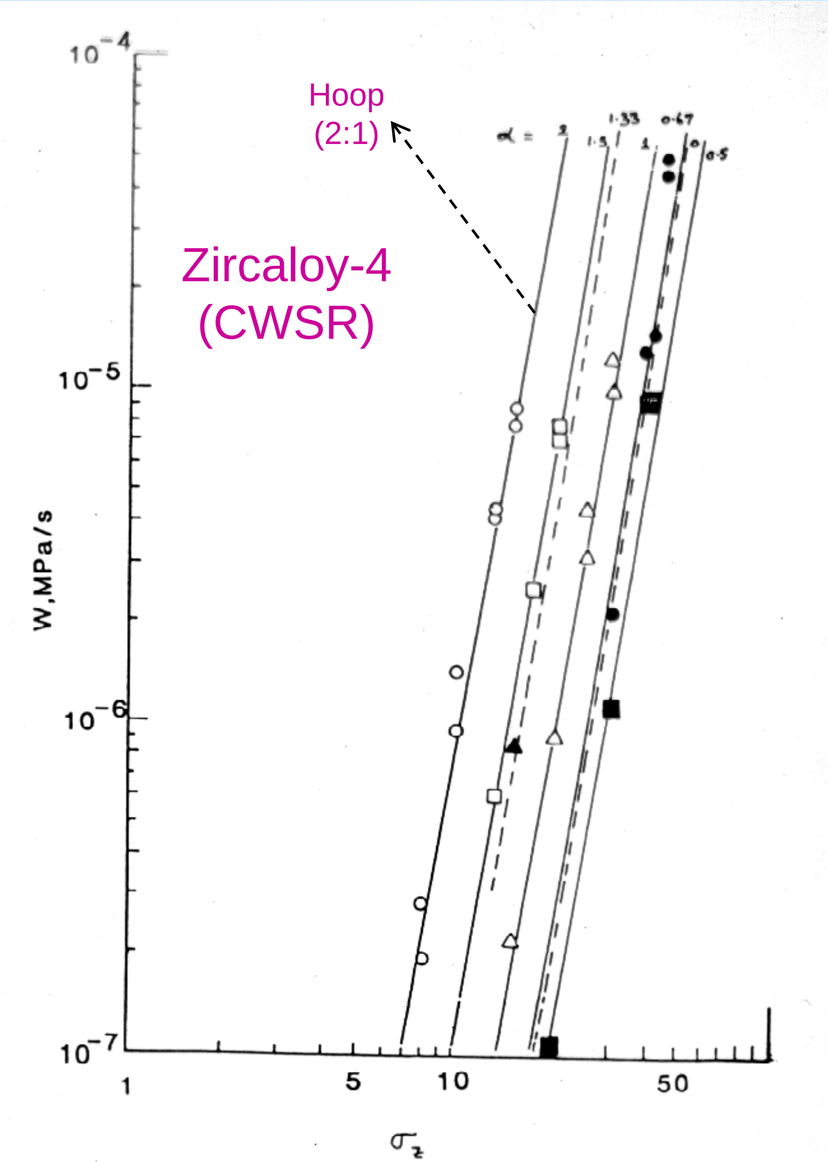


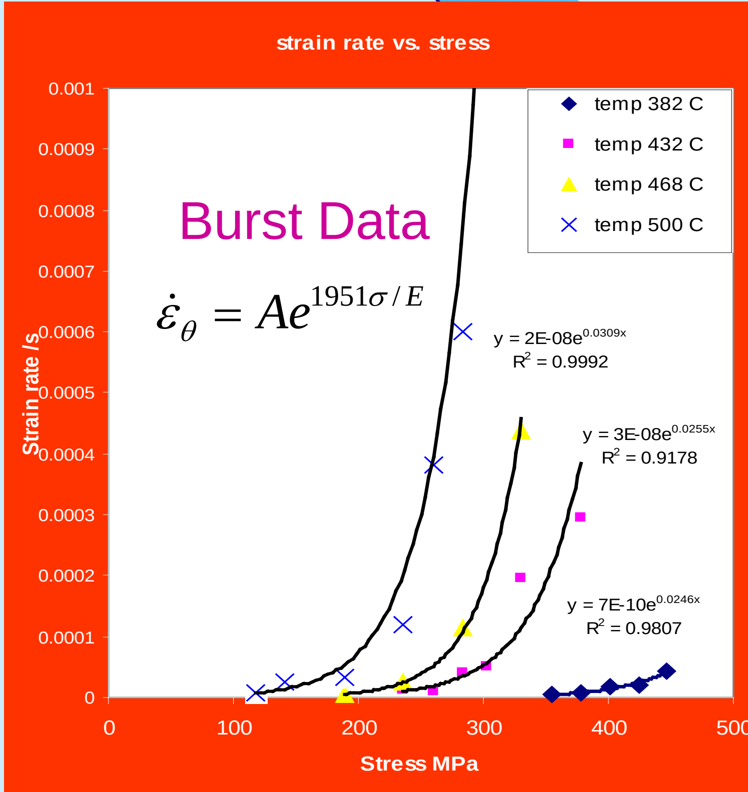
low c/a ratio - prism slip

Biaxial Steady-State Creep Results

$$\dot{\varepsilon}_\theta = Ae^{-60,900/RT}\left(\frac{\sigma_\theta}{E}\right)^5 \text{ for } \sigma < 10^{-3}E$$

$$\dot{\varepsilon}_\theta = 1.9x10^9 D_o e^{-\Delta H/kT}\left(\sinh 500\frac{\sigma}{E}\right)^5$$





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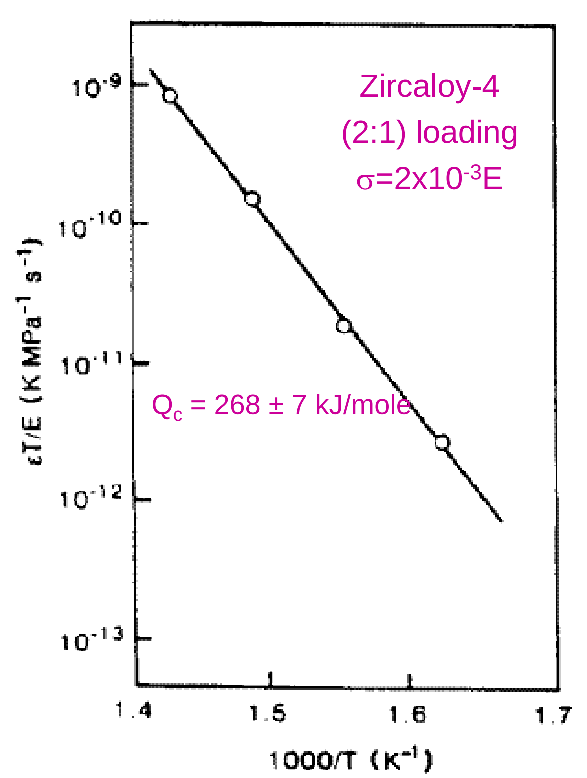
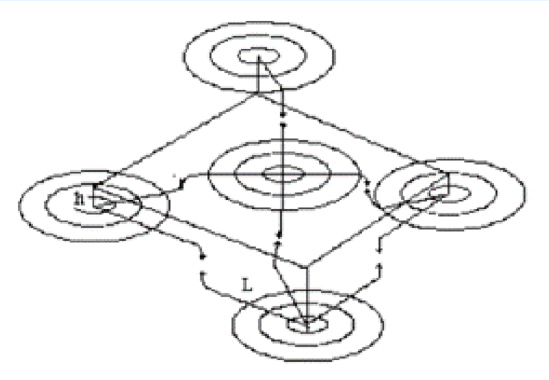
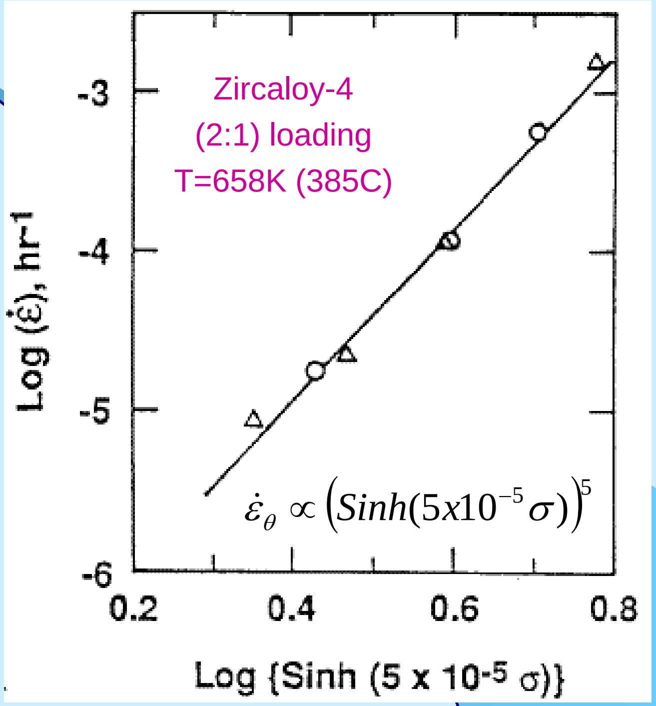
Temperature & Stress Variations of Secondary Creep-Rate





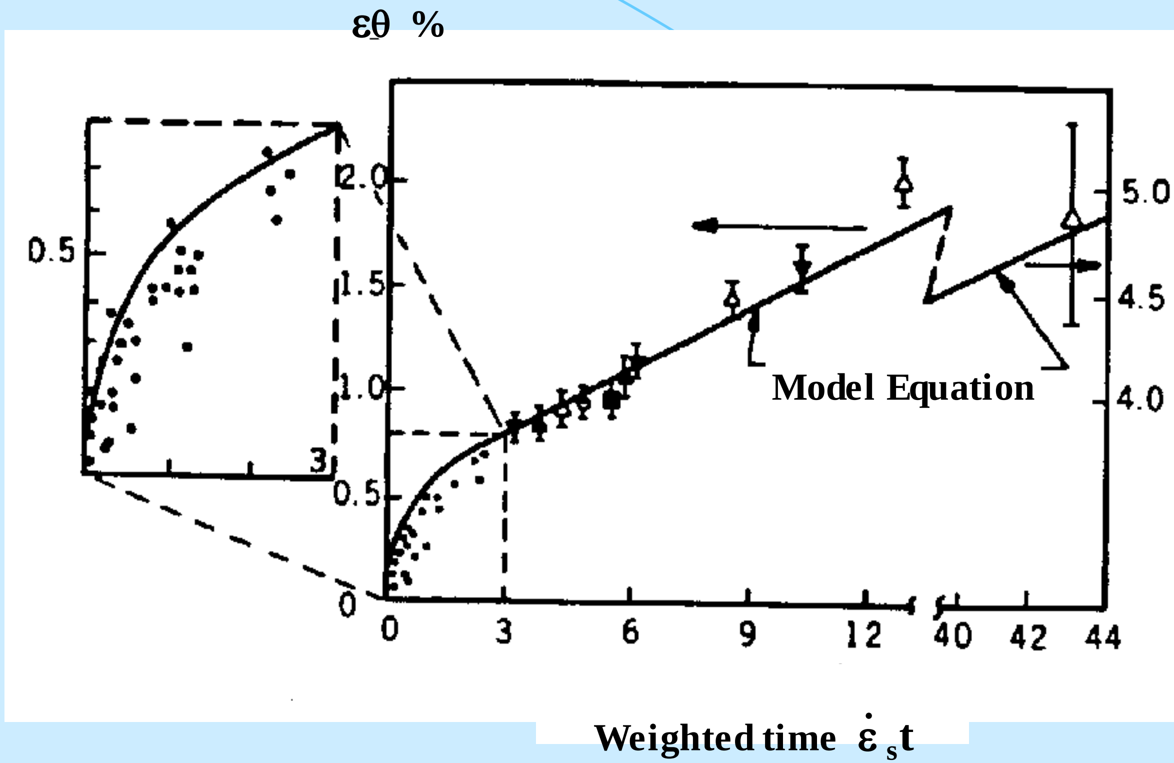


$Q_c = Q_D$ and n~5

Signifies that creep is **climb-controlled** during glide-climb process of dislocations such as is noted in **pure metals and Class-II alloys**.

[note that these are at relatively high stresses ($\sigma > 10^{-3}E$)]

Stress and Temperature Dependence
(internal pressurization – 2:1 tests)



$$\varepsilon = \frac{1+\alpha+\alpha\beta\,\dot{\varepsilon}t}{1+\alpha\beta\dot{\varepsilon}t}\,\dot{\varepsilon}t, \quad \text{where } \alpha = \frac{\dot{\varepsilon}_i}{\dot{\varepsilon}_s}, \; \beta = \frac{1}{\varepsilon_{tr}} \text{ and } \dot{\varepsilon} = f(\sigma, T)$$

$$\frac{\dot{\varepsilon}_s kT}{D_0 E b} = A e^{-\Delta H/kT}\left(\sinh B\sigma\right)^n$$

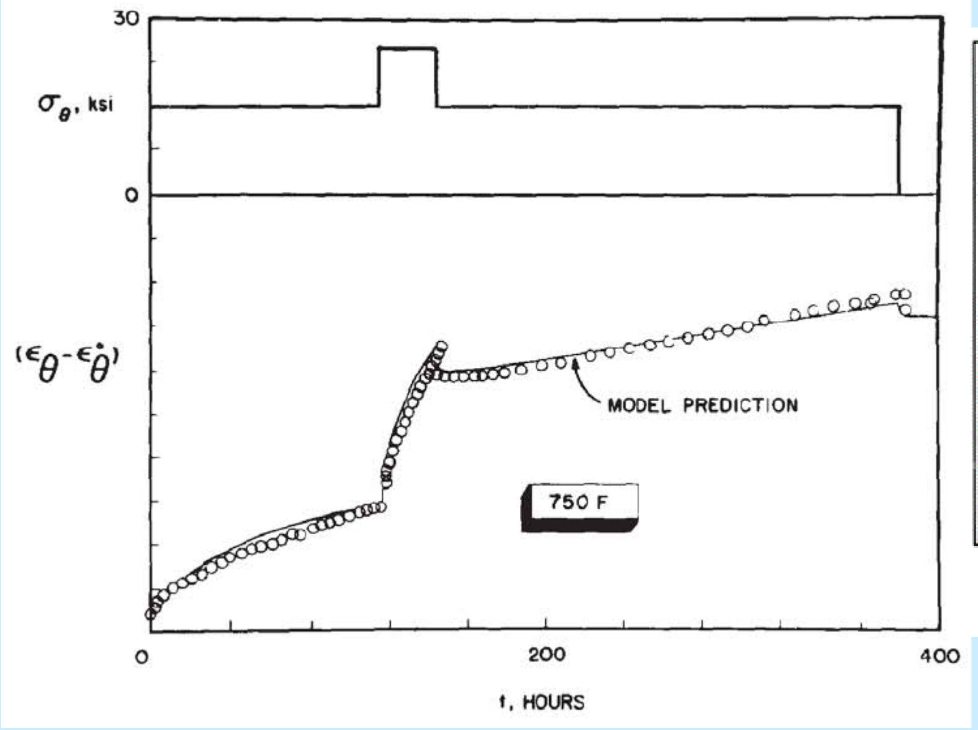
Transients in Creep due to Stress Changes

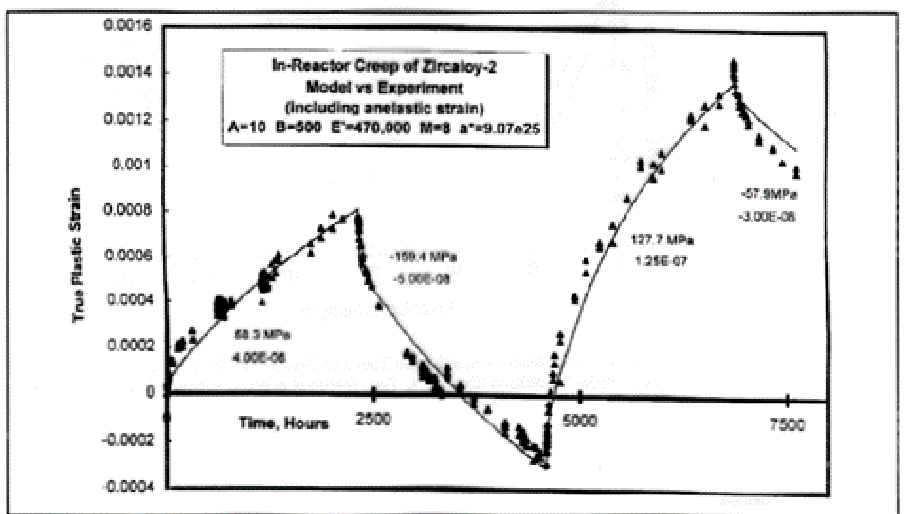
$$\varepsilon = \varepsilon_E + \varepsilon_a + \varepsilon_p$$

$$\varepsilon_E = \frac{\sigma}{E}, \quad E = E_{RT} - E'(T - T_{RT})$$

$$\varepsilon_p = \frac{1 + \alpha + \alpha\beta\,\dot{\varepsilon}t}{1 + \alpha\beta\,\dot{\varepsilon}t}\,\dot{\varepsilon}t, \quad \dot{\varepsilon} = f(\sigma, T)$$

$$\varepsilon_a = \frac{\sigma_f}{E'} - \left\{ \left(\frac{\sigma_f}{E'} - \frac{\sigma_o}{E'} \right)^{1-M} + (M-1)\alpha^*(t - t_o) \right\}^{-1/(M-1)}$$





NCSU Creep-Model predictions compared with in-reactor creep results
(KL Murty, NSF-UCSD/IMM-fellow/minisabbatical, Aug. 1995)

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Model Equations for Predicting In-Reactor Creep

Thermal Creep

Steady-State Creep-Rate: $\frac{\dot{\varepsilon}_s kT}{D_0 Eb} = Ae^{-\Delta H/kT}\left(\sinh B\sigma\right)^n$

Creep Strain: $\varepsilon = \frac{1+\alpha+\alpha\beta\dot{\varepsilon}t}{1+\alpha\beta\dot{\varepsilon}t}\dot{\varepsilon}t$, **where** $\alpha = \frac{\dot{\varepsilon}_i}{\dot{\varepsilon}_s}, \beta = \frac{1}{\varepsilon_{tr}}$ *and* $\dot{\varepsilon} = f\left(\sigma, T\right)$

Effect of Stress Changes: $\varepsilon_a = \frac{\sigma_f}{E'} - \left\{\left(\frac{\sigma_f}{E'} - \frac{\sigma_o}{E'}\right)^{1-M} + (M-1)\alpha^*(t-t_o)\right\}^{-1/(M-1)}$

Generalized Stress & Strain - Rate

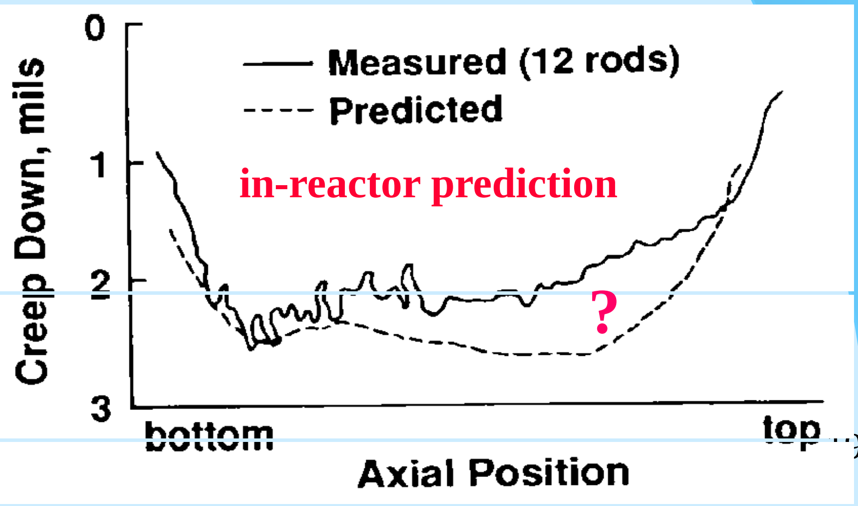
$$\sigma_g = f\{\sigma_\theta, \sigma_z, R, P\}$$

$$\dot{\varepsilon}_g = f'\{\sigma_g, T\}$$

Radiation Creep & Growth

$$\varepsilon_g^{irr} = B'\phi\sigma_g, \qquad \sigma_g = f\left\{\sigma_\theta, \sigma_z, R, P\right\}$$

$$\varepsilon_z^{sf} = B''\phi t$$



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EFFECTS OF NEUTRON IRRADIATION

Deformed Irradiated Recrystallized Zircaloy-2

(c+a) type Dislocations

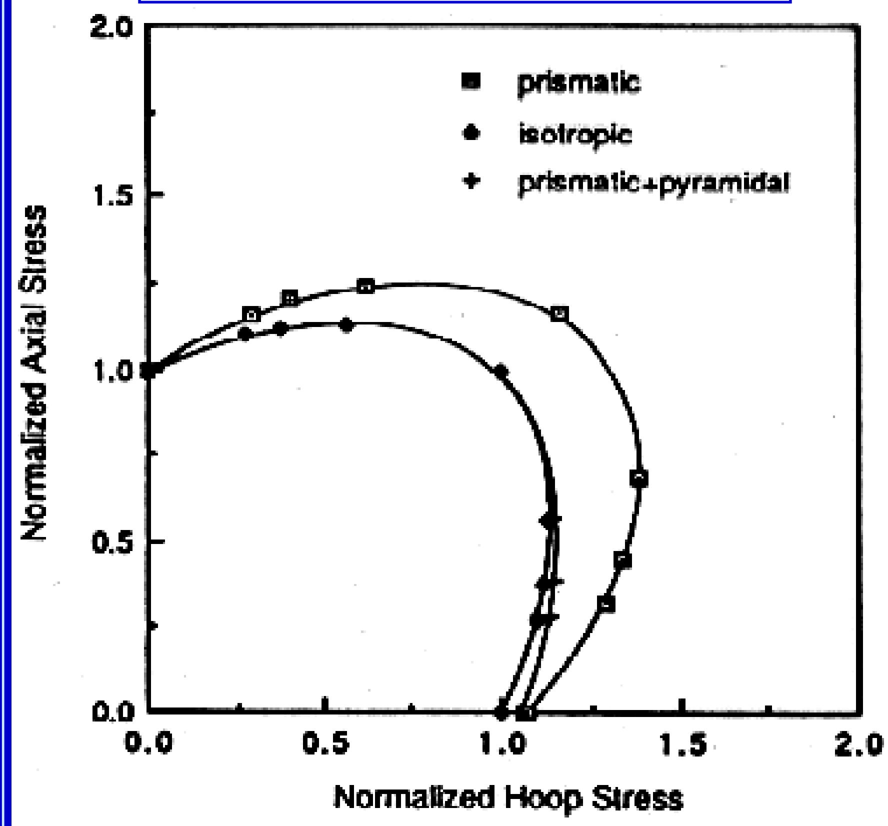
Basal + Pyramidal slip in addition to **prismatic**

Deformation and Creep Anisotropy Decreases

The creep anisotropy of CWSR Zircaloy **unaffected** by irradiation

PIE experiments on Oconee

CODF-CREEP Model



Rx - Decreased Anisotropy

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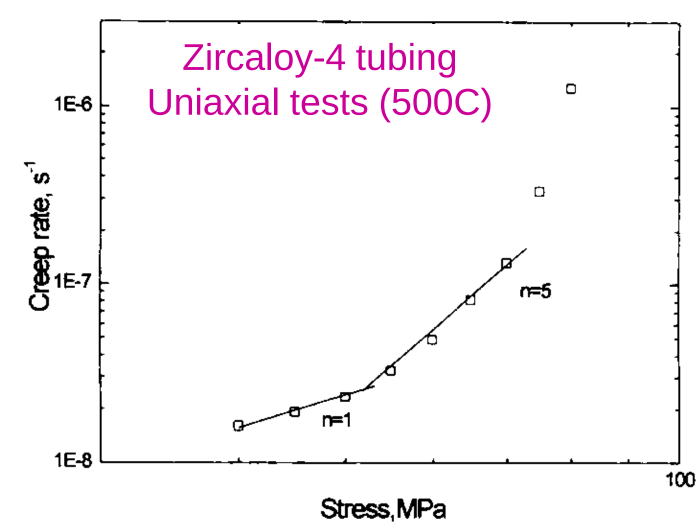
Transitions in Creep Mechanisms

- Most of the hoop data of internally pressurized tubing are at relatively high stresses
- Transitions in creep mechanisms are expected as lower stresses are encountered due mainly to GBS and diffusional creep (Nabarro-Herring and/or Coble)

$$\dot{\varepsilon} = A\frac{\sigma^{n}}{d^{p}}e^{-Q_c/RT}$$

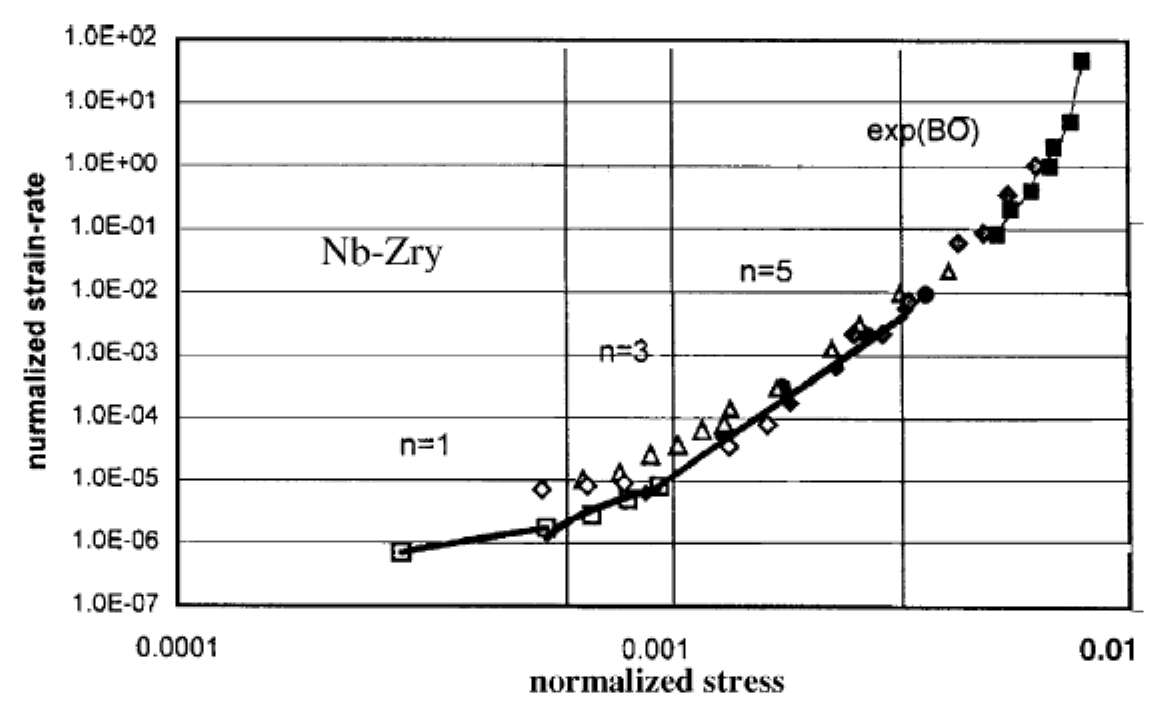
Mechanism	n	p	Q_c	comment
Climb	5	0	Q_D	Pure metals and class-M (class-II) alloys
Glide	3	0	Q_s	Class-A (class-I) alloys
Nabarro-Herring	1	2	Q_D	At high- temp, low σ small d
Coble	1	3	Q_b	At low-temp, low σ and very small d
Superplasticity (GBS)	2	2	Q_D	Intermediate σ and small d

Wang, PhD thesis, NCSU



Nb-Modified Zircaloy-4 Tubing Hoop Creep (2:1)

•Zircaloy-4 +1%Nb (similar to Zirlo)



$Region\ I: 3\times10^{-4} < \sigma/E < 6\times10^{-4}$
Coble Creep

$Region\ II: 6\times10^{-4} < \sigma/E < 1\times10^{-3}$
Dislocation glide controlled creep

$Region\ III: 1\times10^{-3} < \sigma/E < 4\times10^{-3}$
Dislocation climb controlled creep

$Region\ IV: \sigma/E > 4\times10^{-3}$
Power – law breakdown

Alloy class (Class-I) behavior due to locking of dislocations by solute atoms (known as microcreep with n~3) –

at higher stresses dislocations break-away from locking leading to climb controlled creep similar to pure metals (or class-M alloys)

Y. Zhou, B. Devarajan and K.L. Murty, Nucl. Eng. Design, 228 (2004) 3-13

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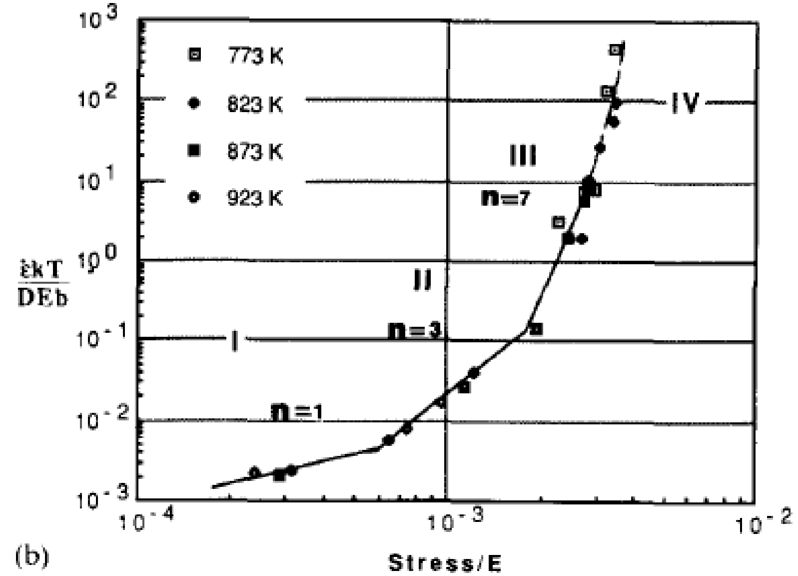
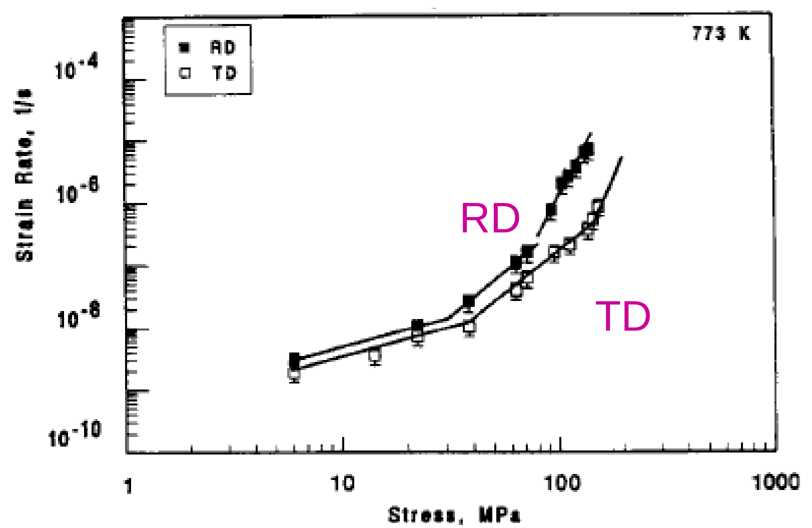
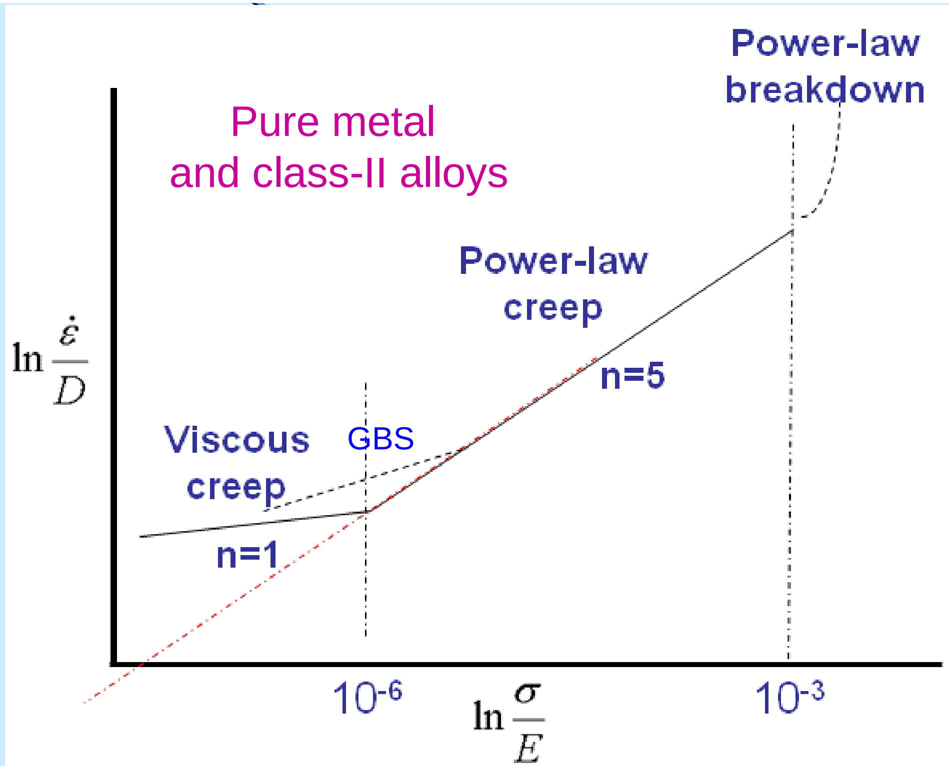
Nb-Modified Zircaloy-4 sheet Uniaxial Creep



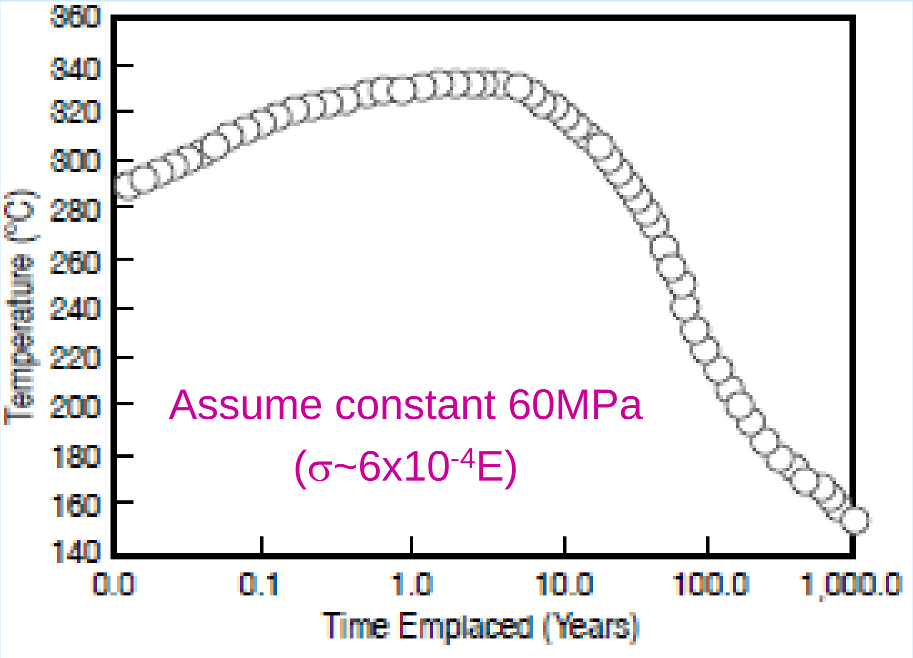
J. Ravi, Wiratmo and K.L. Murty, Nucl. Eng. Design, 156 (1995) pp. 359 – 371

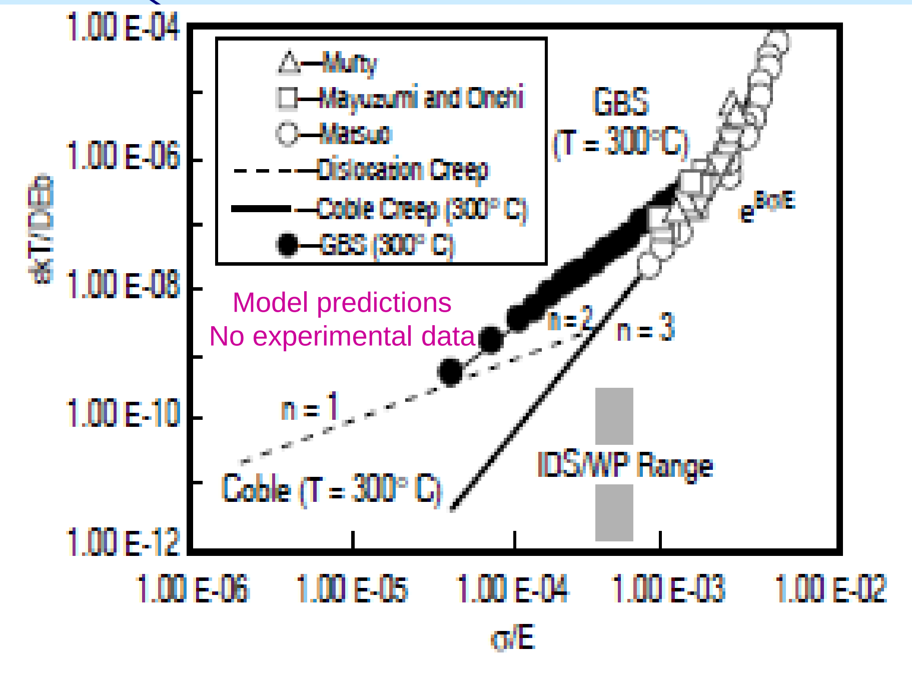


Dangers of blind extrapolation to low stresses



Creep Strain Calculation (inert dry storage conditions)





$$\varepsilon_{total}=\varepsilon_{diffusion}+\varepsilon_{GBS}+\varepsilon_{dislocation}$$

Time Hardening – easier

Strain Hardening – more involved but more realistic

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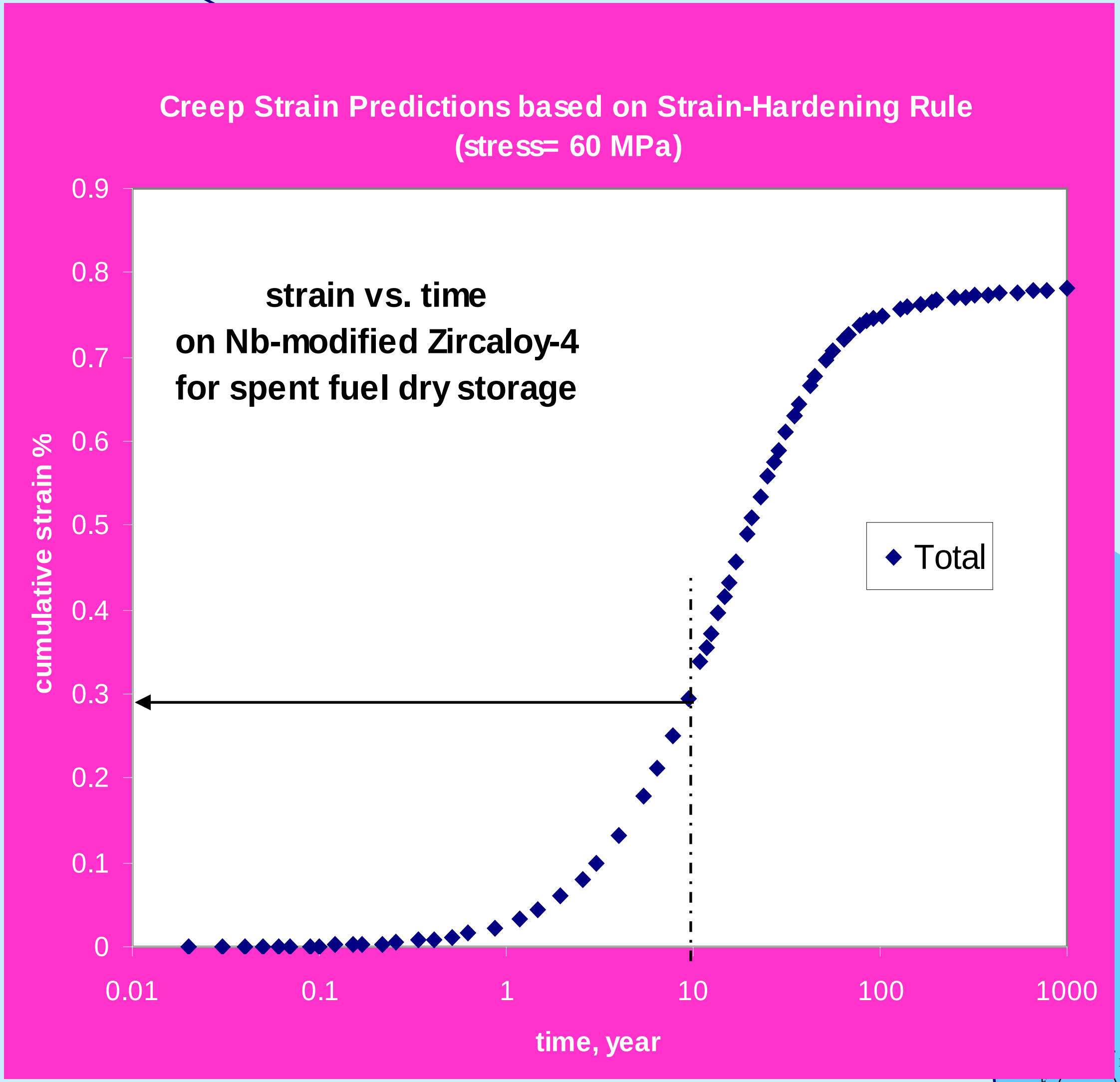
Creep Strain Calculations

Prediction

10-years storage

ε~ 0.3 %

(close to ANL-PNNL results, 2002)



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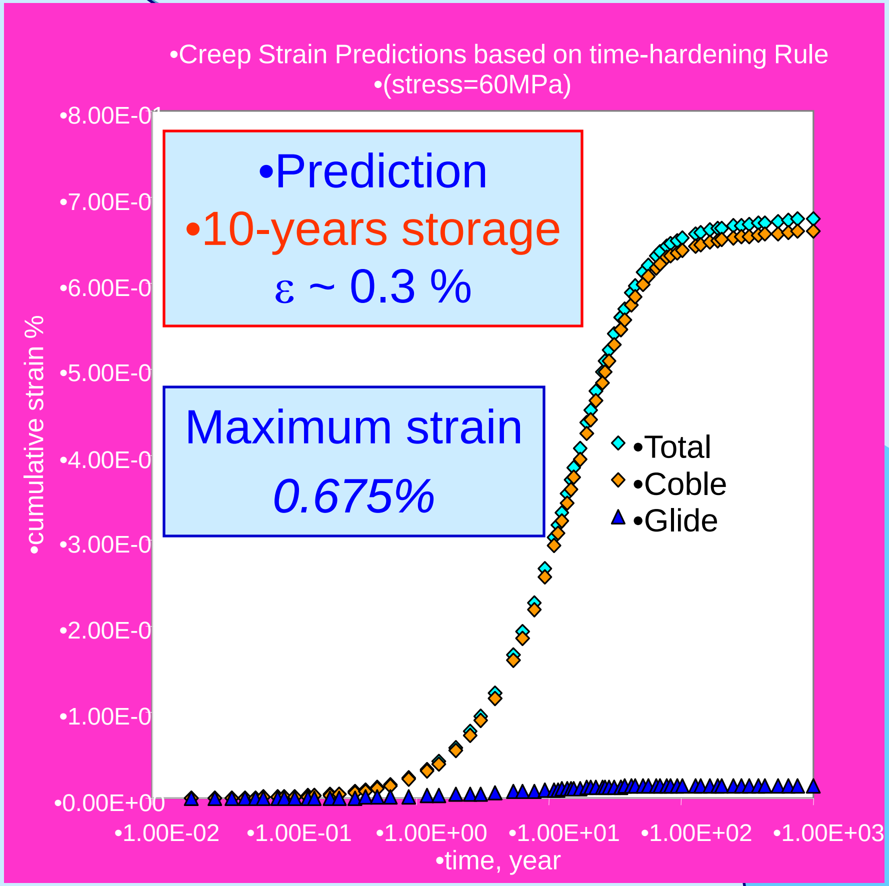
Creep Strain Calculations

- Only thermal creep considered
- Need more concrete data on transitional creep mechanisms in Zircaloys
  (as per recent study on Ti alloy – next slide)
- Effect of radiation – expected to make the material harder and thus these predictions are considered conservative
- Effects of oxidation, hydriding and crud – very limited amount of work to-date
- **Ideal is to perform tests on UNF from cooling pools – small unihoop type suitable in a hot cell**



Further research for proper characterization of transitions

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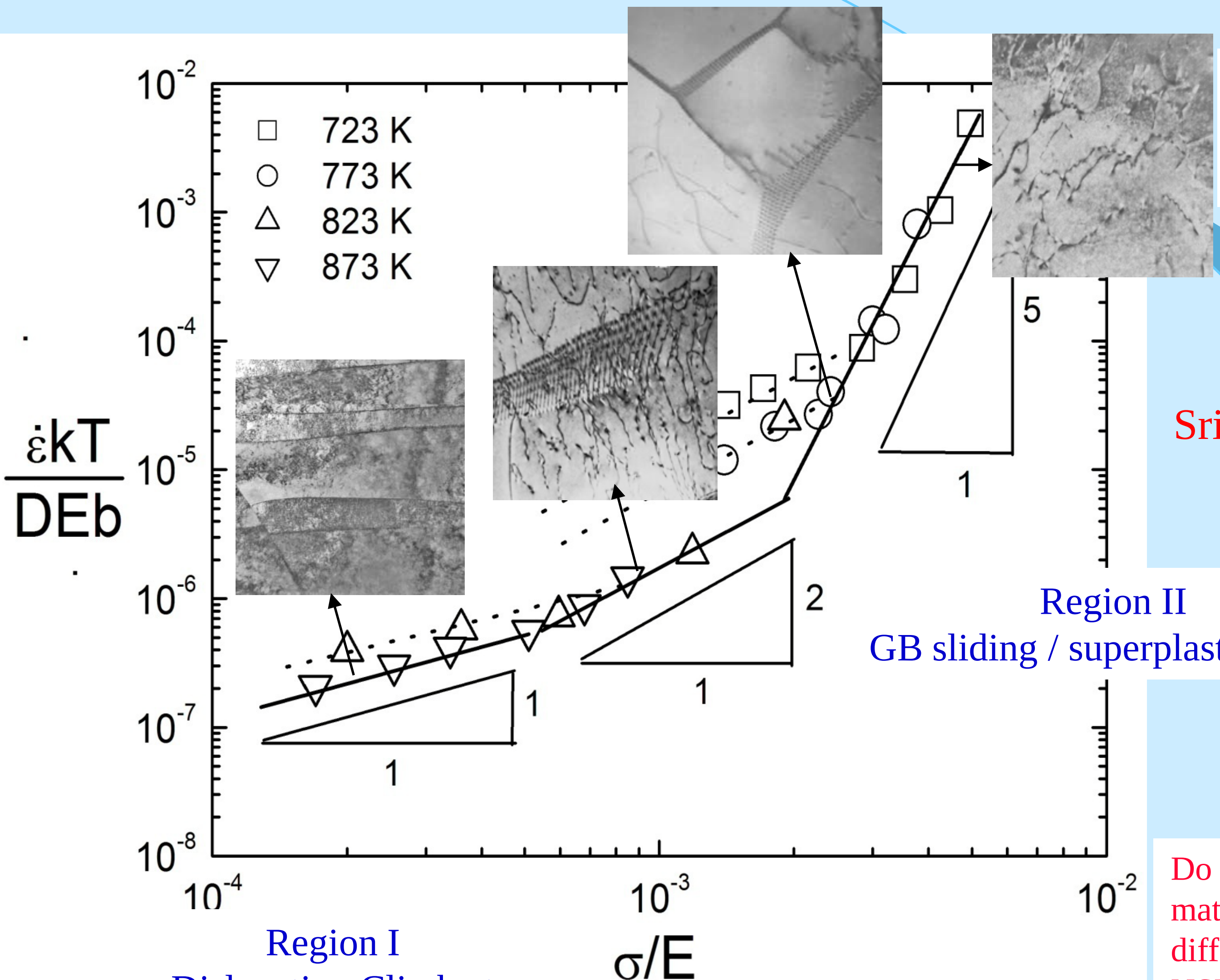
Ti3Al2.5V – Transitional Creep Mechanisms



Region III
Jogged Screw Dislocation
Climb of Edge Dislocations

Srikant et al, Acta Mat, 2008

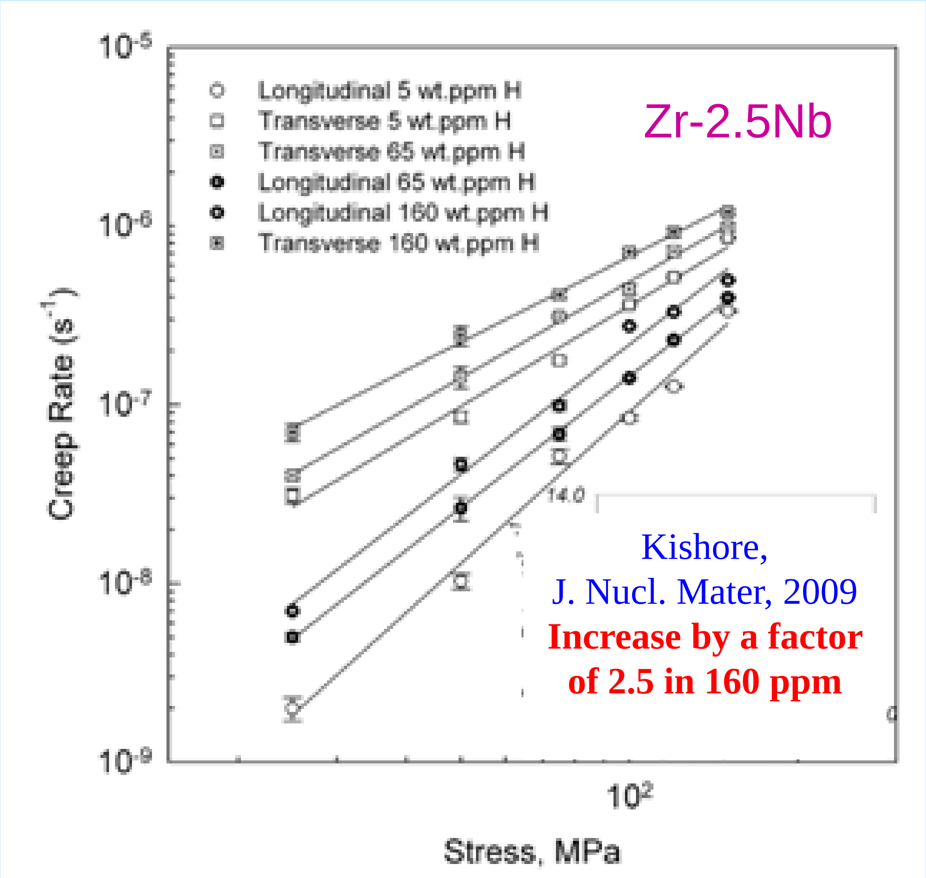
Region II
GB sliding / superplastic creep

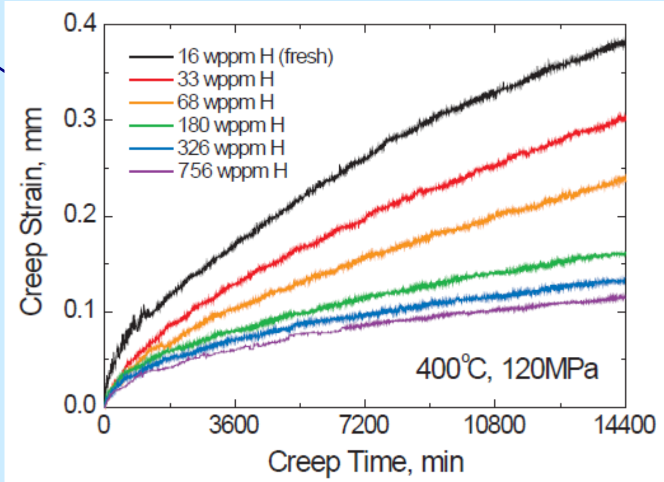
Region I
Dislocation Climb at
slip-band – GB intersections

Do these transitions occur in other materials such as Zr and under different loading conditions? – new NSF grant (9/15/2010 – 2014)

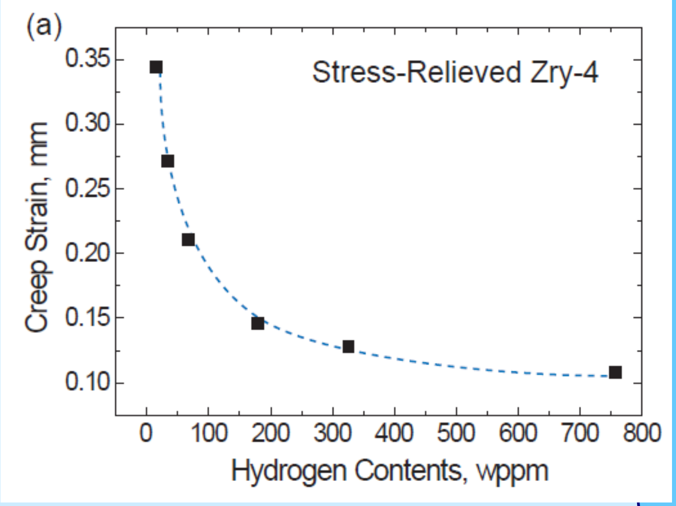
Effect of Hydrogen on Creep of Zircaloys

- Depends on atomic hydrogen vs hydride
- Presence of oxide layer
- CWSR vs Rx
- Alloy composition
- Effect of neutron irradiation (?)





Y. Jung et al, J. Nucl Mater, 2011



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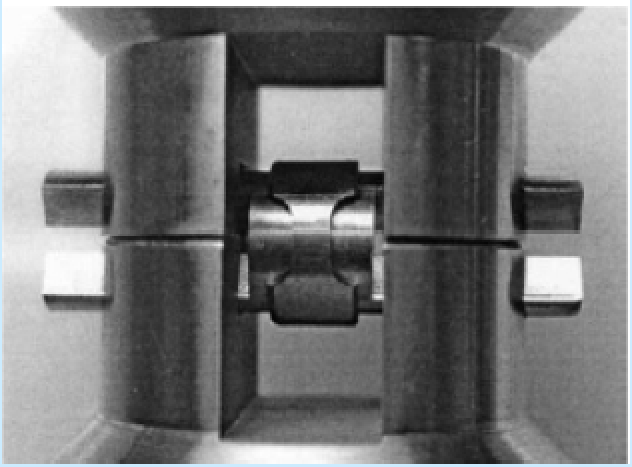
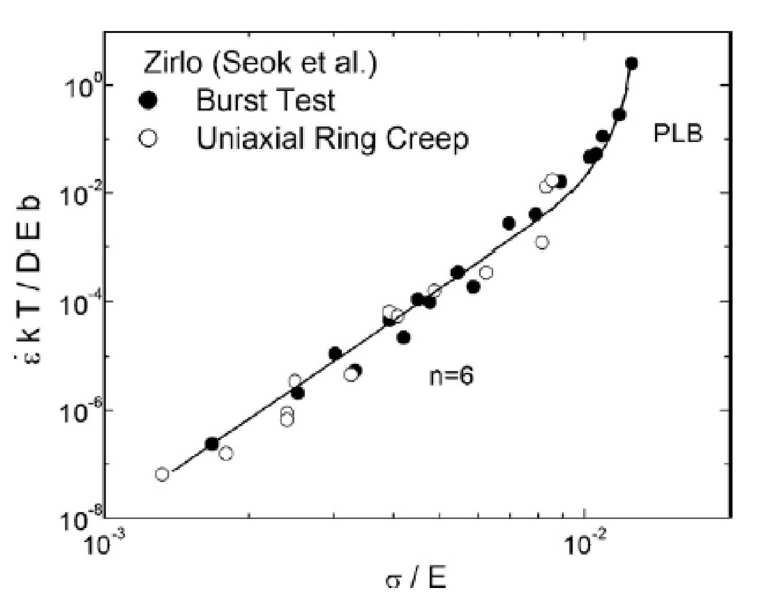
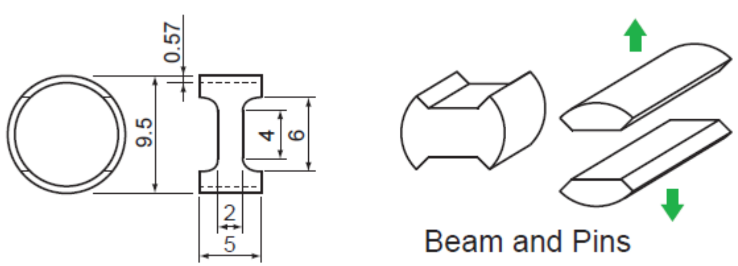
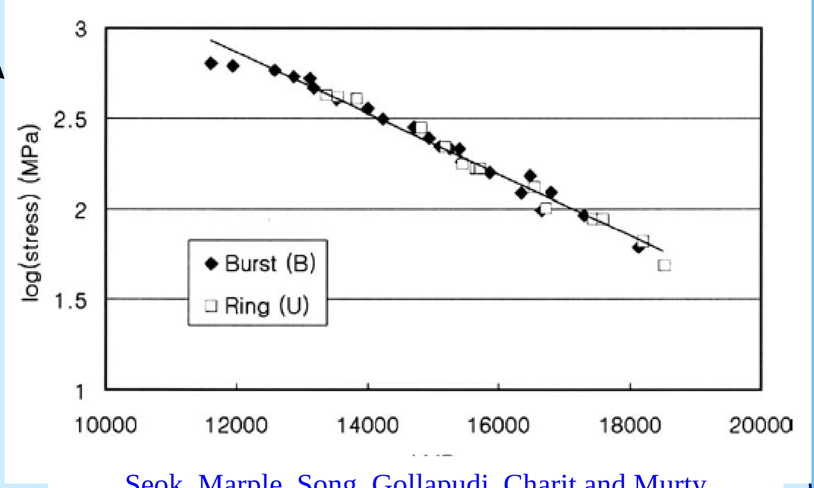
Possible study on UNF
using uni-hoop creep tests





Equivalence between ring tensile and burst tests





Seok, Marple, Song, Gollapudi, Charit and Murty
Nucl Eng Design, 2011

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Relevant NCSU Publications

K.L. Murty and B.L. Adams, "Biaxial Creep of Textured Zircaloy: Part I - Phenomenological and Experimental Descriptions," Mat. Sci. Eng., 70, 169-180 (1985)

B.L. Adams and K.L. Murty, "Biaxial Creep of Textured Zircaloy: Part II - Crystal-Mean Plastic Modeling," Mat. Sci. Eng., 70, 181-190 (1985)

K.L. Murty, "Biaxial Creep of Zircaloy Cladding," Trans. IIM, 39, 357-368 (1986)

P. S. Godavarti and K.L. Murty, "Creep Anisotropy of Recrystallized Zircaloy-4 TREX Using Impression Tests," Proceedings of 9th International Conference on STRUCTURAL MECHANICS in REACTOR TECHNOLOGY (1987), paper C3/4

R.R. Kola, S.T. Mahmood and K.l. Murty, “Creep and Yield Loci of Zircaloy Cladding at Elevated Temperatures,” Textures and Microstructures, vol. 7 (1987) pp. 211-226

W. L. Daugherty and K.L. Murty, "Application of Texture in Predicting Nuclear Fuel Cladding Creep," Nucl. Technology, 80, 443-450

K.L. Murty, "Prediction of In-Reactor Performance of Zircaloy Cladding in PWRs," Proceedings of the International Conference on 'Residual Life of Power Plant Equipment - Prediction and Extension,' LIPREX-89, Jan. '89, paper 7A.2

S.T. Mahmood and K.L. Murty, "Anisotropic Biaxial Creep of Zircaloy Cladding: Effects of Recrystallization and Neutron Irradiation," Proceedings of 10th International Conference on SMiRT, Aug. 1989, paper C

K.L. Murty, “Texture-Based Plasticity Modeling of Biaxial Creep of Zircaloy Cladding: Application to In-Reactor Prediction,” in Proceedings of the International Conference on Physics of Irradiation Effects in Metals (PM’91), Siofok, Hungary, May 20 – 24

J.Ravi, S.T.Mahmood and K.L. Murty, “High Temperature Creep of Nb-Modified Zircaloy,” in Proceedings of the 11th International Conference on Structural Mechanics in Reactor Technology (SMiRT-11), Tokyo, Japan, Aug.18-23 (1991). paper C03/1

K.L. Murty, “Biaxial Creep Behavior of Textured Zircaloy Tubing,” Journal of Metals, Feb. pp.49-55

K.L. Murty, B.V. Tanikella and J.C. Earthman, “Effect of Grain Shape and Texture on Equi-Biaxial Creep of Stress Relieved and Recrystallized Zircaloy-4,” Acta Met., vol. 42 (1994) pp. 3653-3661

J. Ravi, Wiratmo and K.L. Murty, “Transitions in Creep Mechanisms and Creep Anisotropy in a Zr-1Nb-1Sn-0.2Fe Sheet,” Nuclear Engineering and Design, 156 (1995) pp. 359 – 371

K.L. Murty, “Zircaloy Life-Prediction and New Generation Zircaloys for LWRs,” Trans. Ind. Inst. Metals, vol. 50 (1997) pp. 533-562

K.L. Murty, “Creep Studies for Life-Prediction in Water Reactors, “ Journal of Metals, Oct. 1999, pp. 32-39

K.L. Murty, “Internal Pressurization Creep of Thin-Walled Tubing of Zr-Alloys for Dry Storage Feasibility of Nuclear Spent Fuel,” J of Metals, Sep. 2000, pp. 34-38

Y. Zhou, B. Devarajan and K.L. Murty, “Short-Term Rupture Studies of Zircaloy-4 and Nb-Modified Zircaloy-4 Tubing using Closed-End Internal Pressurization,” Nucl. Eng. Design, 228 (2004) 3-13

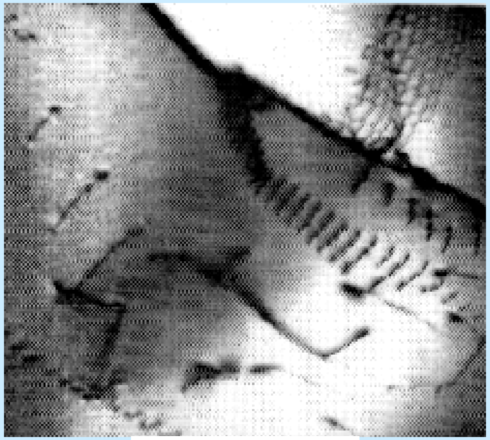
I. Charit and K.l. Murty, “Creep Behavior of Niobium-Modified Zirconium Alloys,” Journal of Nuclear Materials, 374 (2008) 354-363

C.S. Seok, B. Marple, Y.J. Song, S. Gollapudi, I. Charit and K.L. Murty, “High Temperature Deformation Characteristics of Zirlo™ Tubing via Ring Creep and Burst Tests,” Nuclear Engineering and Design, 241 (2011) 599-602

S. Gollapudi, I. Charit and K.L. Murty, “Creep Mechanisms in Ti-3Al-2.5V Tubing Deformed under Closed-end Internal Gas Pressurization,” Acta Mat., 56 (2008) 2406-2419.

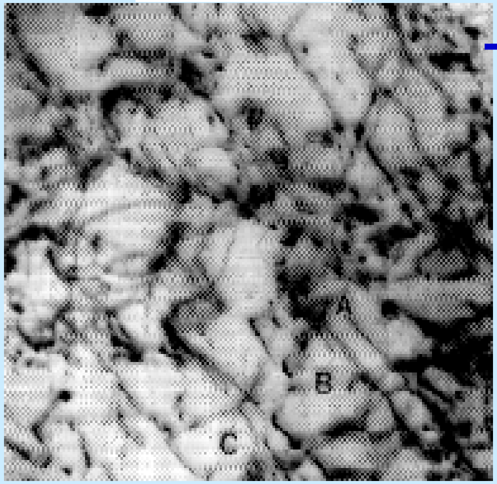
S. Gollapudi, V. Bhosle, I. Charit and K. L. Murty, “Newtonian Viscous Creep in Ti3Al2.5V,” Phil Mag., 88:9 (2008) 1357-1367.

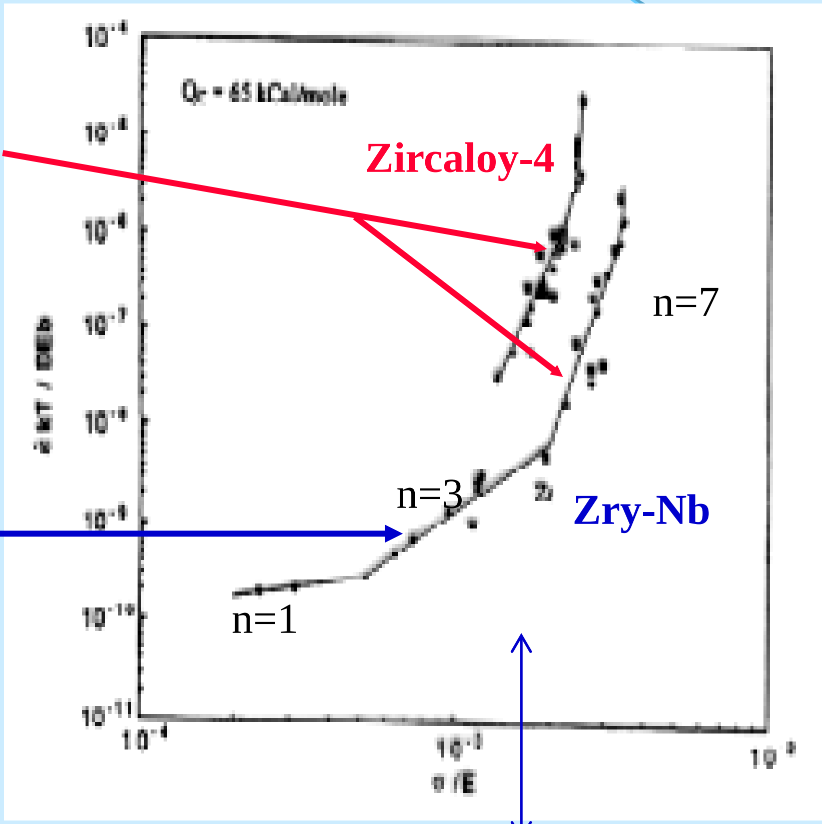
Transitional Creep Mechanisms



Class-M

Class-A



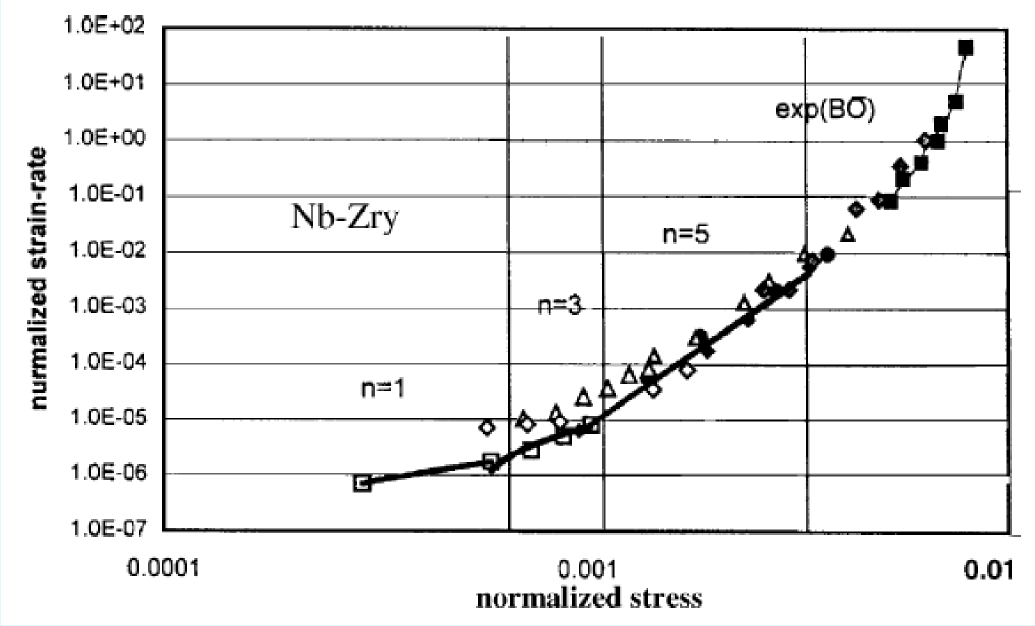


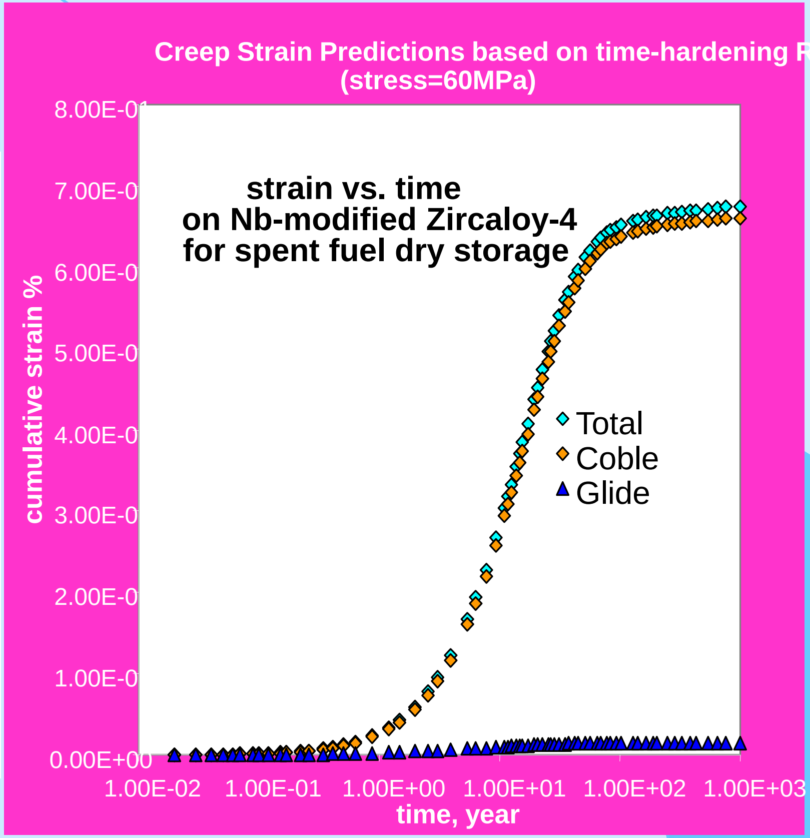
$$\sigma_c = \frac{W_m^2 c_o}{2^{\beta} kTb^3}$$

*** Class-A Alloys ***
Dislocation Break-away at high stresses
Murty (1973)

Transitional Creep Mechanisms
Application to Dry-Storage of SNF

Maximum strain : ***0.675%***





Nucl. Eng. Design, 228 (2004) 3-13

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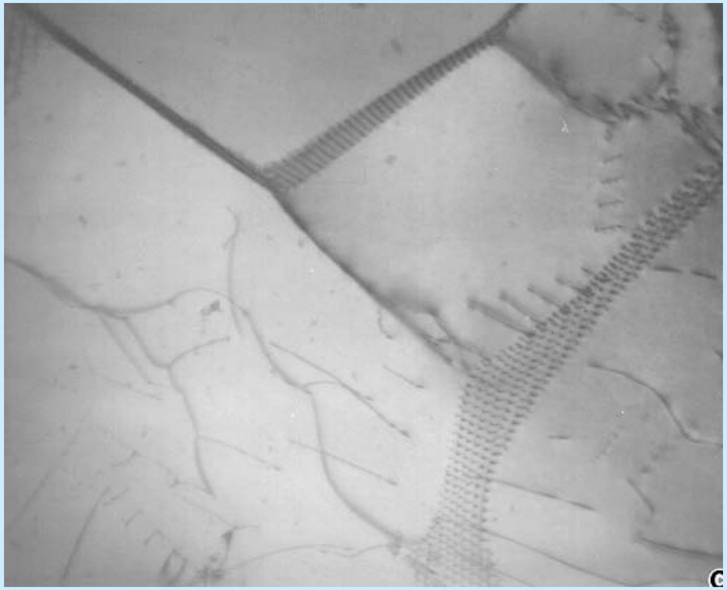
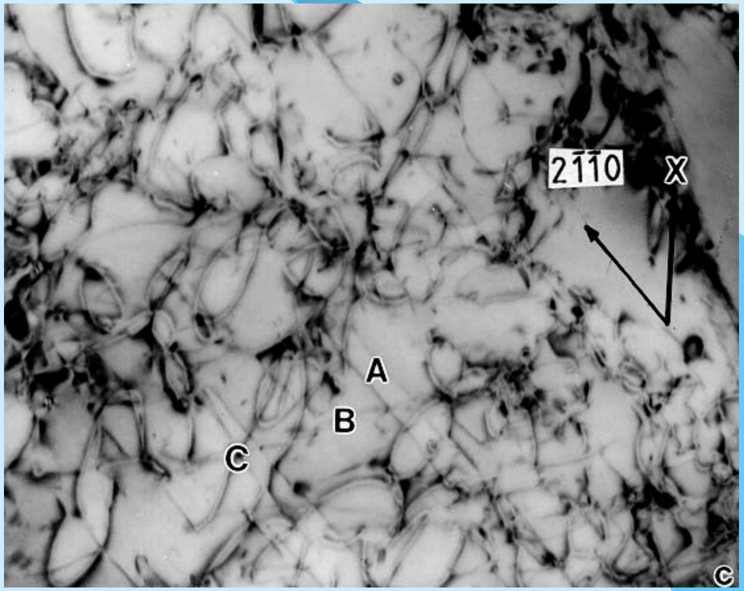
Dislocation Microstructures
Influence of Alloying / Stress-State

cp-Ti vs Ti3Al2.5V

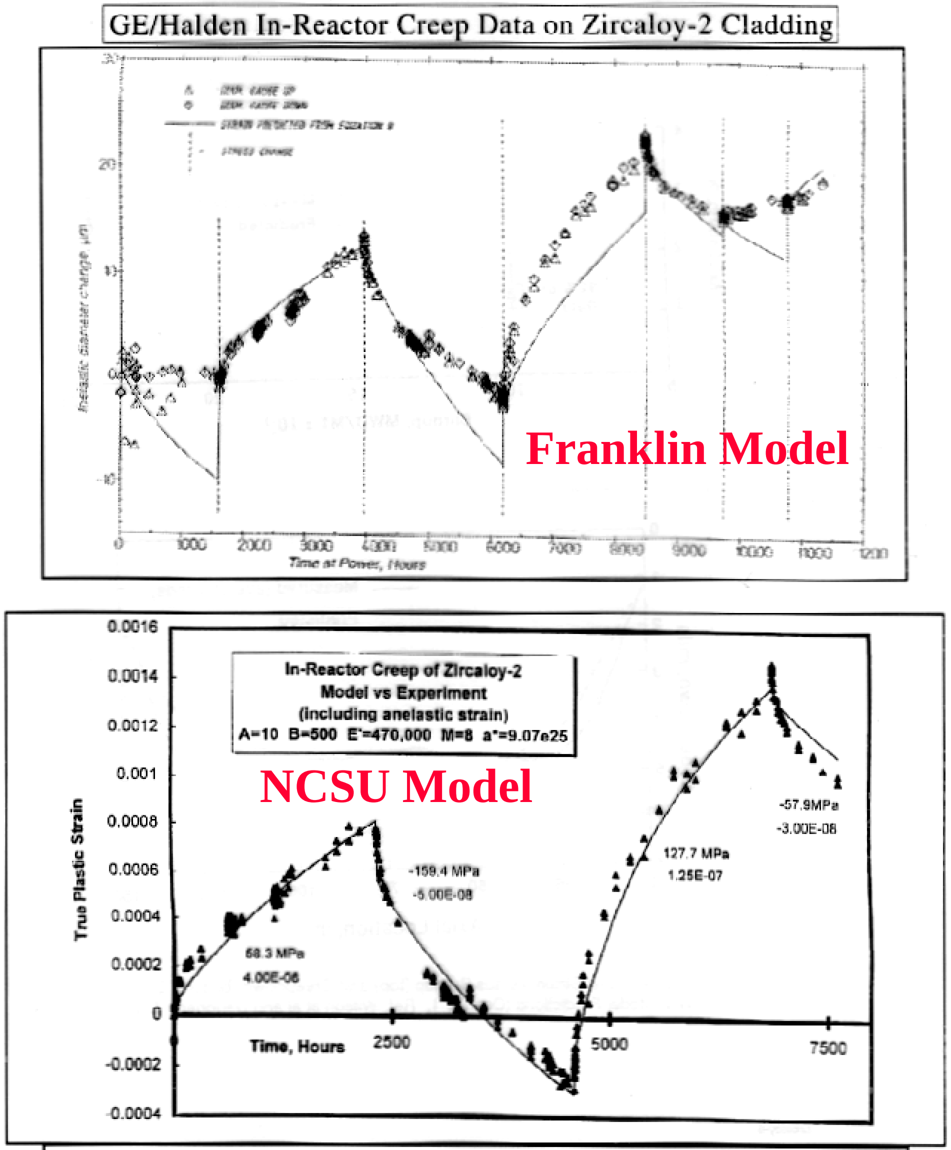




Biaxial Loading vs Uniaxial Loading

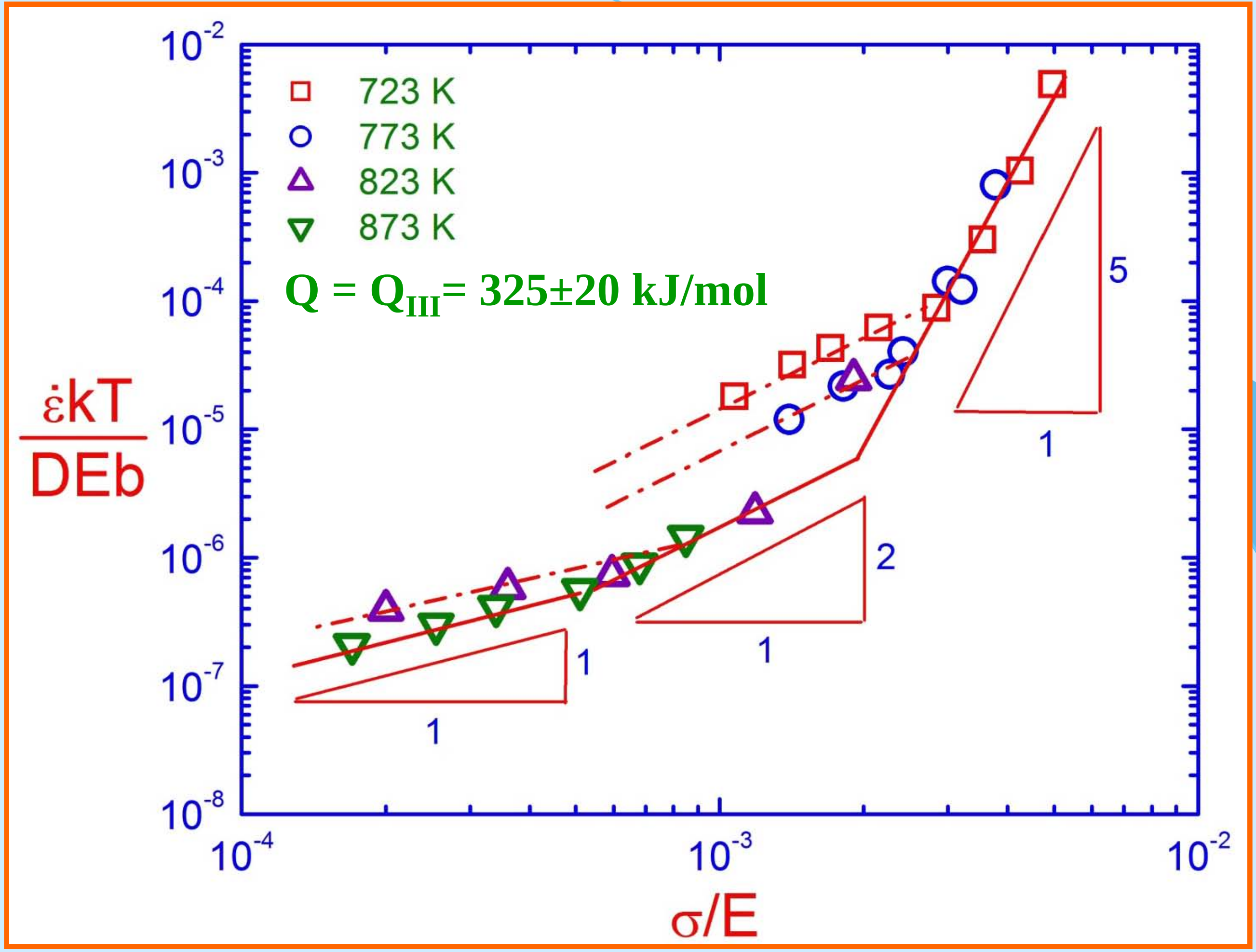
need further investigation

Transients in Creep - Irradiated Zry-2



NCSU Creep-Model predictions compared with in-reactor creep results
(KL Murty, NSF-UCSD/IMM-fellow/minisabbatical, Aug. 1995)

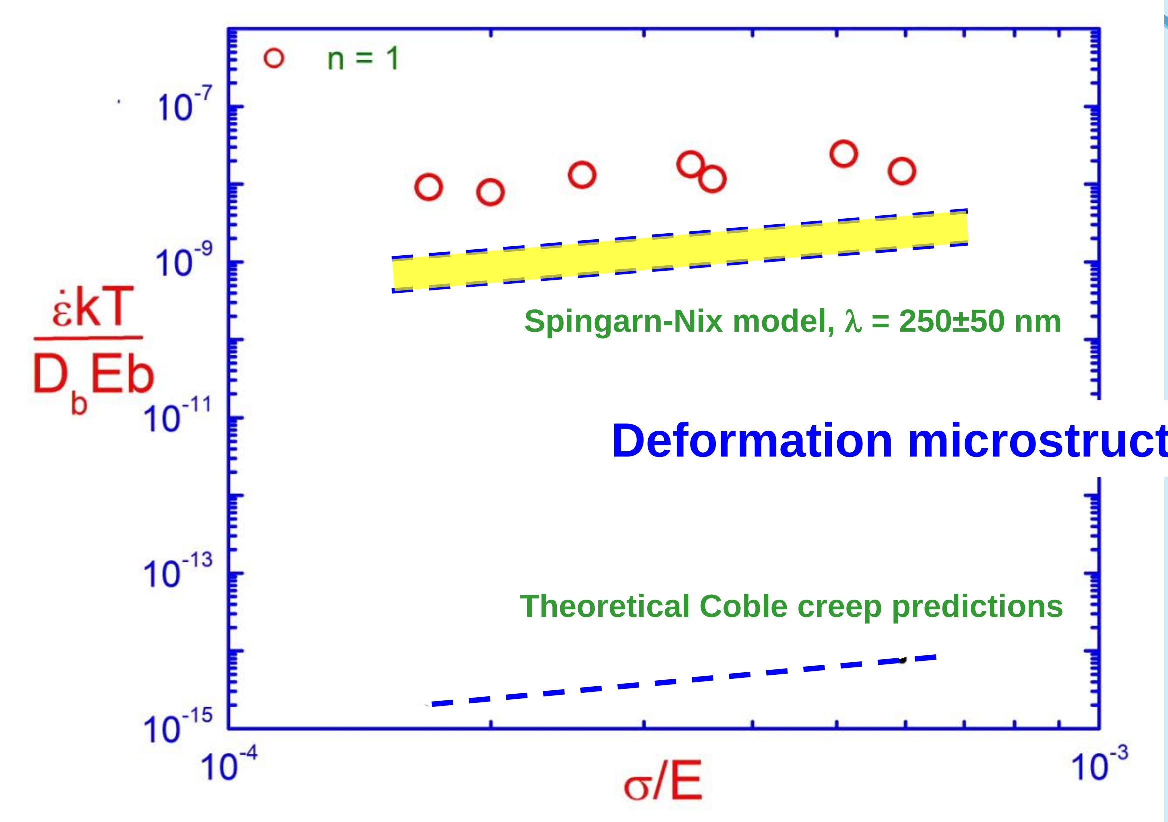
Ti3Al2.5V



Dorn Plot

Acta Mat. 56 (2008) 2406-2419

The Region I: Correlation with Coble Creep Predictions

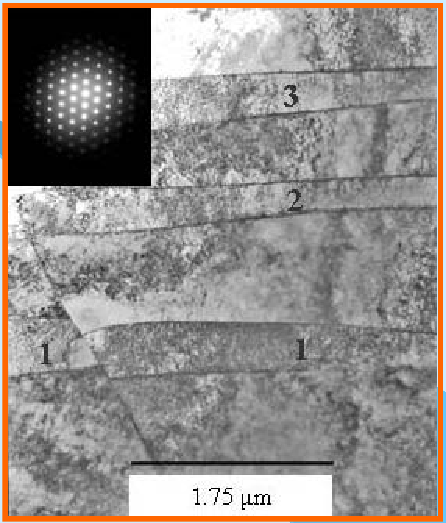


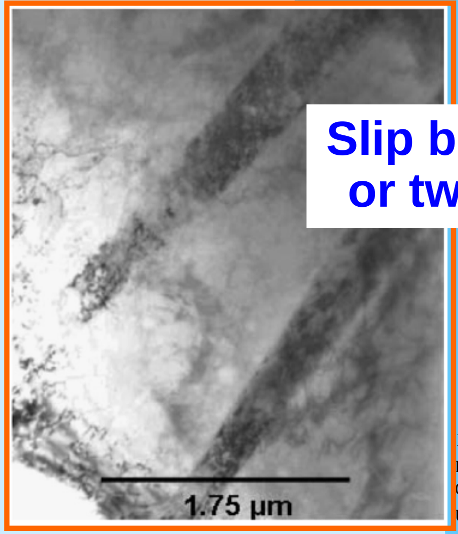
$$\dot{\varepsilon}_{SB} = \frac{50l}{\lambda^4}\frac{\sigma\Omega}{kT}D_B\delta$$

Spingarn and Nix (1979)

Gollapudi, Bhosle, Charit and Murty, Phil. Mag. (2008)

Deformation microstructure





Slip bands or twins?

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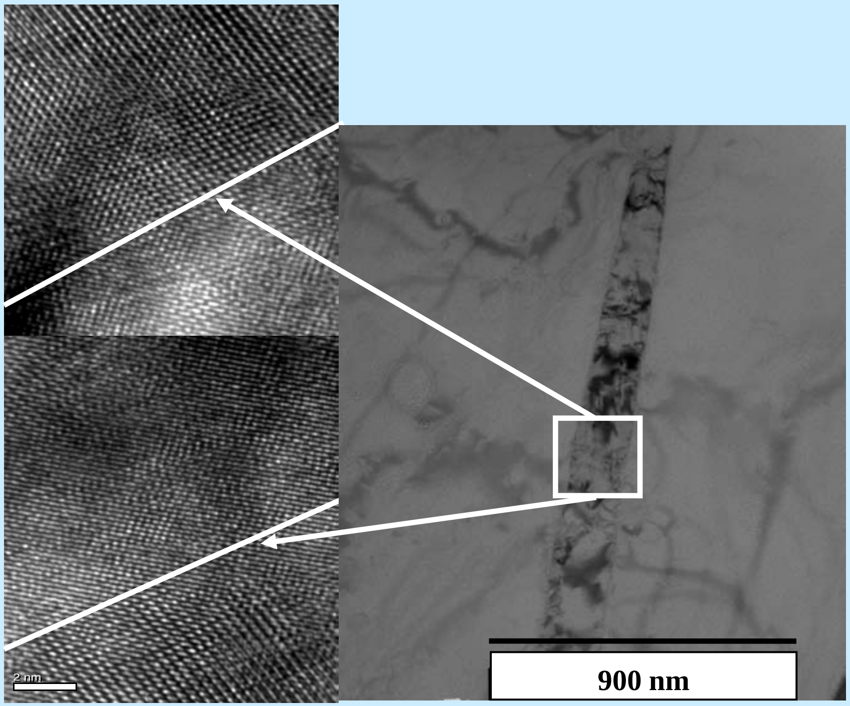
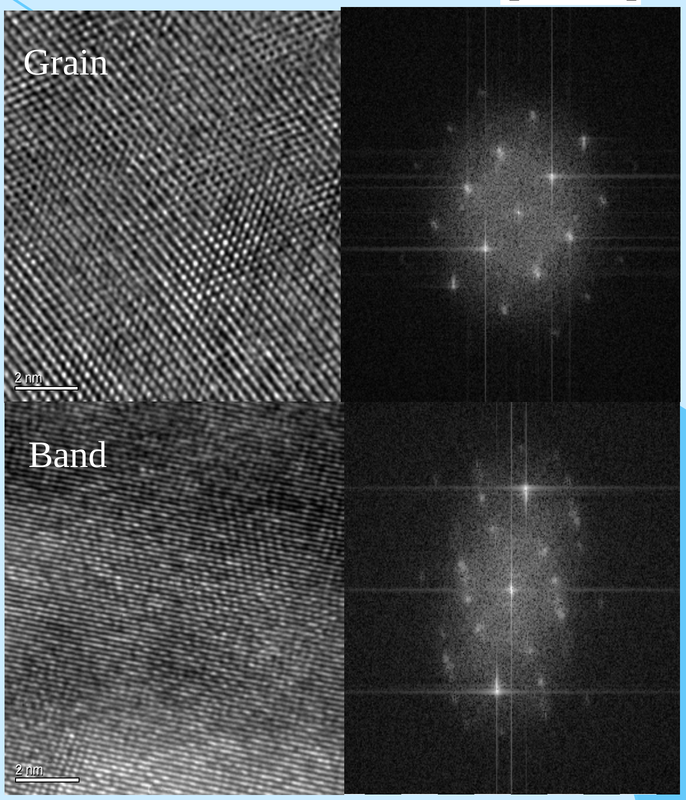
Slip Bands - not Twins

The grain orientation $[\bar{1}2\bar{1}3]$





The band orientation [0001]

HRTEM studies indicated the nature of these bands to be slip bands rather than twins

Phil Mag., 88:9 (2008) 1357-1367

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ZIRCALOY PROPERTIES

Objectives & Goals

Quantify Texture Using CODF

Develop Crystal Plasticity & Creep Models to Predict Anisotropic Creep, Deformation & Formability of Zircaloy Cladding, Sheet & TREX

Applications

Selection of Optimum Textures for tube, Grid & Channel Fabrication

Prediction of In-Service Dimensional Changes of Cladding [PWR & BWR] & Channels [BWR]

Reliability of spent fuel during dry storage and transportation