Helium Embrittlement: Progress and Future Research

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Presentation Outline

- Key Experimental Observations and Design Implications;
- Helium Transport;
- Critical Bubble Models;
- Creep Constrained Cavitation & Rupture;
- Alloy Design for Helium Management;
- Future Research Needs.

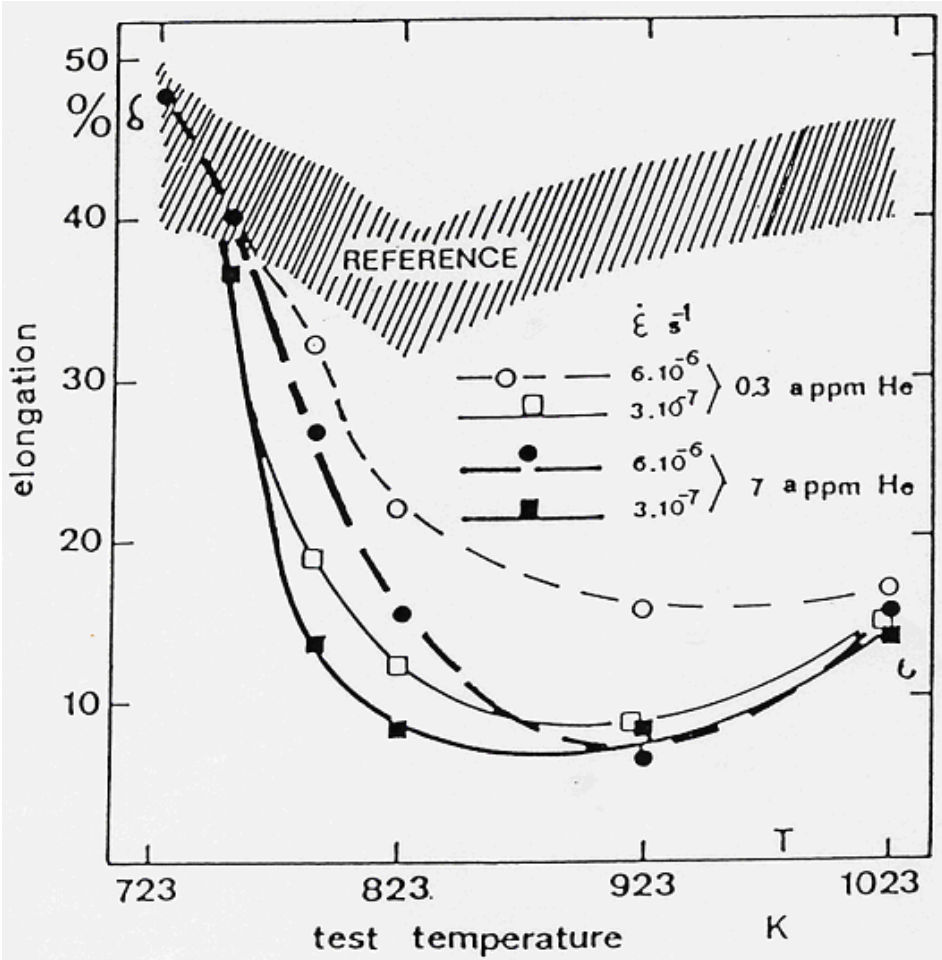
Test Temperature Effects

- **Creep Rupture Time:** Can be reduced by a factor of 10 for a few appm He → Nucleation Control.

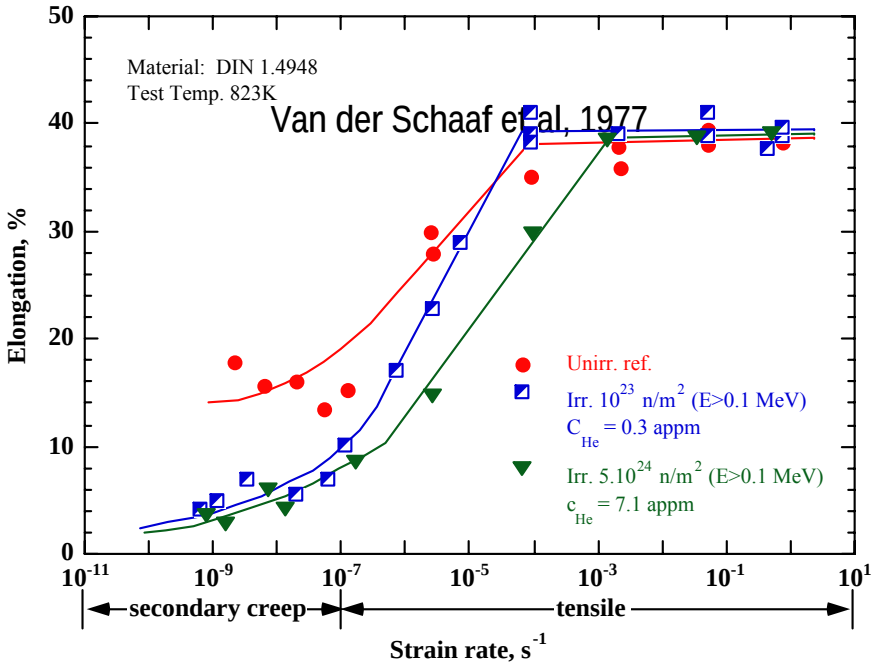
- **Test Temperature Effects:** Loss of ductility starts at ~500 ^{O}C. Partial recovery at T> 700 ^{O}C

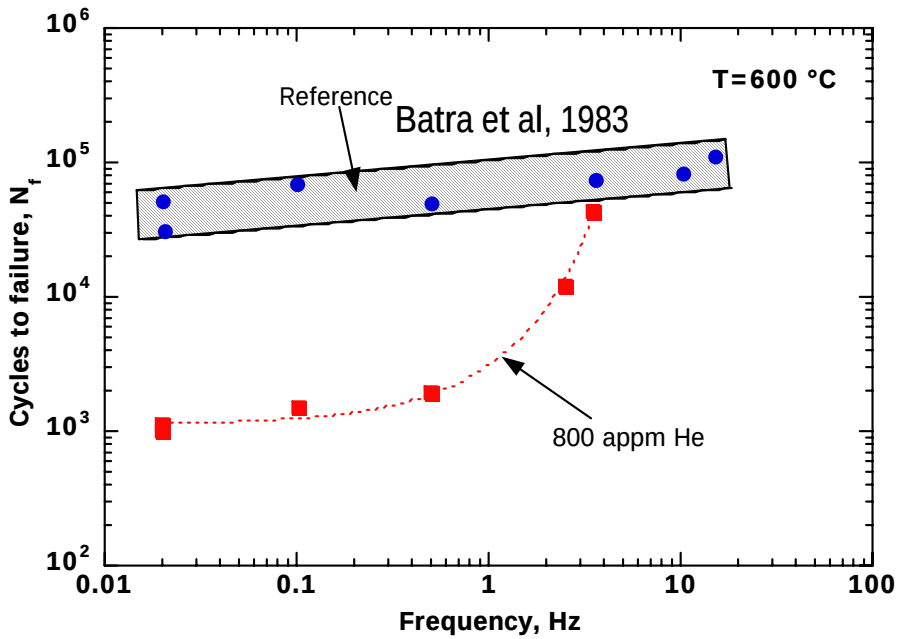
  → Bubble mobility and coalescence.

De Vries et al



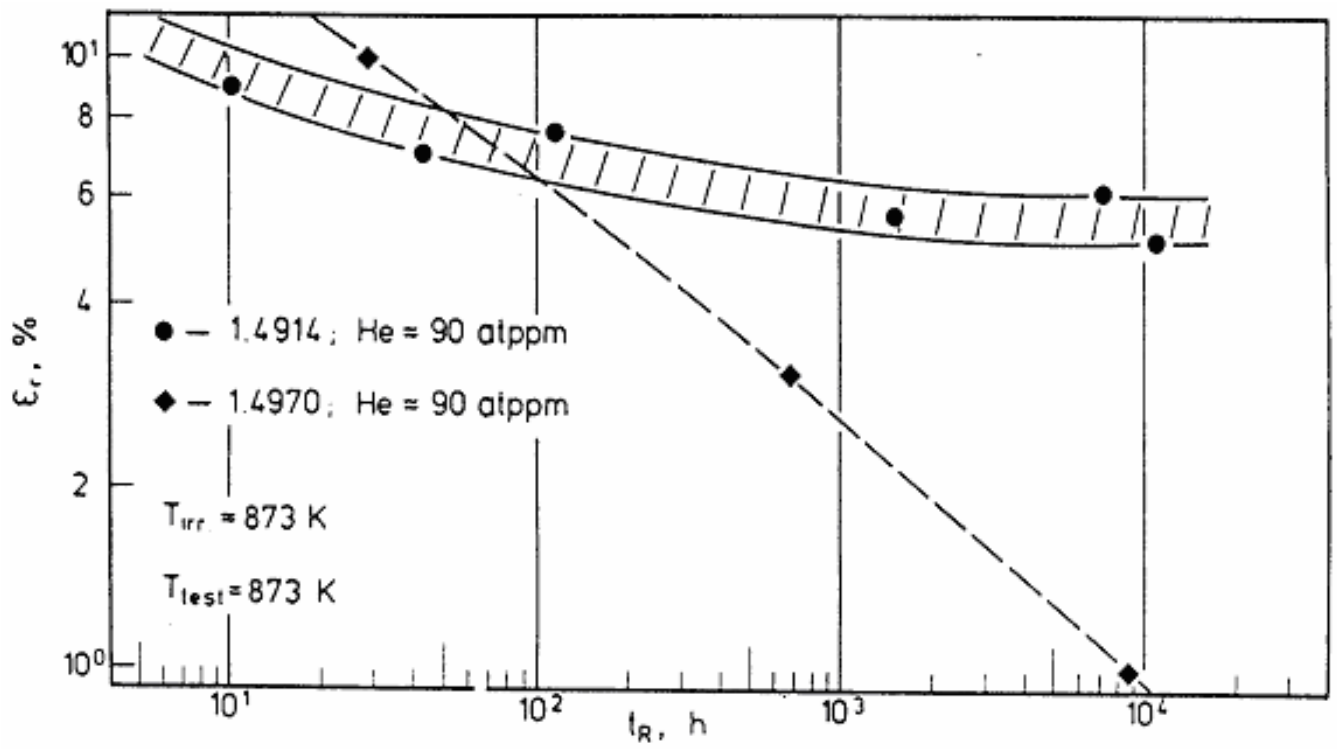
Strain Rate Effects





- Most drastic at low strain rates (10^{-7}-10^{-6} s^{-1})
- No effect at high strain rates (>1 s^{-1})
- ➡ Slow material transport
- (e.g. diffusion / plasticity).

Ferritics vs Austenitics

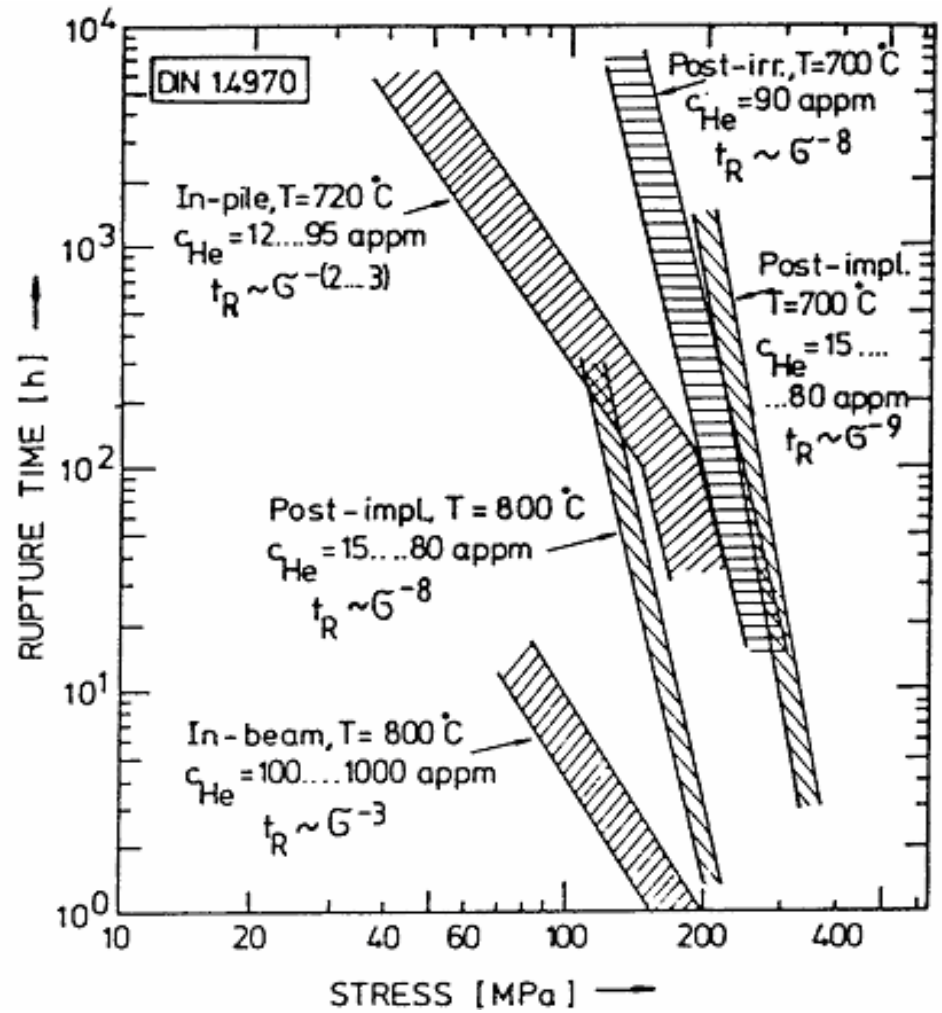


- Austenitics ductility decreases significantly with strain rate
- Ferritics are more resistant to helium embrittlement
- → Cavity growth is constrained in Ferritics.

In-pile vs pre-implanted

Shroeder's Data

- In-pile/in-beam shows a milder stress dependence of rupture time $t_R \approx \sigma^{-(2..3)}$
- Pre-implanted shows $t_R \approx \sigma^{-(8..9)}$
-
- Complex role of microstructure.
- Irradiation Creep (Odette)

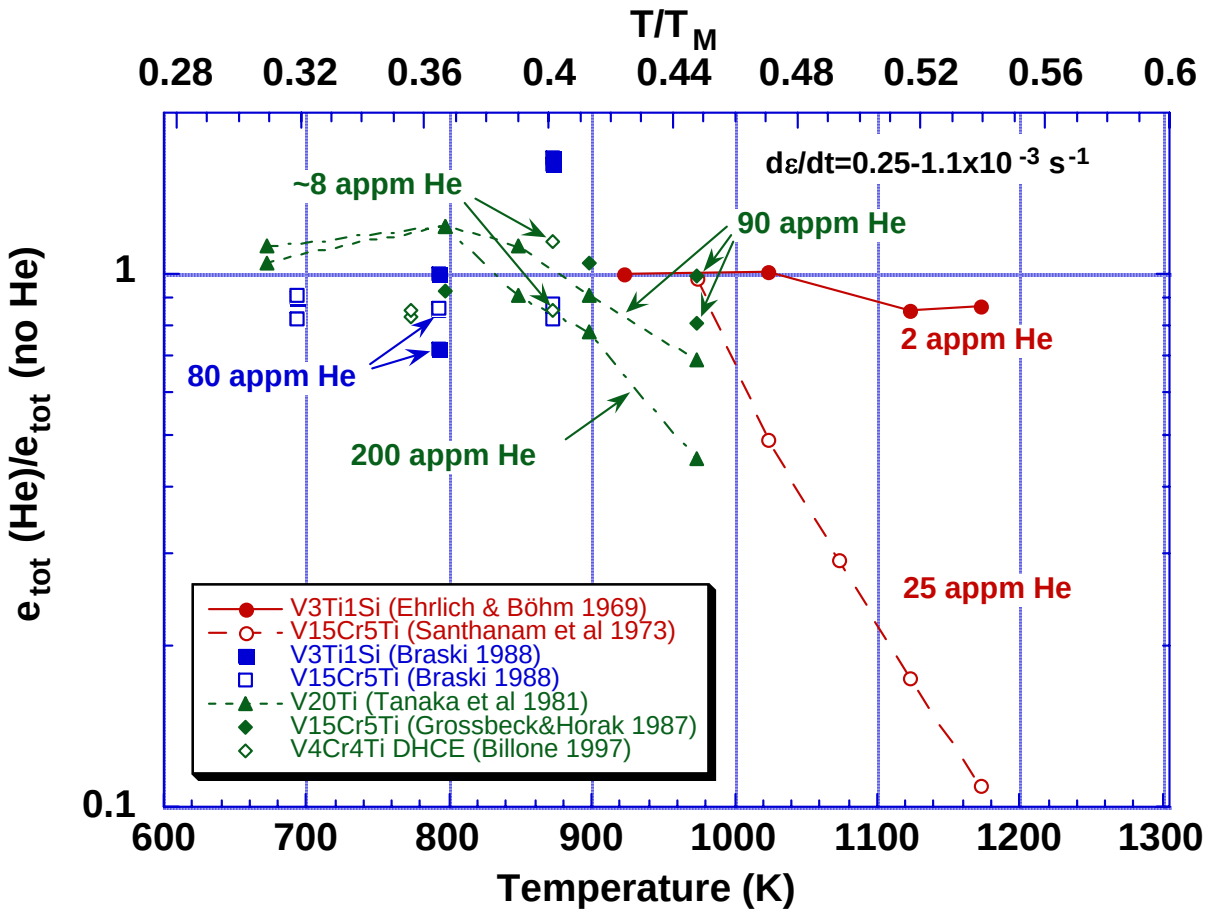


Effects of He on Vanadium

He embrittlement in V alloys becomes evident in tensile tests for:

T > 700°C

C_{he} > 100 appm



Helium Transport

1) Essentially insoluble in metals, extrinsic interstitial migration energy is very low (0.14 eV in Ni by Philips & Sonnenberg);
2) Transport to grain boundaries is highly-dependent on the microstructure;
3) Strong coupling between matrix bubble nucleation/growth and helium flow to grain boundaries;
4) Complex possibilities:
    1) Thermal trapping/ detrapping in vacancies and vacancy clusters;
    2) Replacement of substitutional helium by SIAs;
    3) Extrinsic interstitial diffusion;
    4) Dynamic displacement by cascades;
    5) Pipe diffusion along dislocation cores;
    6) Stress-induced transport by dislocation motion;
    7) Bubble thermal & stress gradient transport.

Dominant Mechanisms*

1) Radiation Displacement Regime:

$$D_{He}^{eff} = \frac{D_{01} g G}{K_{01}^{10} \nu_{01} \exp(-E_{He}^{M} / kT) C_{10} + gG}$$

1) SIA-Replacement Regime:

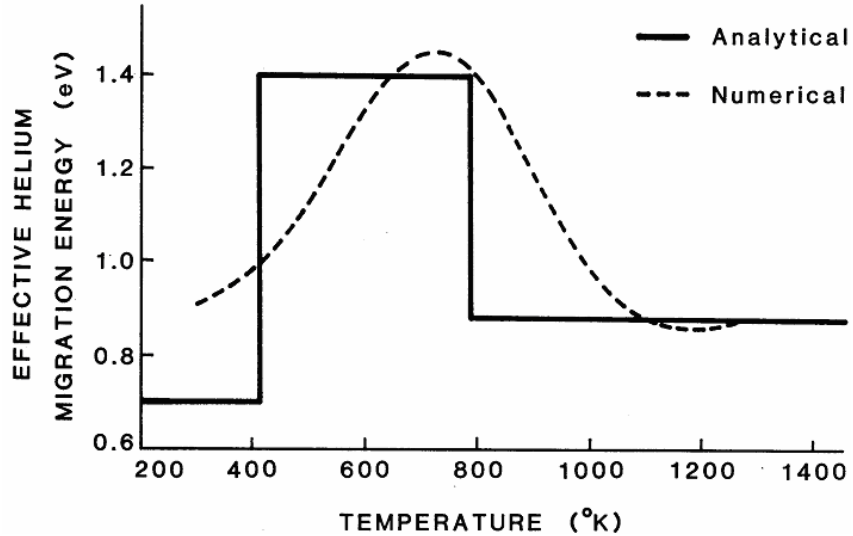
$$D_{He}^{eff} = \eta D_v, \quad \text{and } \eta = K_I^{11} / K_{01}^{10}$$

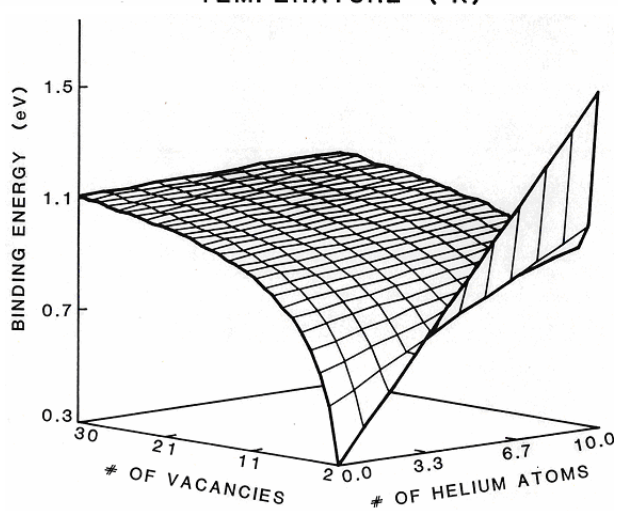
2) Thermal De-trapping Regime:

$$D_{He}^{eff} = \frac{\alpha \exp\left[-\left(E_{Hev}^{B} + E_{He}^{M} - E_v^{F}\right)\right]}{1 + \beta \exp\left[-\left(E_{Hev}^{B} - E_v^{F}\right)\right]}, \text{ and } \beta = \frac{K_{11}^{v}}{K_{01}^{10} \exp\left(-E_v^{F} / kT\right)}$$

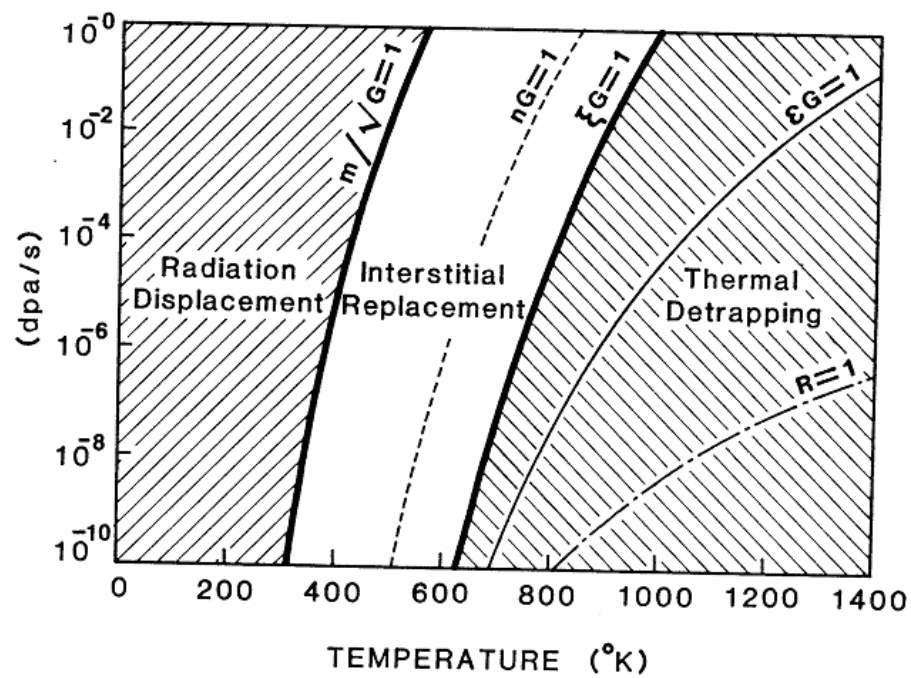
*N. M. Ghoniem, S. Sharafat, J. Williams, and L. K. Mansur, "The Theory of Helium Transport and Clustering in Materials Under Irradiation", *J. Nucl. Mater.*, **117**:96-105, 1983

Effective He Diffusion

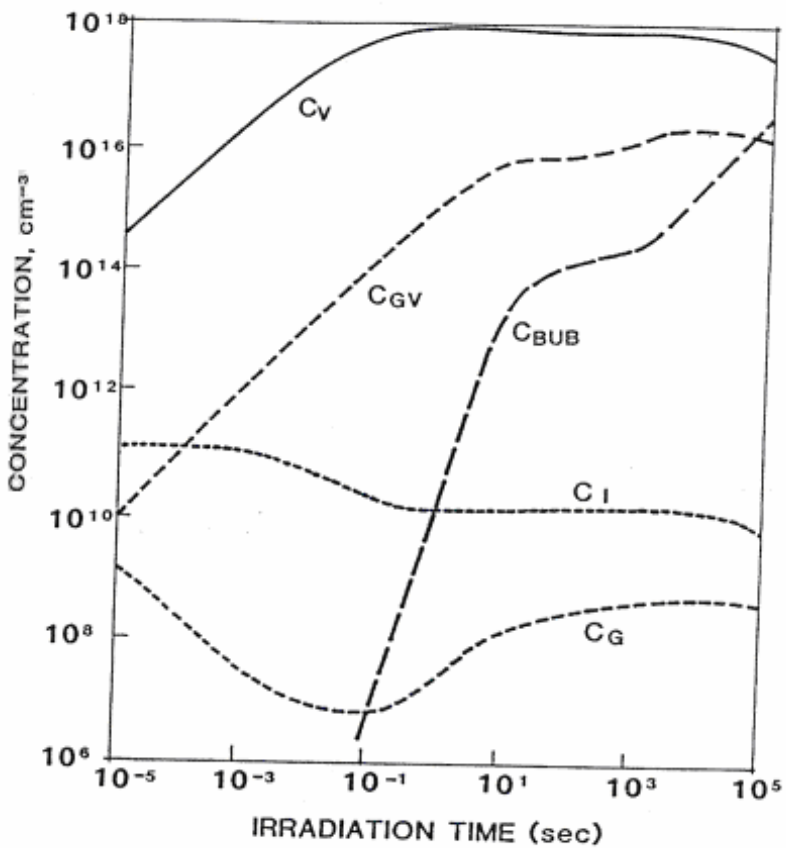


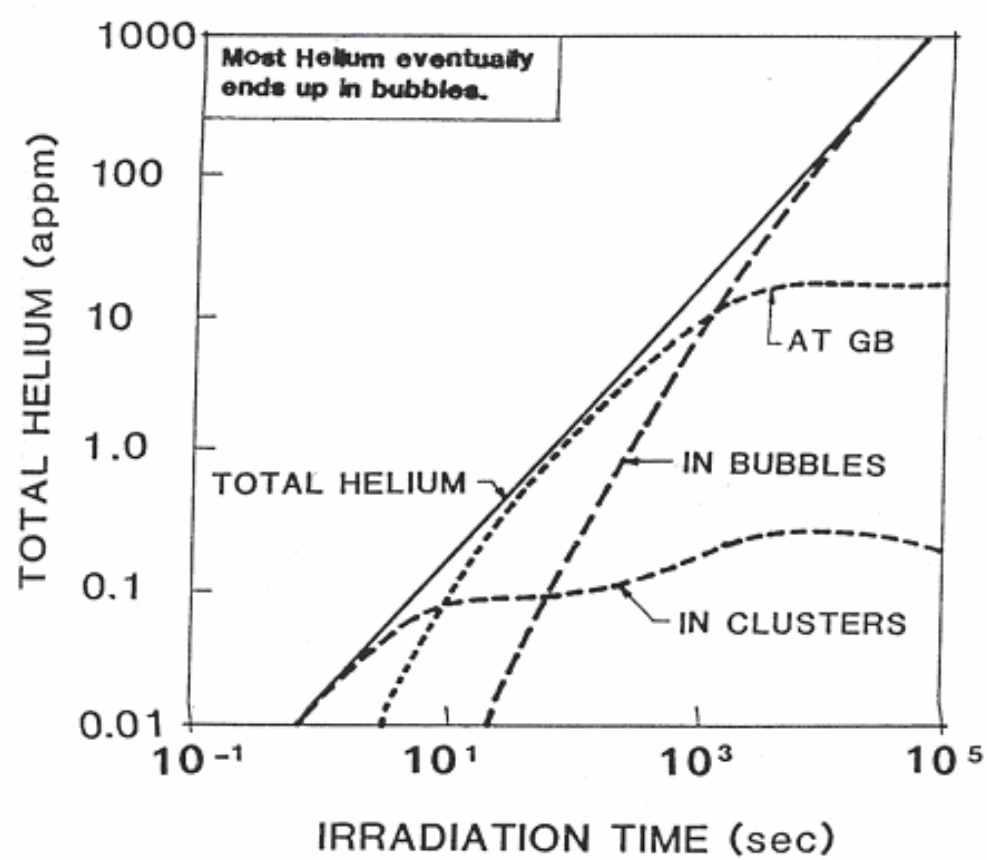


BINDING ENERGY AS A FUNCTION OF VACANCY AND He

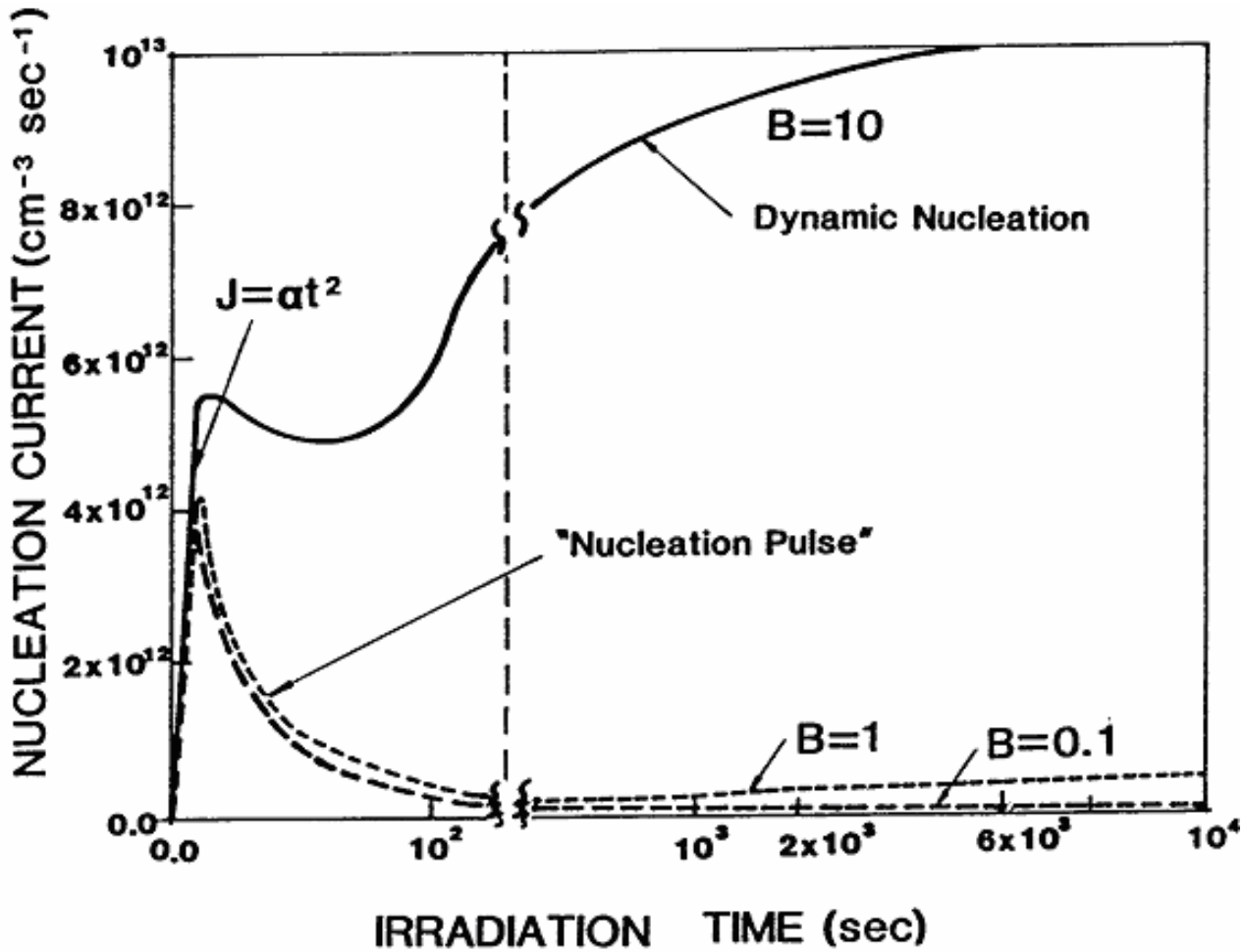


Helium Transport in the Radiation Field





Dynamic (secondary) Nucleation. Is it real?



Creep Rupture Models

Hull & Rimmer (1959)
Speight & Harris (1967)
Weertman (1973)
Raj & Ashby (1975)
Speight & Beere (1975)

→ $\sigma t_R = const.$

Hancock (1976)
Beere & Speight (1978)
Edward &Ashby (1979)

→ $\dot{\varepsilon} t_R = const.$

Ishida & Mclean (1967)
Ashby (1972)
Burton (1973)
Harris (1973)
Beere (1980)

→ **Source Control Of Vacancies**

Raj & Ashby (1975)

→ *Shape Factors
*Nucleation and growth

Evans, Rice, Hirth (1980)
Argon, Chen, Lau (1980)
Lau (1981)

→ Stress Evolution and Relaxation at TPJ and Particles

Hyam & Sumner (1962)
Lewthwaite (1967)
Brailsford & Bullough (1973)

→ Growth Instability Criterion

Beere (1981)

→ Stress + Pressure Driven Growth

Trikanus & Ulmaier(1979)

→ Stress + Pressure Driven Nucleatin &Growth

Beere (1984)

→ Small Bubble Sweeping At Grain Boundaries

Critical Bubble Concept

- **Hull & Rimmer**

$$\text{Void:}\quad R_{crit} > \frac{2\gamma}{\sigma}, \quad \text{and} \quad \frac{dR}{dt} = K\frac{D_{gb}}{kT}\left(\sigma - \frac{2\gamma}{R}\right)$$

- **Hyam & Sumner**

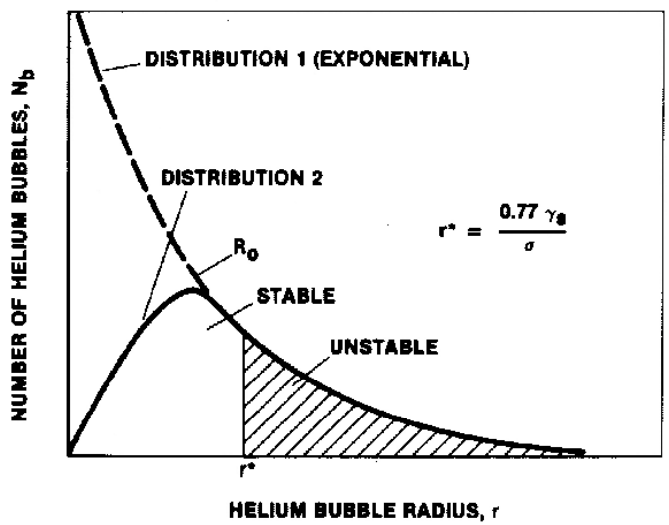
$$\text{Bubble:}\quad R_{crit} > \frac{0.77\gamma}{\sigma}, \quad \text{and} \quad \frac{dR}{dt} = K\frac{D_{gb}}{kT}\left(\sigma + p - \frac{2\gamma}{R}\right)$$

Helium Embrittlement Models -1

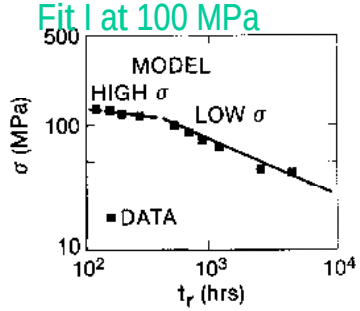
- In-situ creep-rupture: Nabarro-Herring flow of vacancies from PFZ plus irradiation creep (B) leads to simple scaling laws:

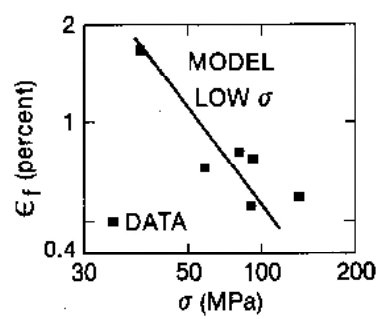
  @ low σ: $t_r \approx I/[(He/dpaB)^{1/2}\sigma^2]$ and $\varepsilon_r \approx I\sqrt{B}/[(He/dpaB)^{1/2}\sigma^2]$

Odette 1984

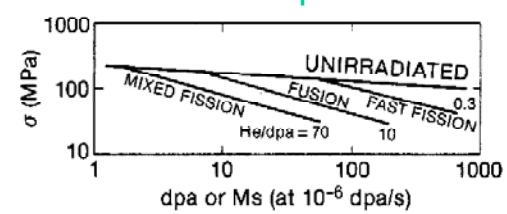


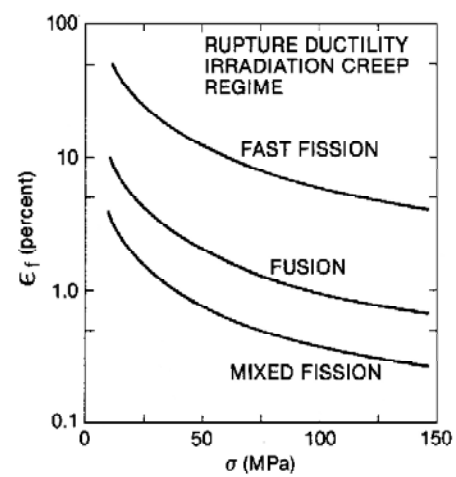
Fit I at 100 MPa





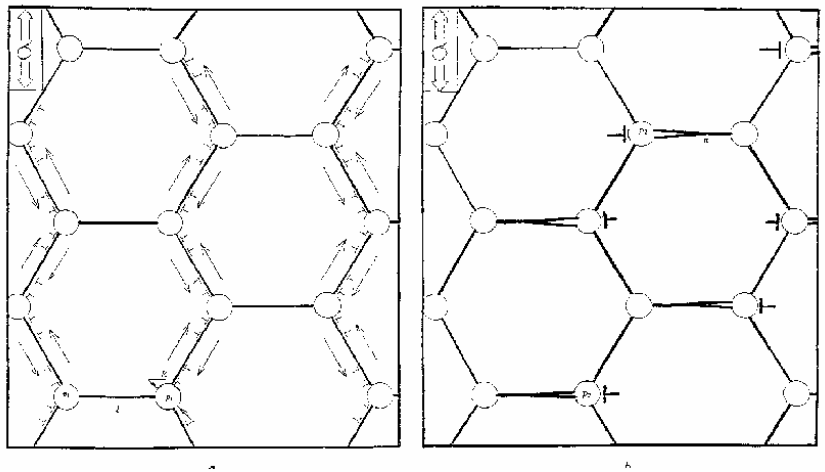
Predicted Implications





Helium Embrittlement Models -2

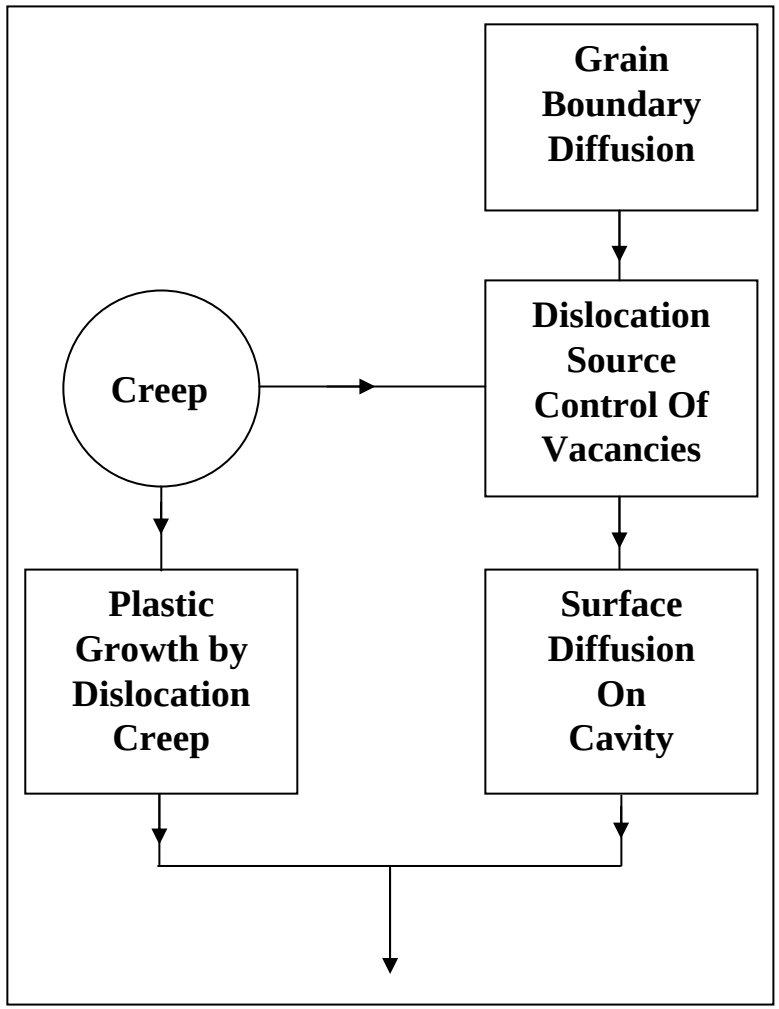
- 2-D model of cylinders at TPJs;
- GB sliding is represented by dislocation pileups;
- Modified Griffith criterion for gas-filled TPJ cracks.
- Instability is obtained by minimization of energy w/t crack length and gas pressure.
- Does not explain slow creep rupture.

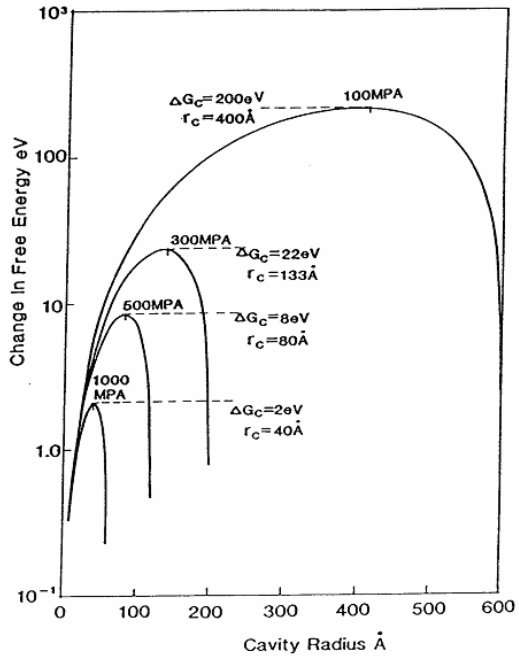


Ryazanov-Trinkaus, 96

Helium Embrittlement Models -3

Alhajji-Ghoniem, 85

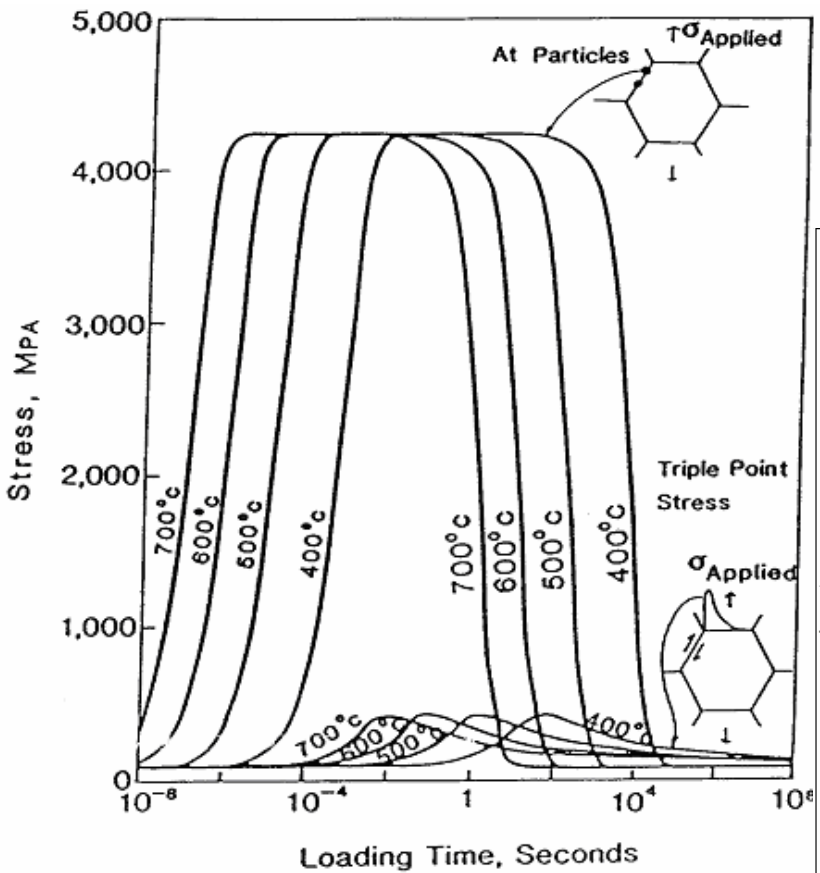


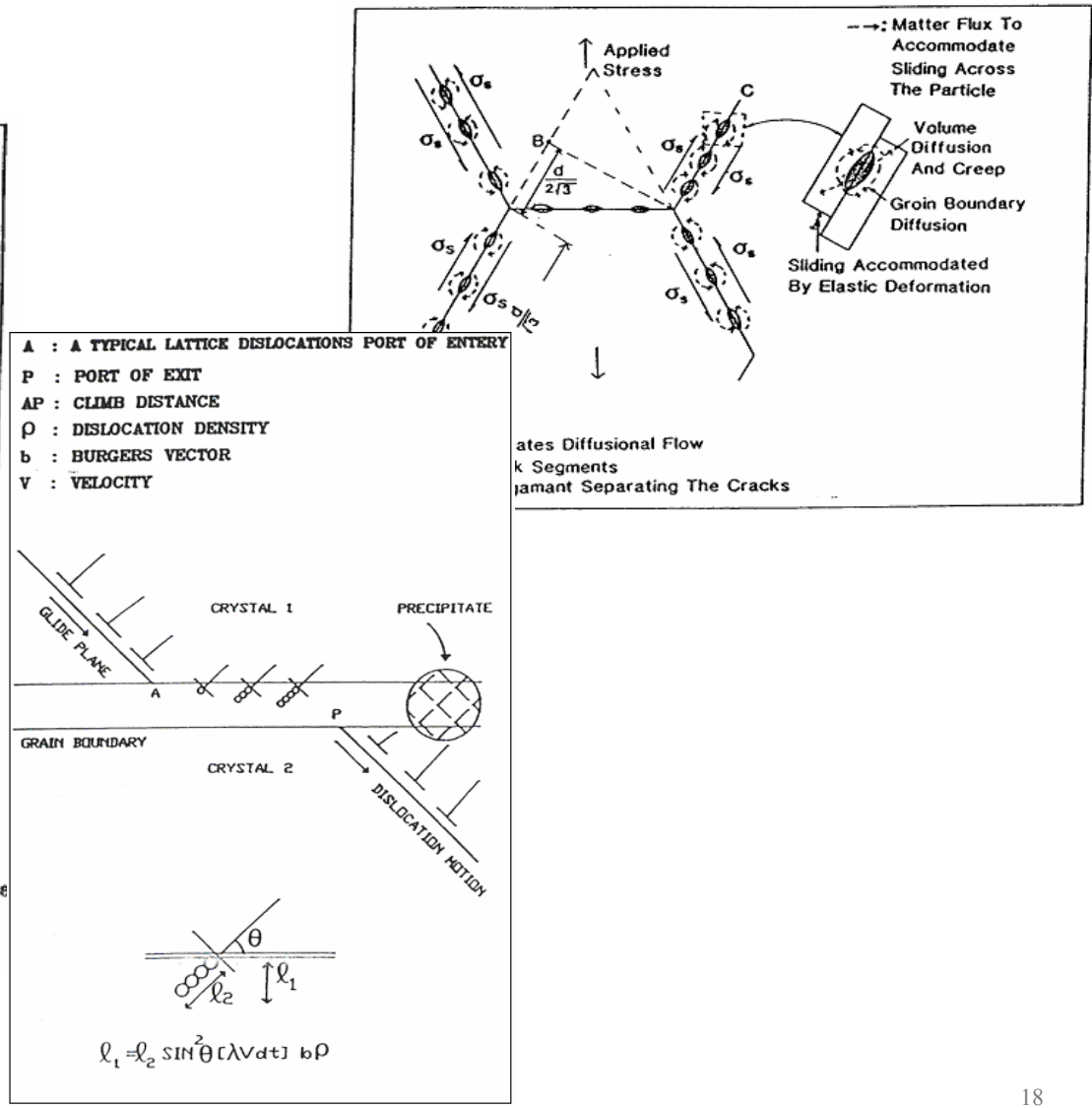


$$\dot{V} = \dot{V}_{DC} + \frac{\dot{V}_{BD}\dot{V}_{SC}}{\dot{V}_{BD} + \dot{V}_{SC}}$$

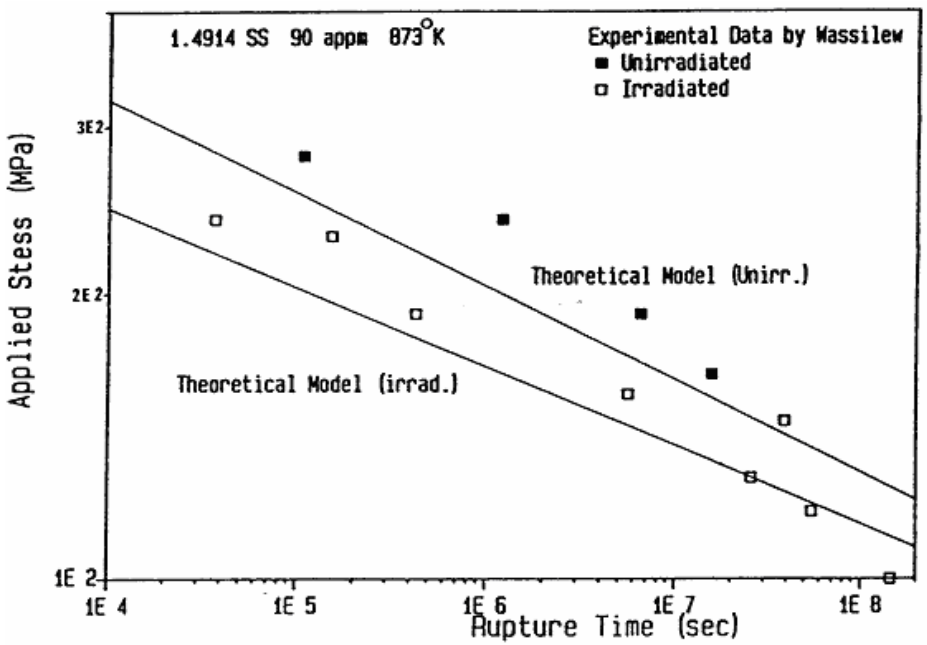
Helium Embrittlement Models -3

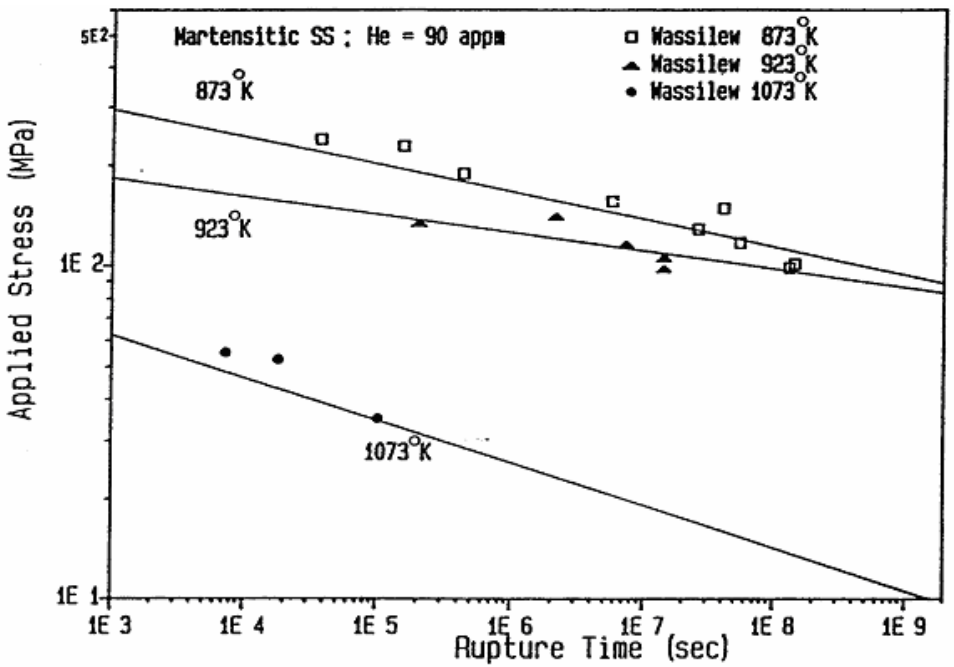
Alhajji-Ghoniem, 85





Helium Embrittlement Models -3





Austenitic

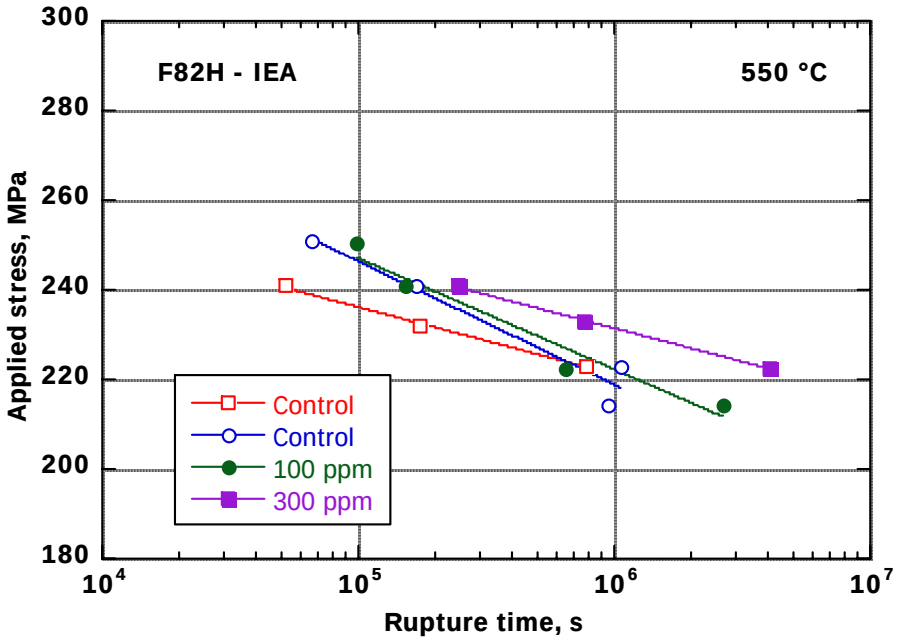
Martensitic

Alloy Design for Helium Management

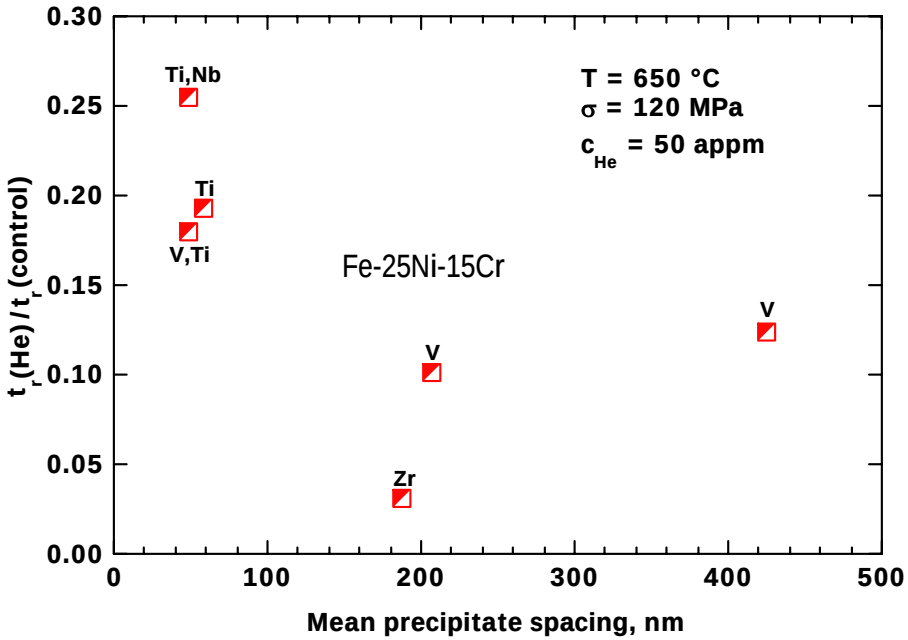
1. Limit grain boundary dislocation climb by increasing the precipitate density at grain boundaries.
2. Reduce thermal creep rates. This limits bubble growth by creep deformation and reduces the vacancy supply by dislocations entering the boundary.
3. Increase the bubble surface energy (segregation?) to delay bubble growth instability.
4. Increase the matrix precipitate density to reduce helium flux to grain boundary.
5. Increase the matrix bubble density to reduce the sink strength of grain boundaries.

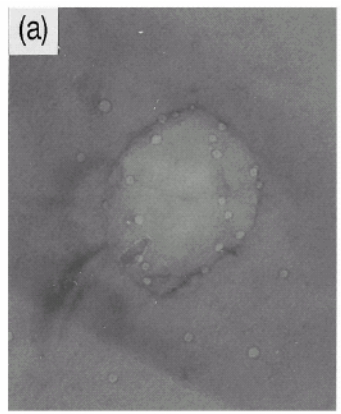
Microstructure Design Improves Resistance to He Embrittlement

Yamamoto et al, 2000



Yamamoto et al, 1998





Titanium additions to austenitic alloys improves creep rupture by capture of MC-type precipitates of (implanted?) helium.

Future Research Needs
1- Atomistic

1. KMC & MD of helium transport. Rate theory conclusions firm?
2. Ab-initio/ MD of He-V cluster binding energies;
3. Ab-initio/ MD of He-precipitate (MC-type) binding energies;
4. Cascade-bubble interaction (helium re-solution & re-nucleation);
5. KMC of bubble migration & coalescence (matrix, GBs, dislocations?)
6. Relationship between GB structure and He bubble density/ size;
7. KMC of GB bubble/ void transition & instability;
8. Dislocation-GB interaction in BCC vs FCC & vacancy source.

Future Research Needs
2- Micromechanics

1. 3-D Micro-mechanics/ FEM (e.g. ABACUS) Models of Grain Boundary Sliding and Stress Evolution;
2. DD simulations of Dislocation-GB interaction & Cross-slip for FCC and BCC;
3. Coupled DD/FEM (or mesh-free) for matrix creep/ GB evolution and sliding of polycrystals.
4. Eshelby-Based KMC for He flow to GBs;

$$u_i^m(\mathbf{R}_m) = \sum G_{ij}^{mn}(\mathbf{R}_m, \mathbf{R}_n)\, f_j^s$$

Future Research Needs 3 - Experimental

- Well-characterized, model-driven, single-variable experiments to resolve remaining He effects questions;
  - Bi-crystals: Helium implanted and loaded?
  - Helium desorption of (1) samples with precipitates; (2) samples with different grain size &/or structure.
- Proposed US/Japan collaborative experiments with possible He effects studies
  - DOE/JAERI (F82H and model ferritic alloys; He effects study would concentrate on 300 & 400˚C tensile, fracture behavior)
  - DOE/MEXT Jupiter-II (V alloys; experimental matrix for 1st irradiation to be decided this fall)

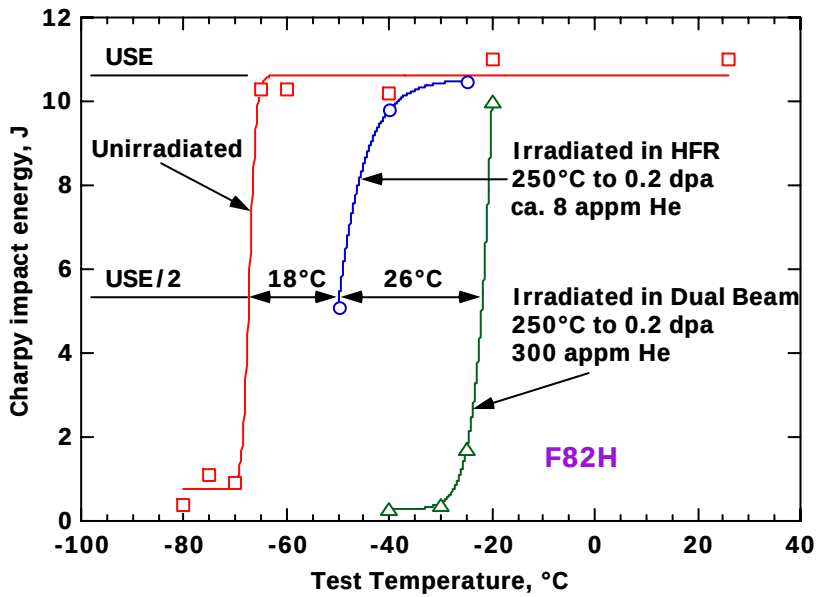
Additional Slides

Modeling Results and Observations

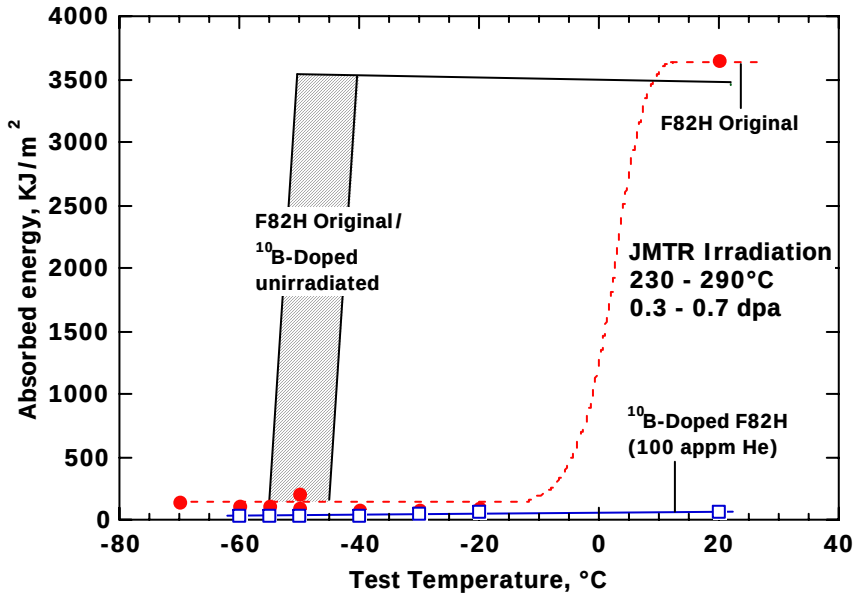
- Precipitates at GB's are necessary for cavitation in the absence of irradiation.
- t_R is controlled cavity growth (nucleation is fast).
- Irradiation results in a significant enhancement of GB cavity density.
- Higher helium implantation rates result in a higher grain boundary bubble density.
- The grain boundary bubble density reaches a steady state at large amounts if injected helium (>10 appm).
- Out of pile creep tests do not accurately describe the in-pile creep behavior.
- The resistance of martensitics to helium embrittlment is a result of vacancy source control.

Low-temperature Helium Embrittlement

Lindau, et al., 1999



Shiba & Hishinuma, 2000



Effects are masked by data scatter (related to initial microstructure) and a number of experimental uncertainties.

Design Implications

- Time-dependent Allowable Stress (S_t): Lowest value of
- (a) 2/3 of minimum stress to cause creep rupture in time t
- (b) 80% of minimum stress to cause tertiary creep in time t
- (c) Minimum stress to produce 1% strain in time t

- Extension of Standard ASME rules may be problematic

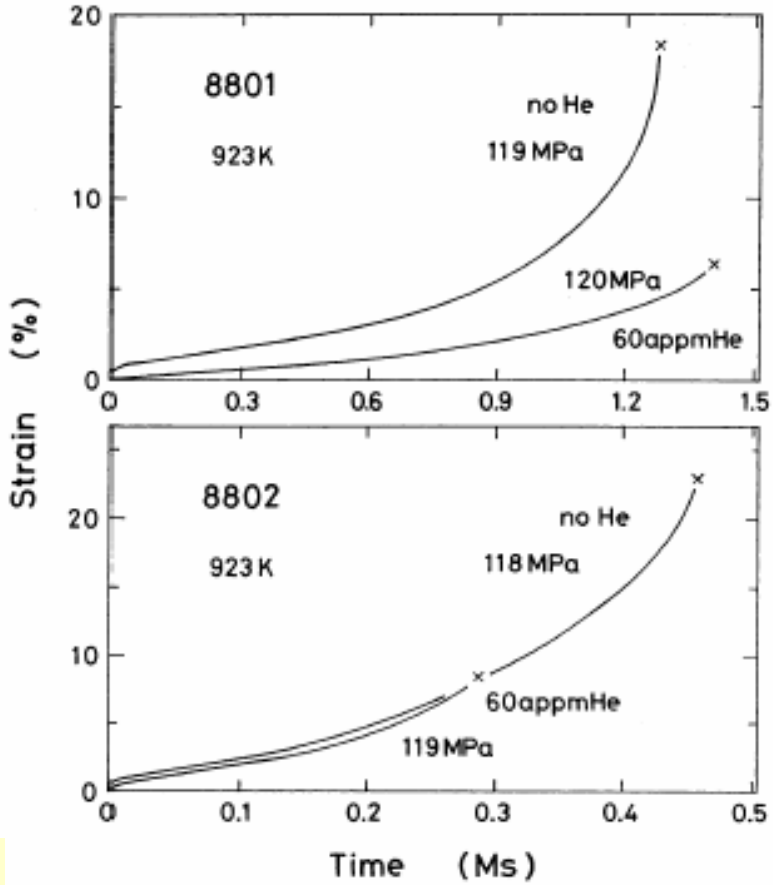


Fig. 3. Comparison of creep curves of thermomechanically treated Alloys 8801 and 8802 in helium free and implanted (C_{He} = 60 appm) conditions tested at 923 K, 120 MPa.