Impact of H and He Transmutation Products on Radiation Effects in Materials

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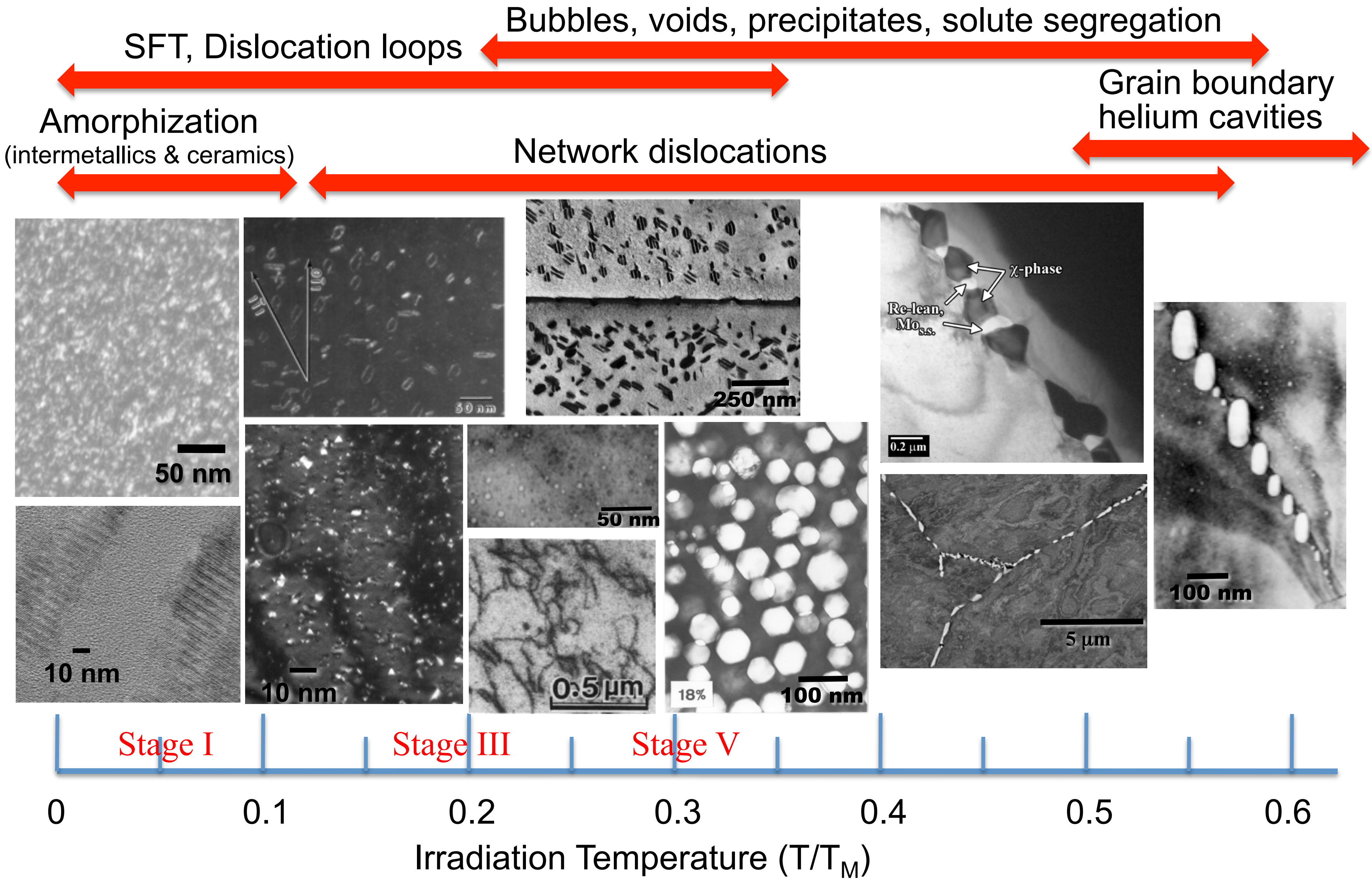
DEPARTMENT OF NUCLEAR ENGINEERING

OAK RIDGE National Laboratory

Outline

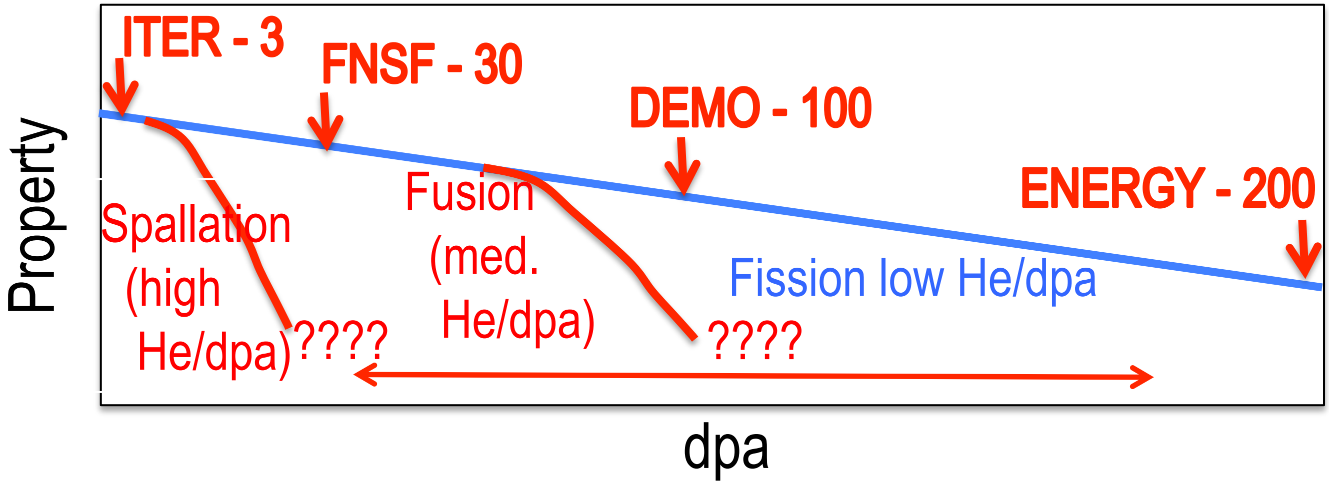
- **Low temperature phenomena: Hardening and embrittlement**
  - **Major effects observed in ferritic steels for C_{He}>500 appm**
- **Medium temperature phenomena: Cavity swelling**
  - **Major effects observed in austenitic steels for C_{He}>100 appm; ferritic steels for C_{He}>500 appm?**
- **High temperature phenomena: High temperature He embrittlement of grain boundaries**
  - **Major effects observed in austenitic steels for C_{He}>1-100 appm; ferritic steels C_{He}>500 appm?**
- **Influence of H is less pronounced (per atom) than He**
  - **H microstructural influence is mainly via chemical/chemisorption effects, vs. insoluble cavity precipitation (vacancy trapping) for He**
  - **H trapping in cavities at intermediate temperatures can be an important safety issue for DT fusion energy systems**

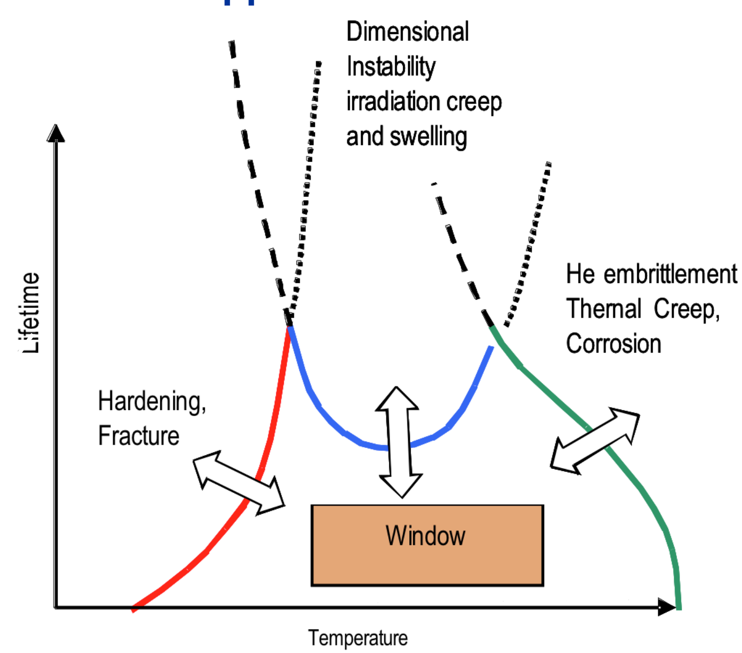
Temperature-dependent irradiated microstructures



Zinkle & Snead, Ann. Rev. Mater. Res. 44 (2014) 241

Challenge of understanding and mitigating property degradation during irradiation in He-rich environments

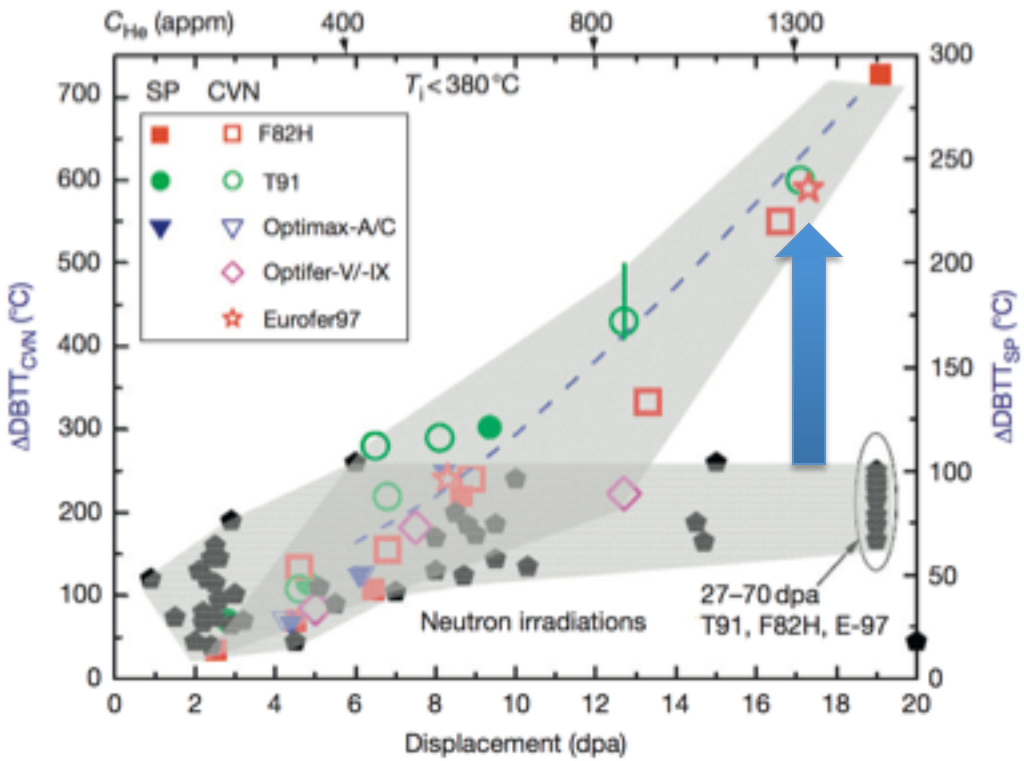




- Potential Mitigation – nanostructured ferritic alloys (NFA) – also known as ODS steels

Evidence for enhanced low temperature embrittlement due to high He production has been observed in simulation studies

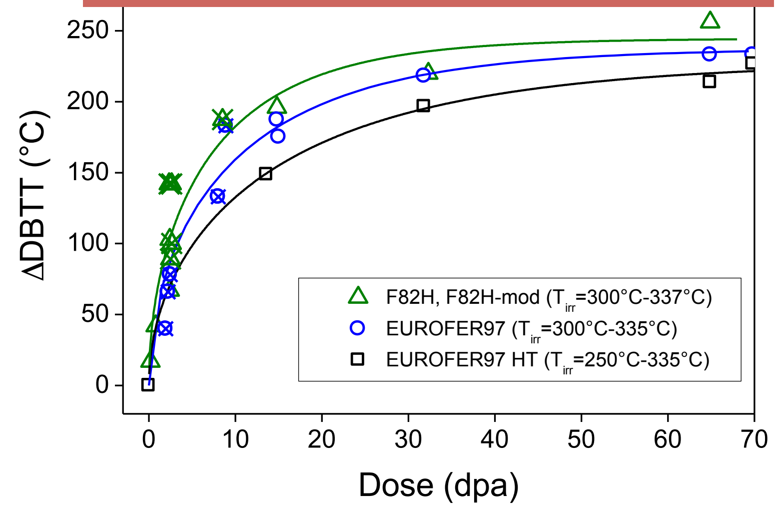
DBTT shift in ferritic/martensitic steel after fission and spallation (high He/dpa) irradiation



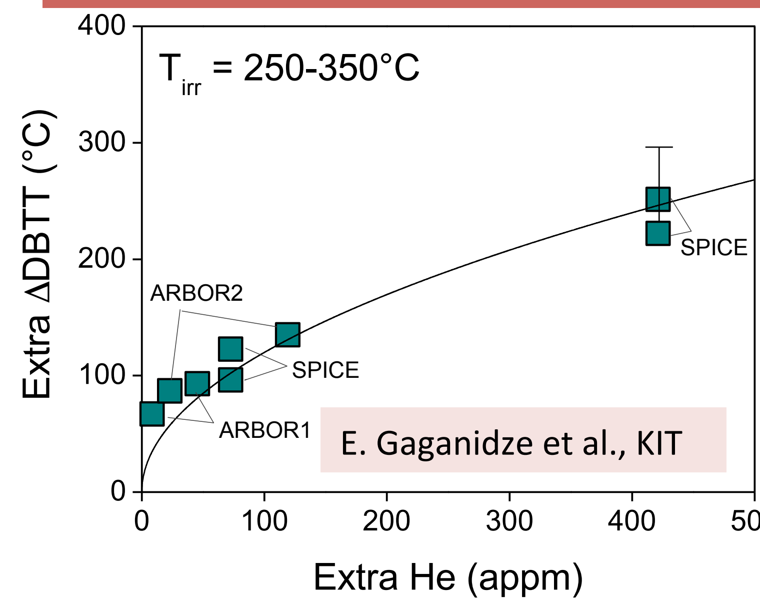
Y. Dai, G.R. Odette, T. Yamamoto, Comprehensive Nuclear Materials, vol. 1, R.J.M. Konings, Ed (2013) p. 141

Open question: Are B-doping and He-injector (Ni foil) simulation tests prototypic for actual fusion reactor condition?

EUROFER, <10 appm He



EUROFER, 10-500 appm He

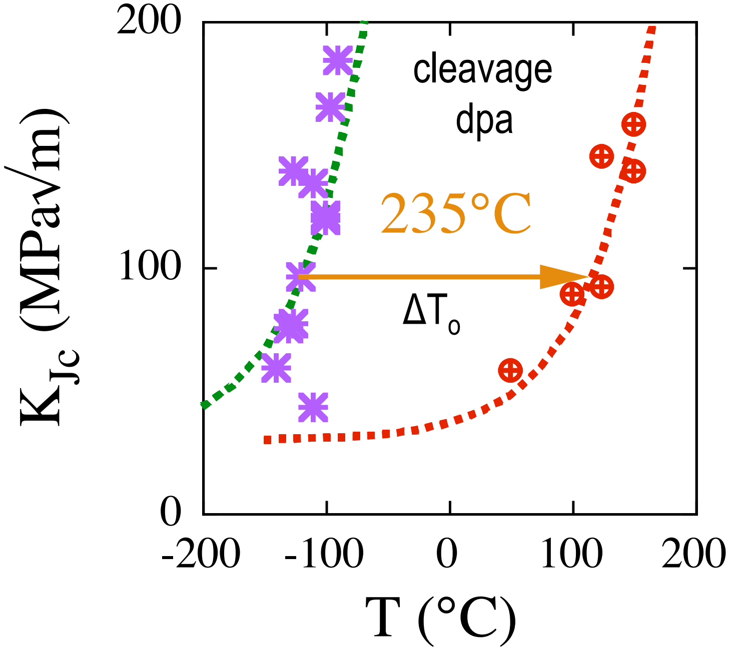


Evolution in understanding of fracture behavior in ferritic/martensitic steels containing high He

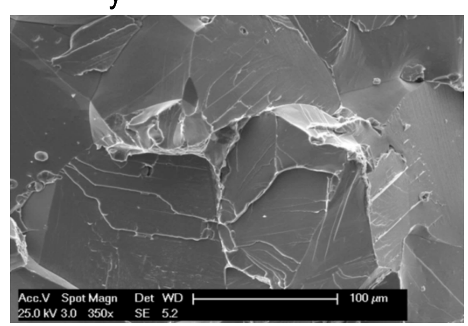
- Bulk He effects on fracture obtained mostly from STIP irradiations (PSI) up to ≈ 50 appm/dpa and ≈ 20 dpa (≈ 2000 appm He)
  - Tempered martensitic steels exhibit enormous ductile-brittle shifts at high He
  - Transition to intergranular fracture (IGF) occurs at high He
  - Elastic tensile fracture at very high He
  - Synergistic hardening ($\Delta\sigma_y$) + lower grain boundary cohesive strength (He)
  - Onset of severe temperature shifts at ≈ 500 appm He
  - Nanocomposited Ferritic Alloys (NFAs) do not experience IGF fracture or large ΔT shifts

NRG, UCSB

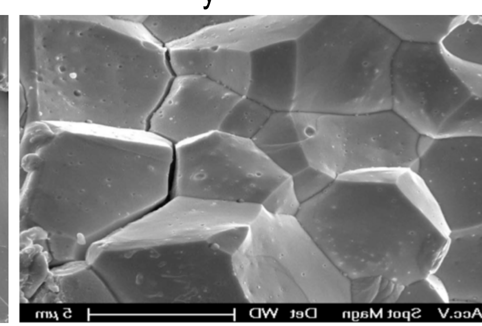
F82H 5dpa @ 300°C



$\Delta\sigma_y$ cleavage ΔT



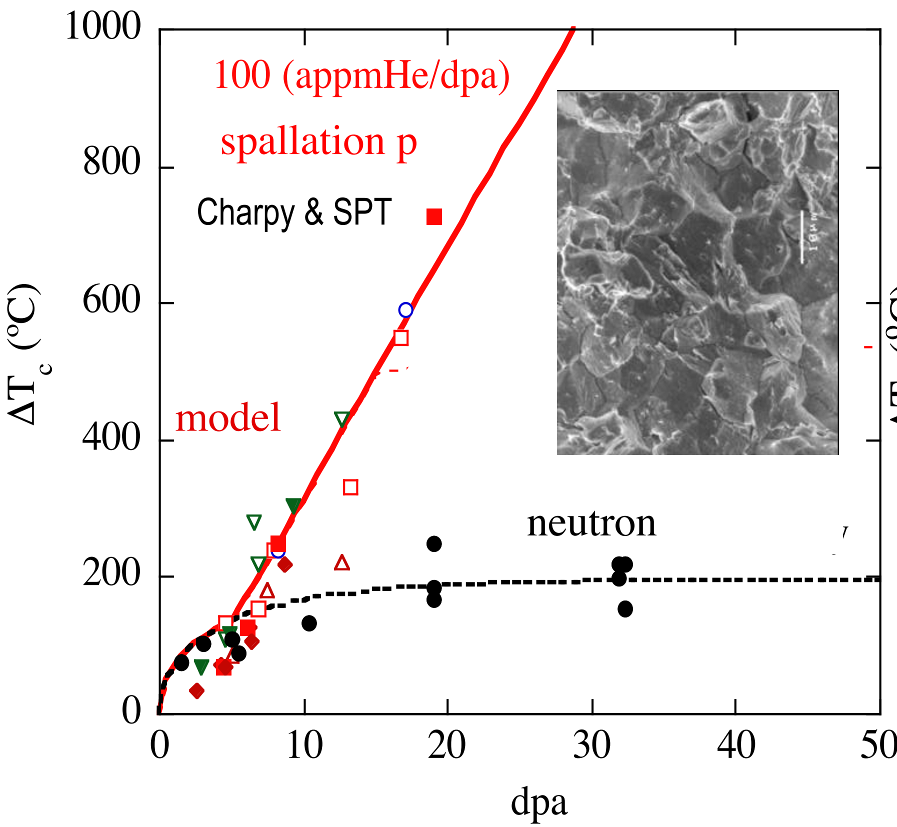
<< $\Delta\sigma_y$+ He IG ΔT

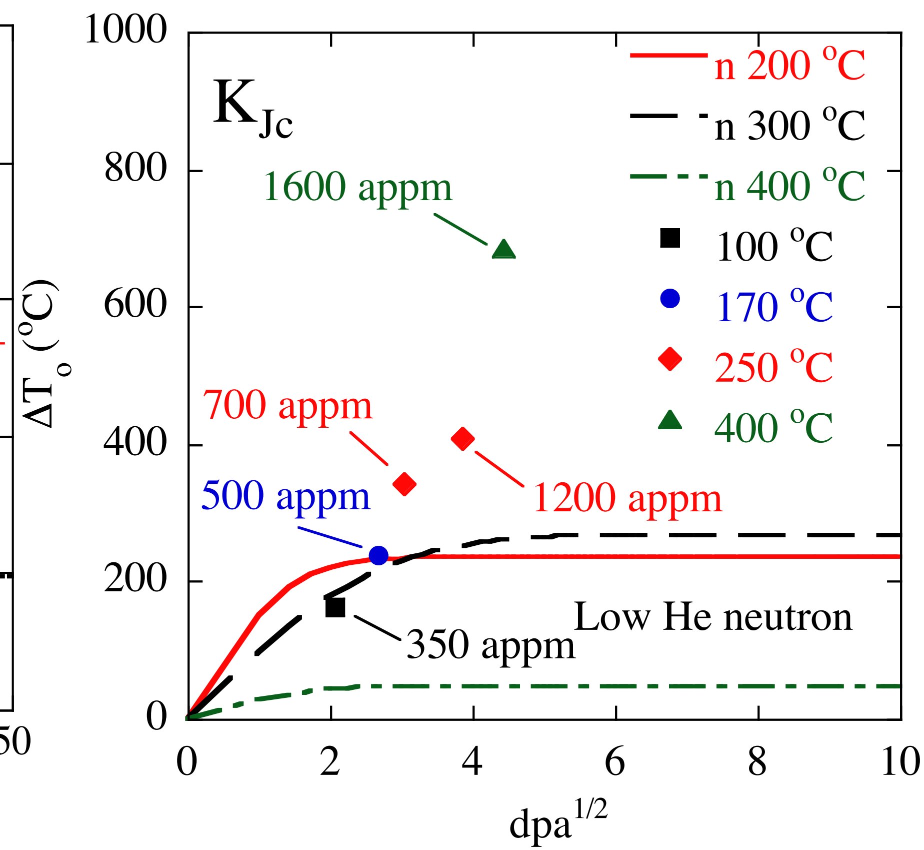


PSI, UCSB

Helium Effects on Fast Fracture - I

Severe He - $\Delta\sigma_y$ - ΔT synergisms and intergranular fracture starting at > ≈ 500 appm ***partly*** due to He weakening grain boundaries

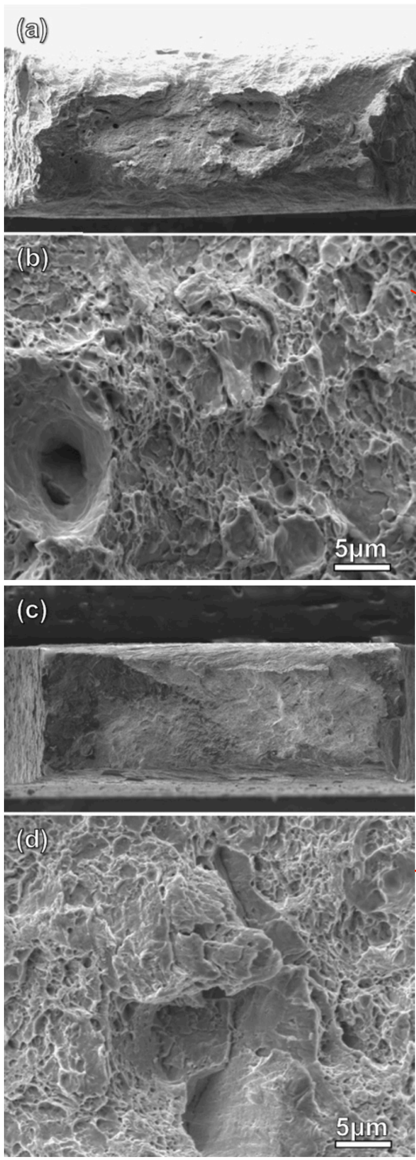




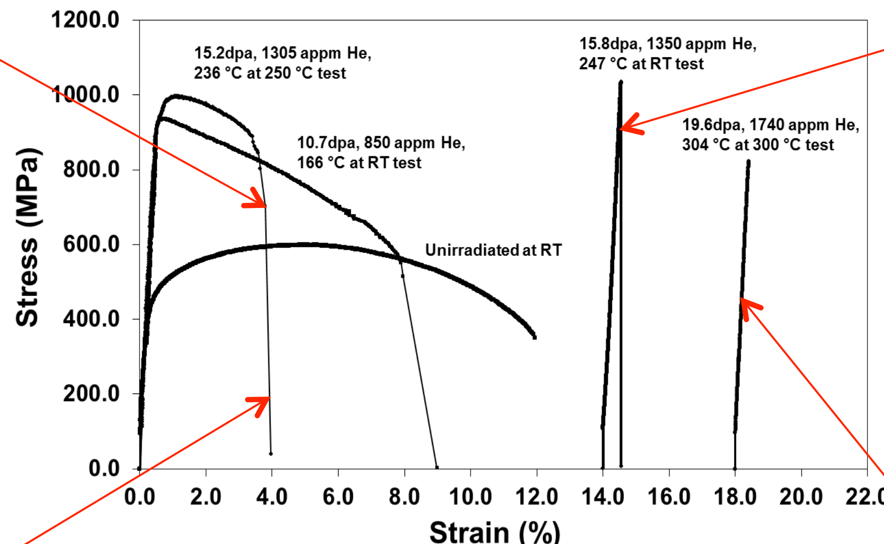
PSI, UCSB

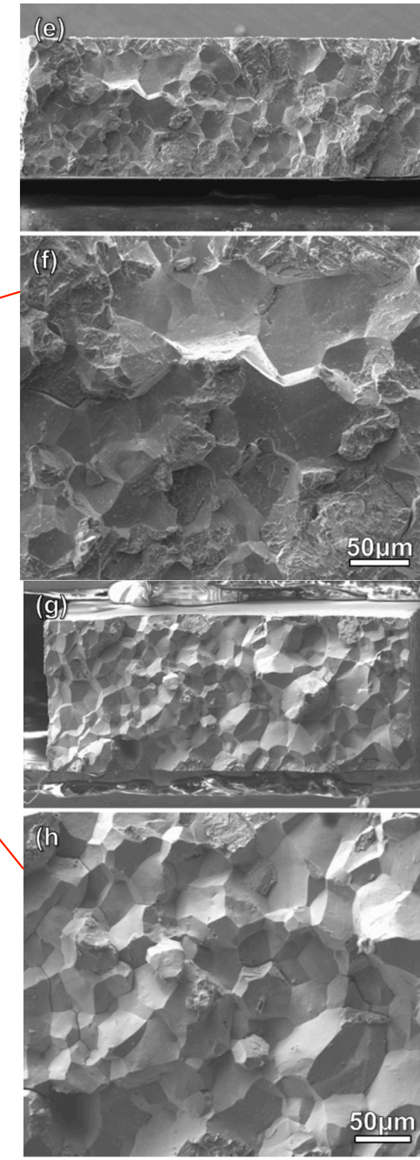
Y. Dai, G.R. Odette, T. Yamamoto, Comprehensive Nuclear Materials, vol. 1, R.J.M. Konings, Ed (2013)

Helium Effects on Fast Fracture - II



Elastic fracture in tensile tests (>1300 appm He) without cracks or stress concentrations



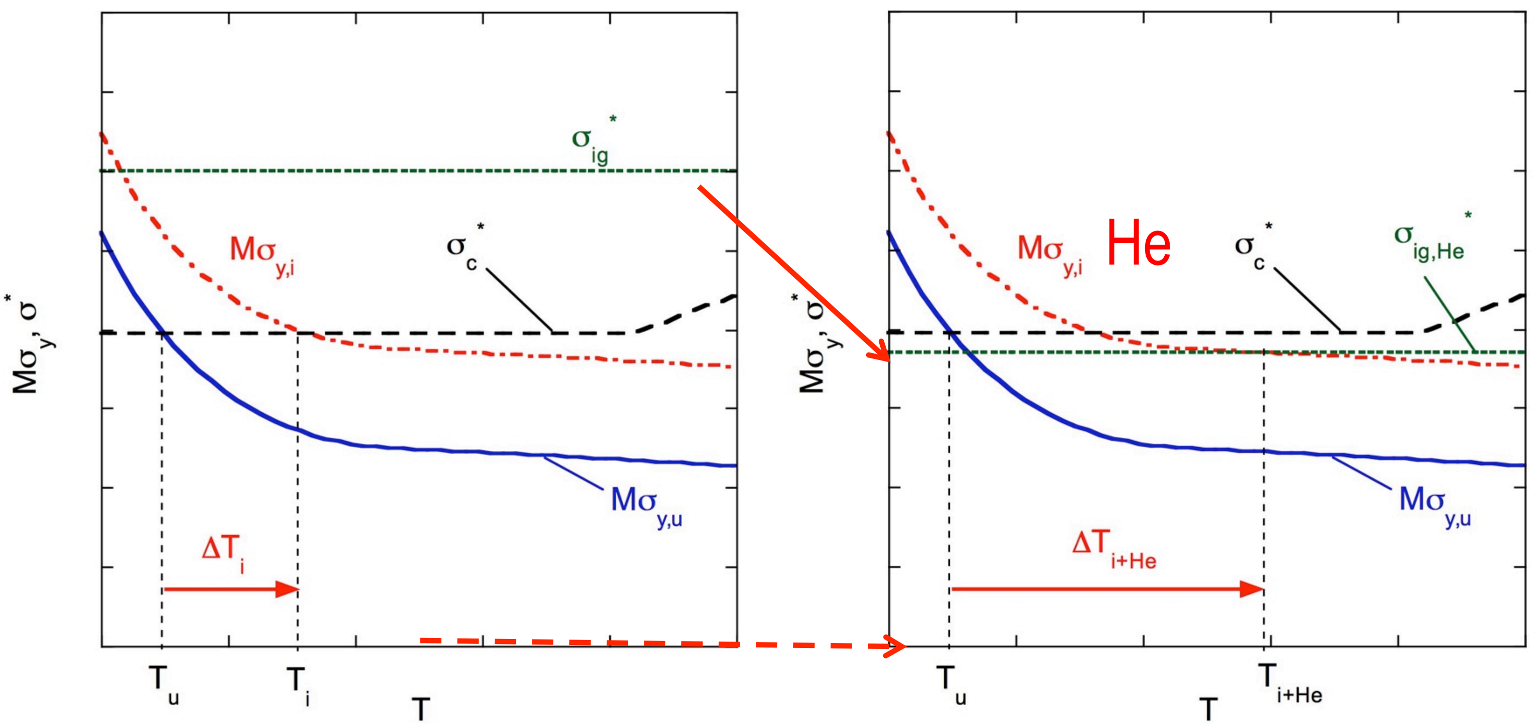


Wang, et al, to be published

PSI

A Potential Helium Embrittlement Mechanism: reduced IG fracture stress due to He

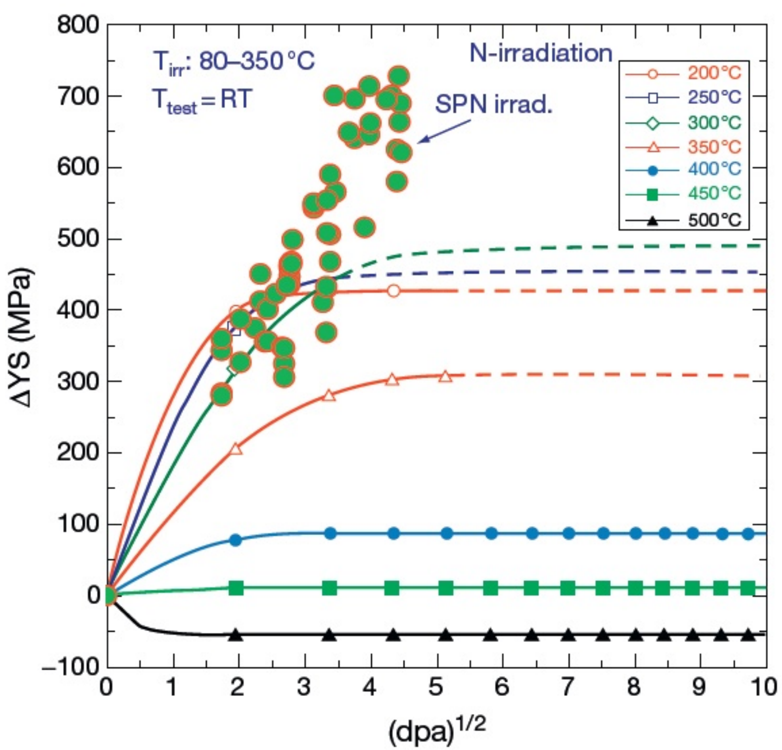
- Fracture *crack tip stress* $\sigma_t = M\sigma_y \geq$ critical stress σ^* at higher T for irradiated $\sigma_{ti} = M(\sigma_y + \Delta\sigma_y)$
- σ^* normally determined by transgranular cleavage σ_c^*, but He weakens GB so intergranular fracture at σ_{ig}^* (He) < σ_c^*
- ΔT reaches large values when $\sigma_{ig}^* < M(\sigma_y + \Delta\sigma_y)$ at high T

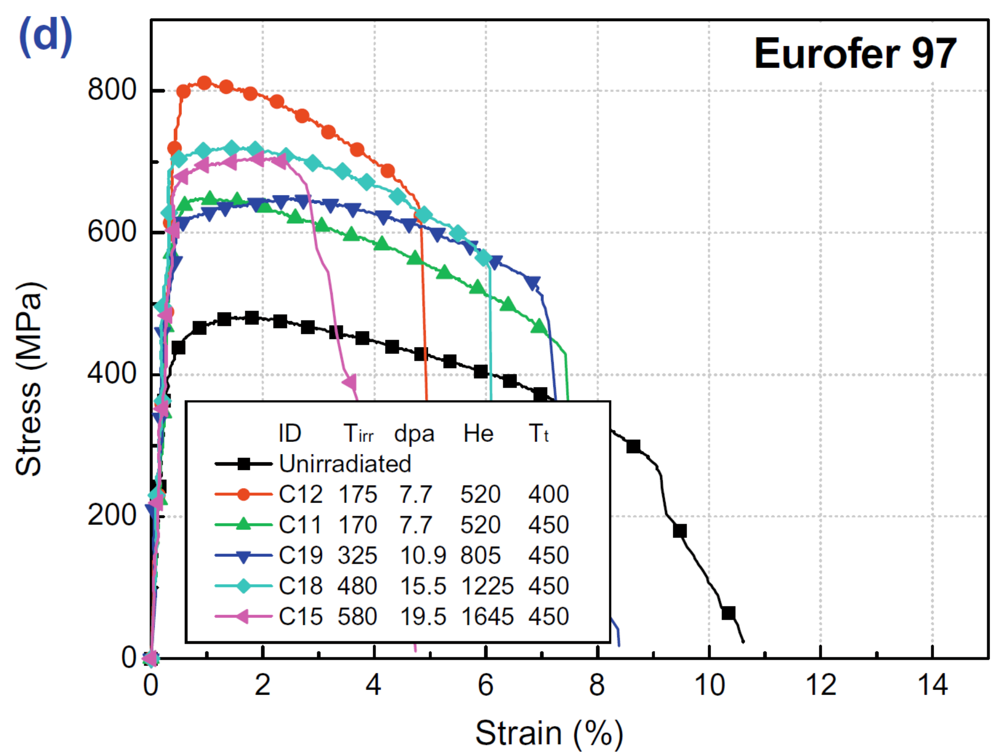


Y. Dai, G.R. Odette, T. Yamamoto, Comprehensive Nuclear Materials, vol. 1, R.J.M. Konings, Ed (2013)

Helium Effects on Yield Stress in irradiated ferritic/martensitic steels

High He extends $\Delta\sigma_y$ to > 700 MPa, and produces significant hardening even at 400-450°C

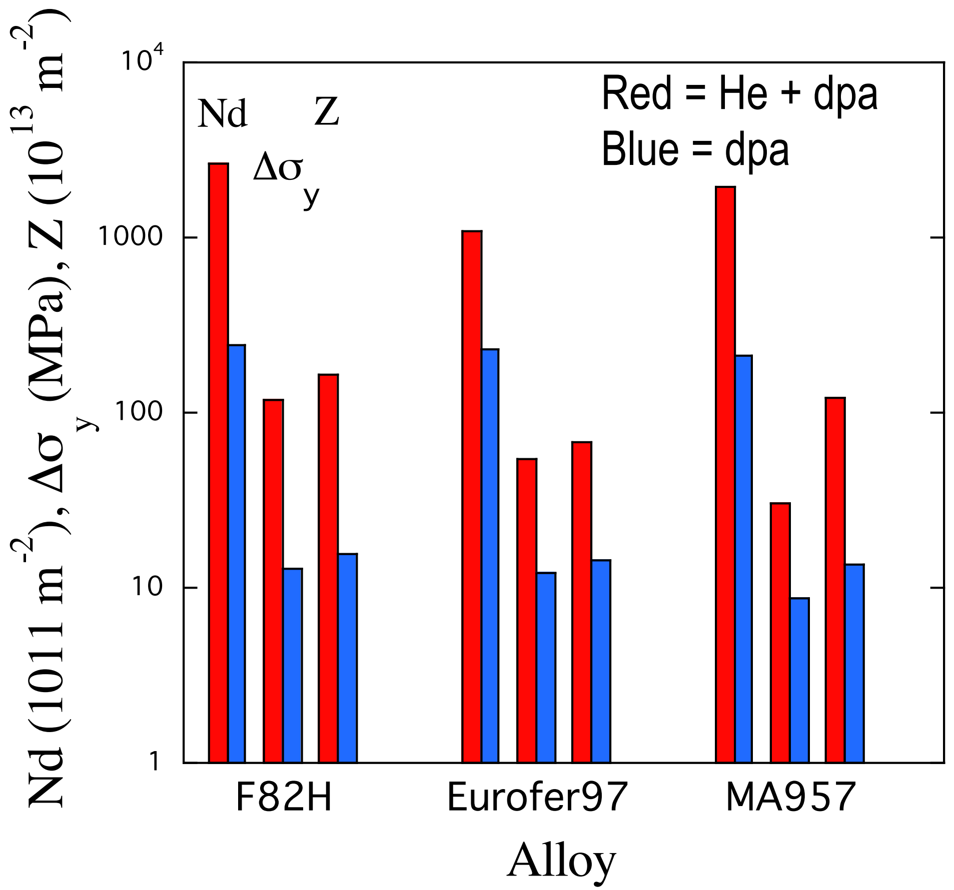




Y. Dai, G.R. Odette, T. Yamamoto, Comprehensive Nuclear Materials, vol. 1, R.J.M. Konings, Ed (2013)

Helium Effects on Loops, ΔYield Stress, and Sink Strength From ISHI Experiments

- High He also promotes dislocation loops with higher density, N, and larger size, d
- N*d ≈ 5 – 10 times greater than for displacement damage only – thus higher $\Delta\sigma_y$ and sink strength Z



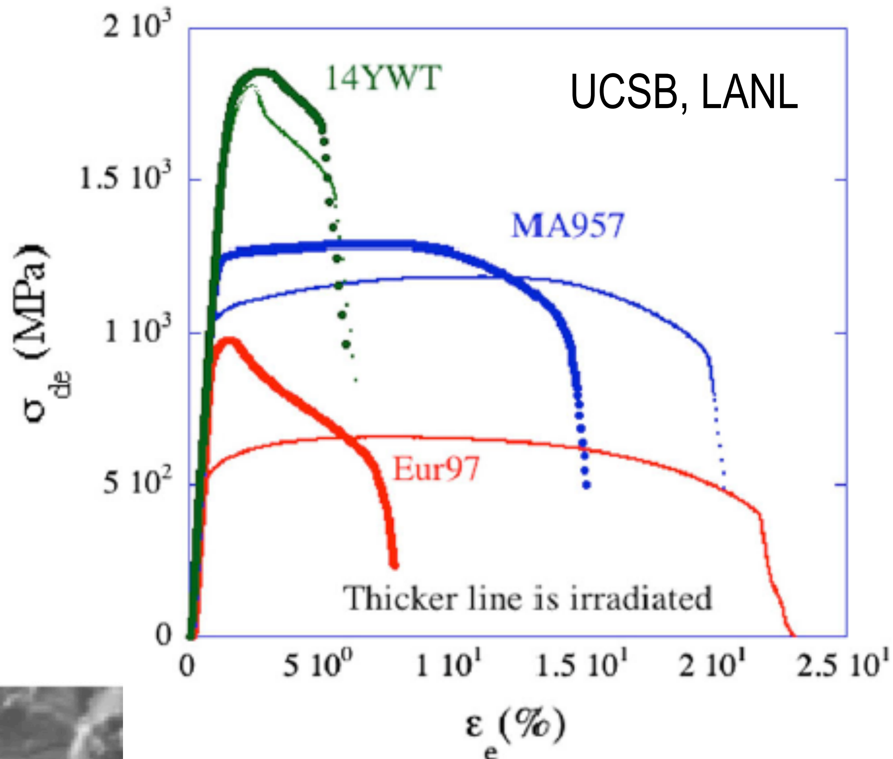
Predicted embrittlement due to enhanced He hardening extends to 500°C (vs. 400°C for fission neutron conditions

Predicted ΔT_0 is less than observed, suggesting possible role of He weakening of grain boundaries

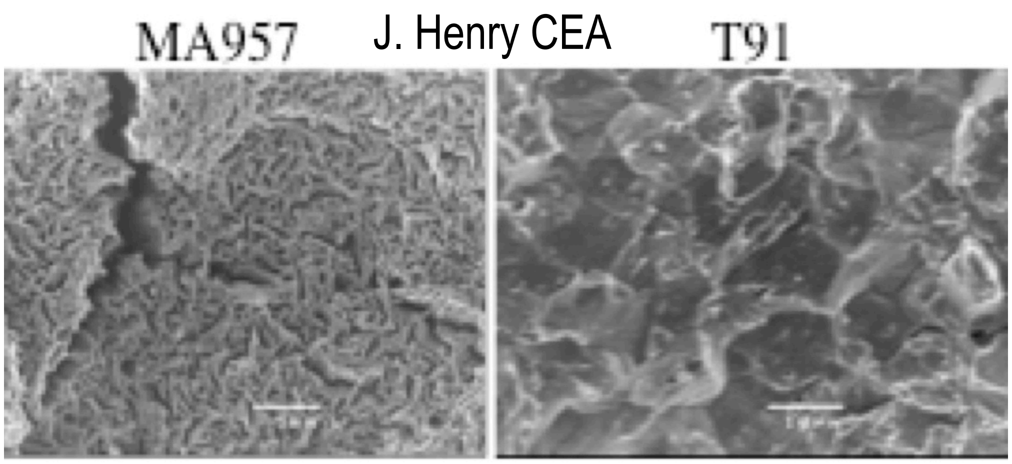
PNNL, UCSB

NFA Yield Stress Increases and Fracture Mechanism

- Irradiated NFA have lower $\Delta\sigma_y$ and greater uniform stain/strain hardening versus tempered martensitic steels
- Ductile tensile fracture surfaces after STIP irradiations to ≈ 19 dpa and 1750 appm He at ≈ 300°C
- Recent NFA have moderate but adequate toughness and strength mediated tearing



J. Henry CEA



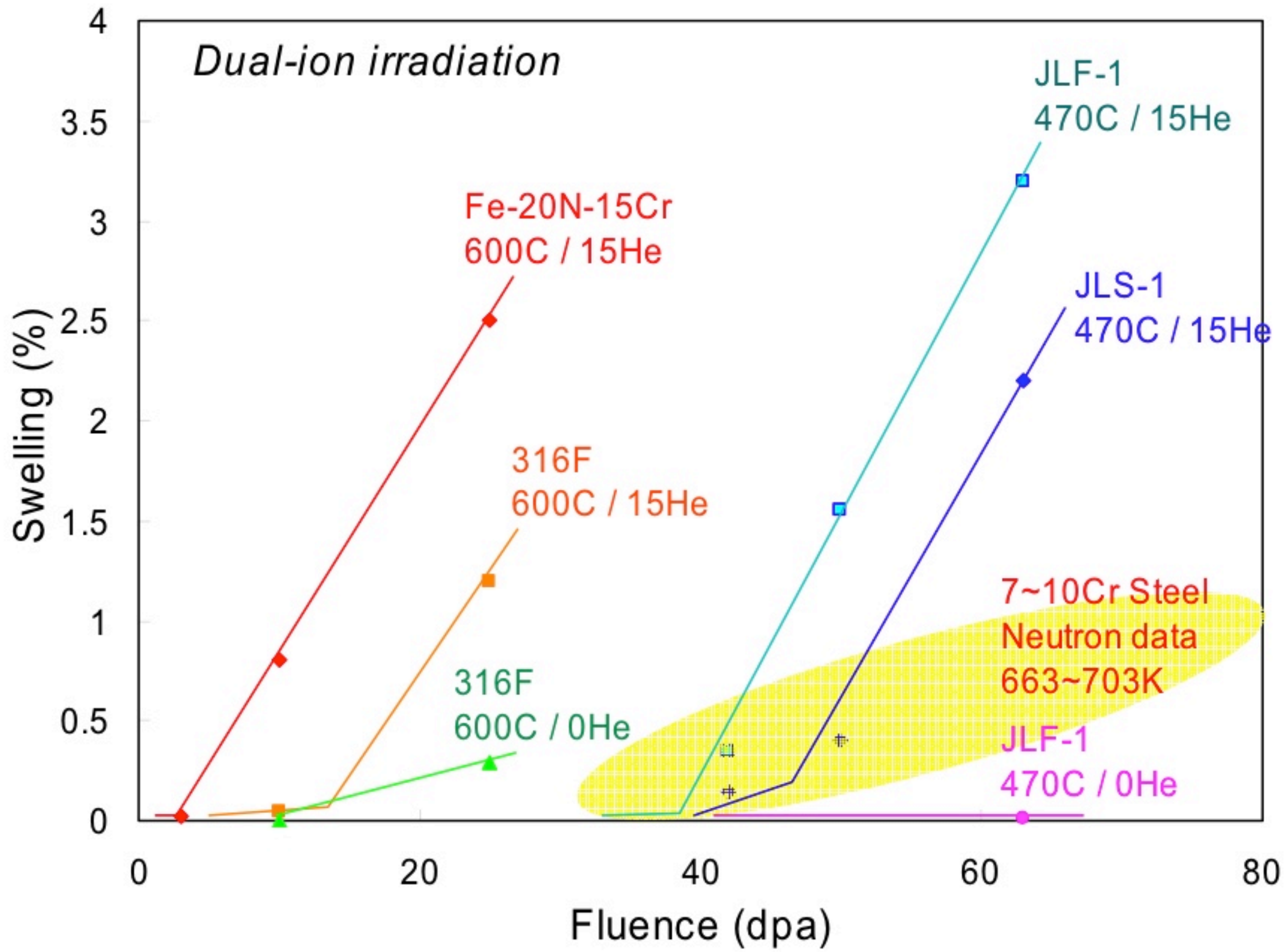
G. R. Odette, JOM 66, no.12 (2014) 2427

S. A. Maloy et al., JNM 468 (2016) JNM 232

Summary - Mechanical Properties & Helium Effects

- Tempered martensitic steels (TMS) experience enormous DBTT shifts
- The onset of more severe He embrittlement is at ~ 500 appm He
- The transition to intergranular fracture occurs at high He levels due to synergistic hardening + lower grain boundary cohesive strength
- At very high He levels hardening does not saturate at moderate dpa or decrease rapidly at high temperatures, and may cause change of deformation mode from dislocation glide to twinning
- **He extends large synergistic temperature shifts in TMS to higher dpa (no saturation) and temperatures (> 500°C)**
- Nanostructured ferritic alloys harden less, maintain considerable uniform stain and strain hardening capacities and are resistant to severe embrittlement and intergranular fracture

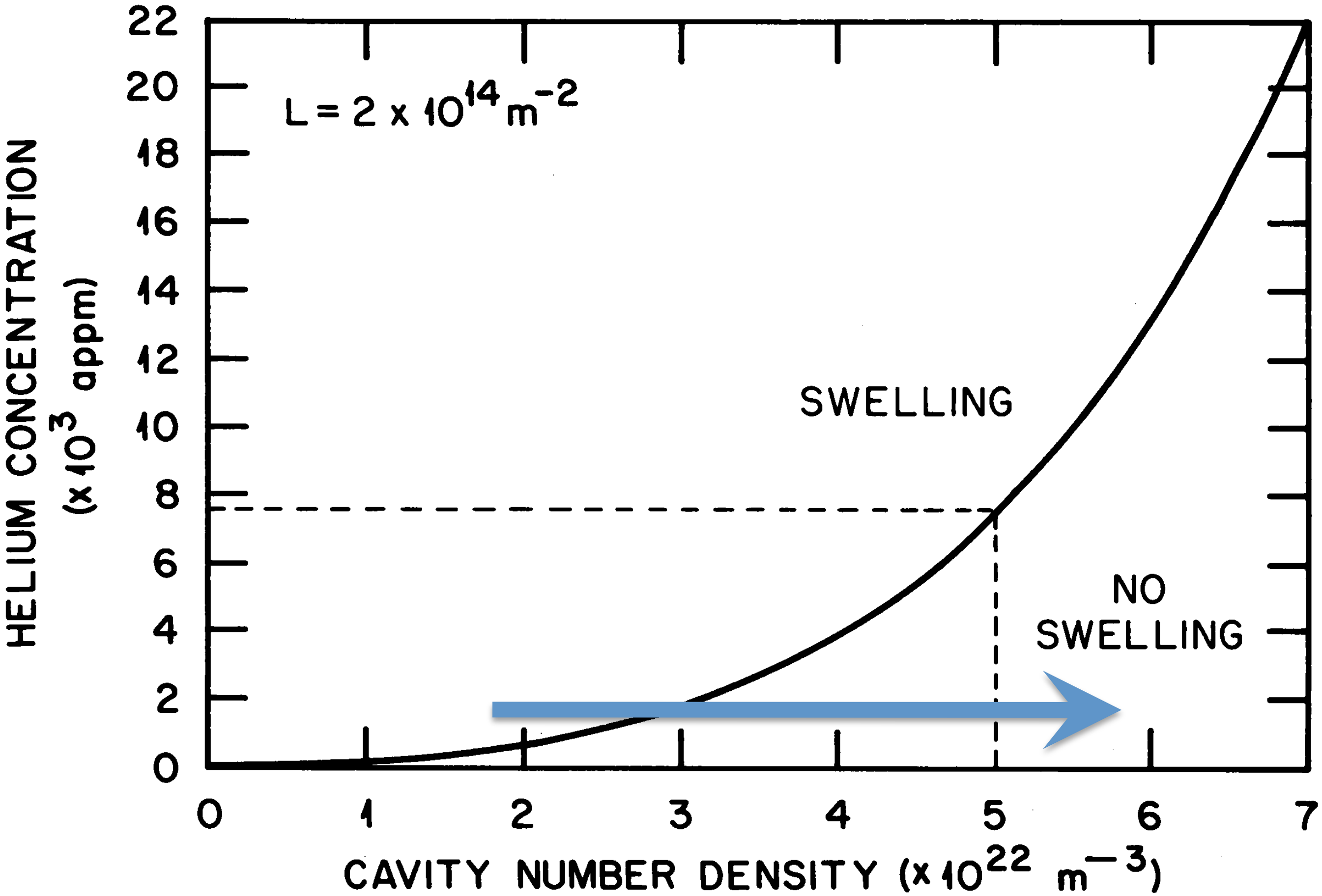
Cavity swelling of Ferritic/martensitic Steel is a Concern for Fusion-relevant He/dpa ratios



Y. Katoh et al., JNM (2003)
H. Ogiwara et al. JNM (2002)

- Swelling behavior of RAFMs and austenitics at around peak-swelling temperatures are similar in the presence of helium, except for incubation dose.

Bubble to void conversion requires a much higher He concentration when cavity number density is high

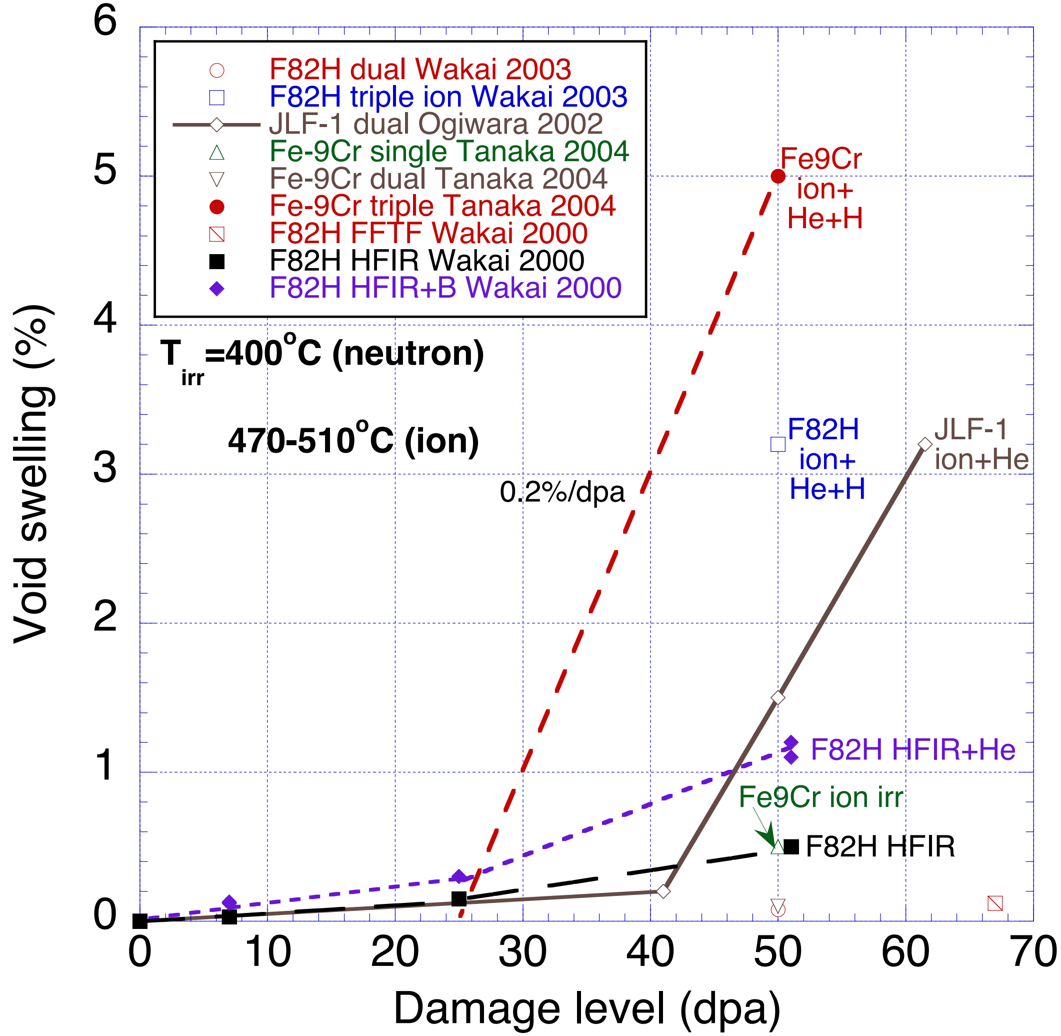


Moderate He/dpa ratio (e.g., DT fusion neutrons)

High He/dpa ratio (e.g., spallation neutrons)

L.K. Mansur & E.H. Lee, J. Nucl. Mater. 179-181 (1991) 1021

Cavity swelling in neutron-irradiated 8-9%Cr ferritic-martensitic steels may become significant for fusion-relevant (~10 appm/dpa) He/dpa values



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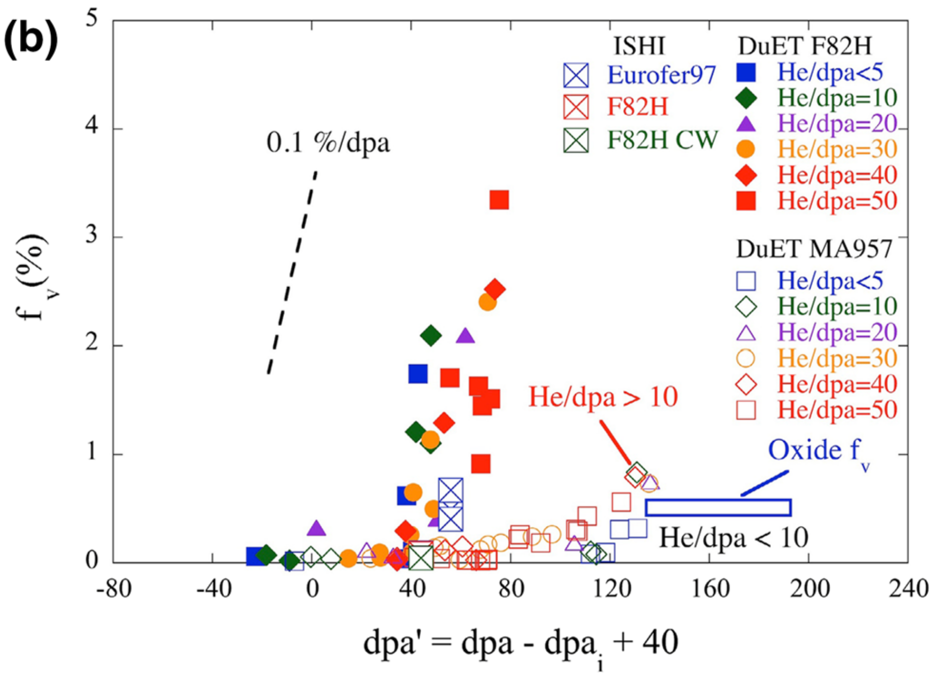
S.J. Zinkle, et al., Nucl. Fusion (2016), special issue on Fusion Materials, in press

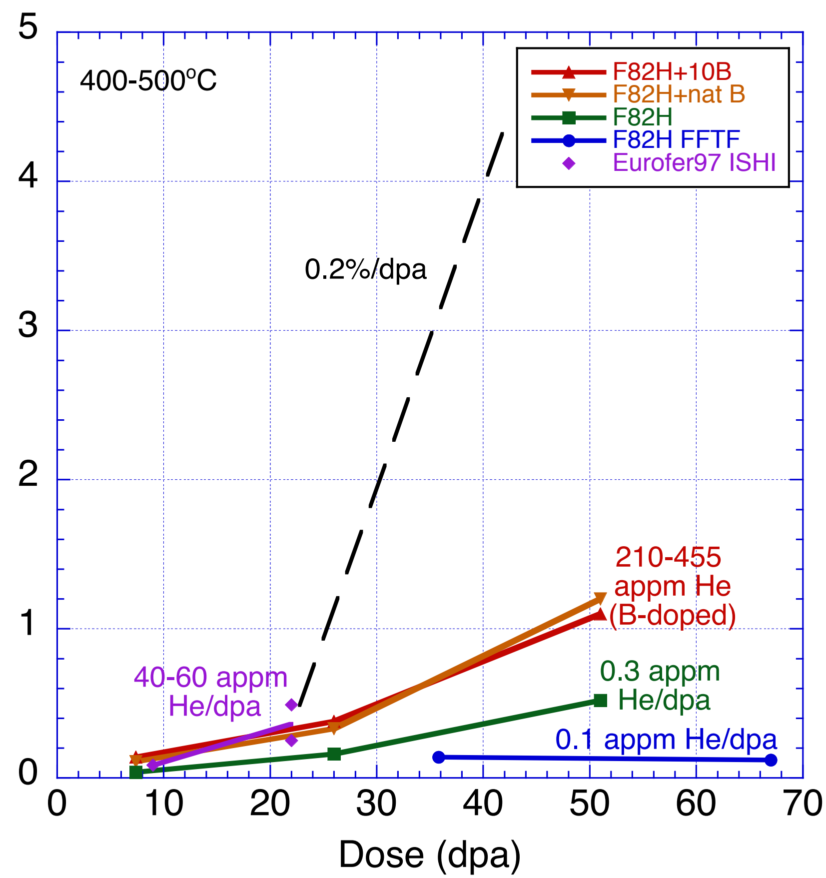
Cavity swelling in irradiated 8-9%Cr reduced activation ferritic-martensitic steels may become unacceptable above ~50 dpa (~500 appm He)

Dual Ion irradiation
(6.4 MeV Fe + 0.2-1 MeV He)



G.R. Odette, JOM 66, 12 (2014) 2427

Fission neutron irradiation



Zinkle, Möslang, Muroga & Tanigawa, Nucl. Fusion 53, 10 (2013) 104024

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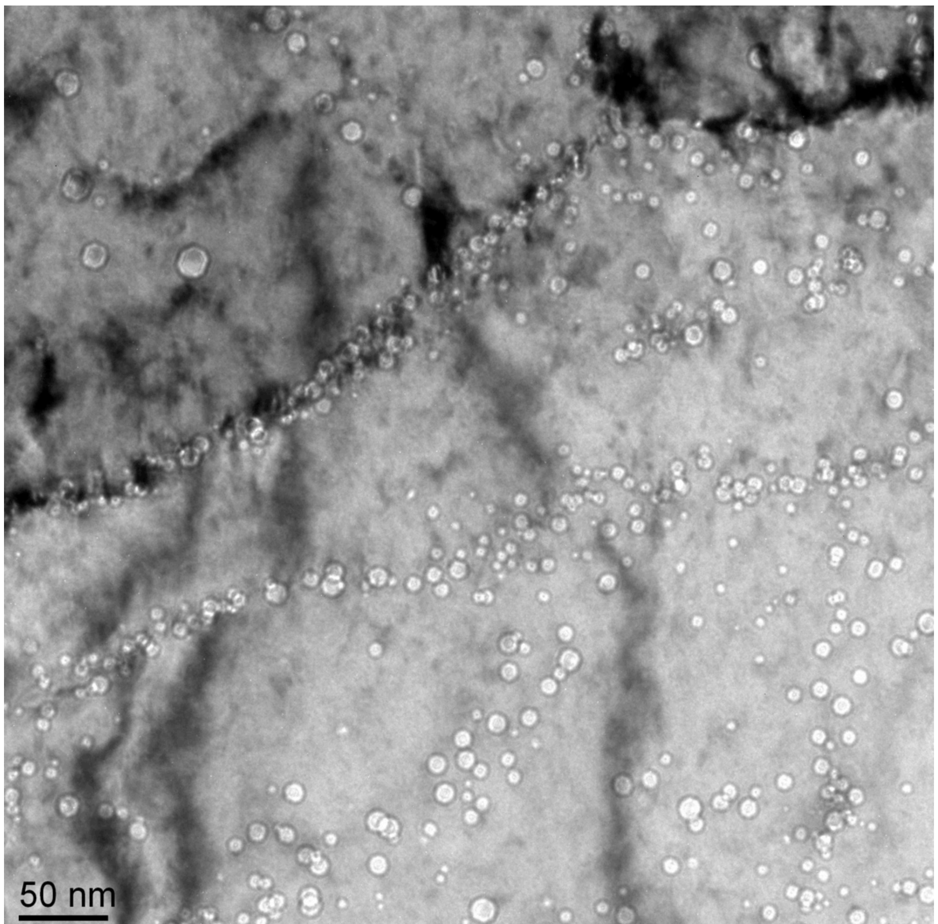
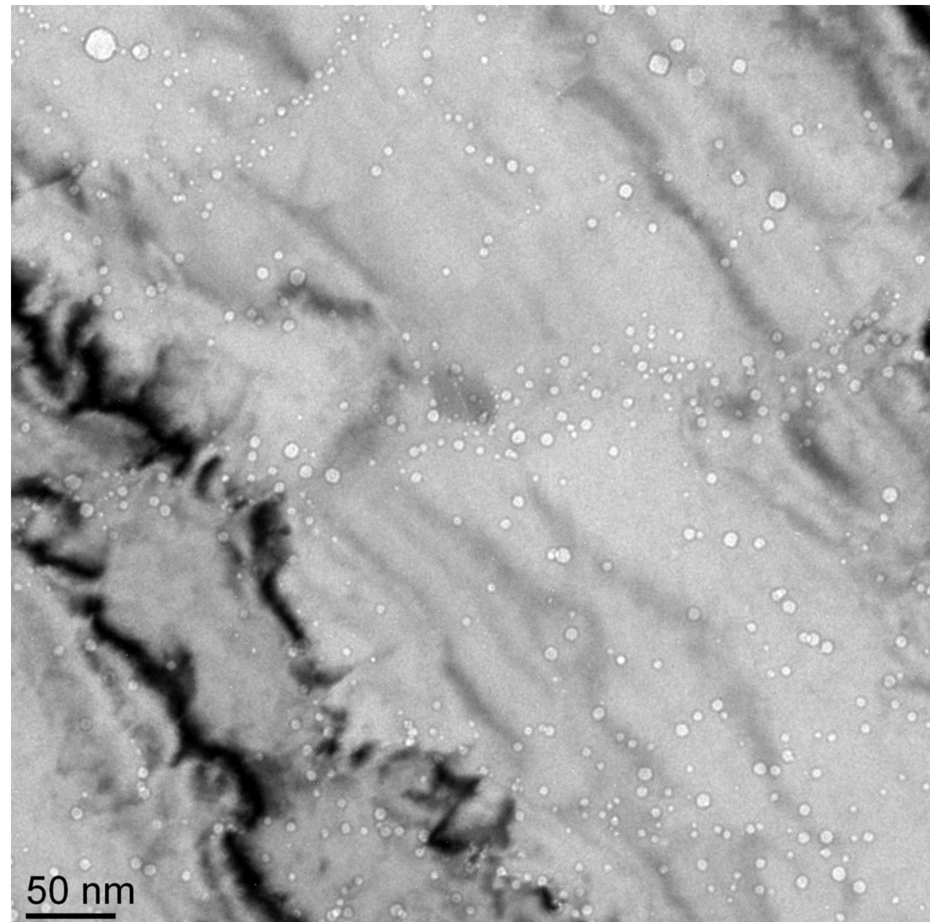
Cavity Microstructure in Dual Ion Irradiated F82H Mod 3 ferritic/martensitic steel

UCSB



As Tempered

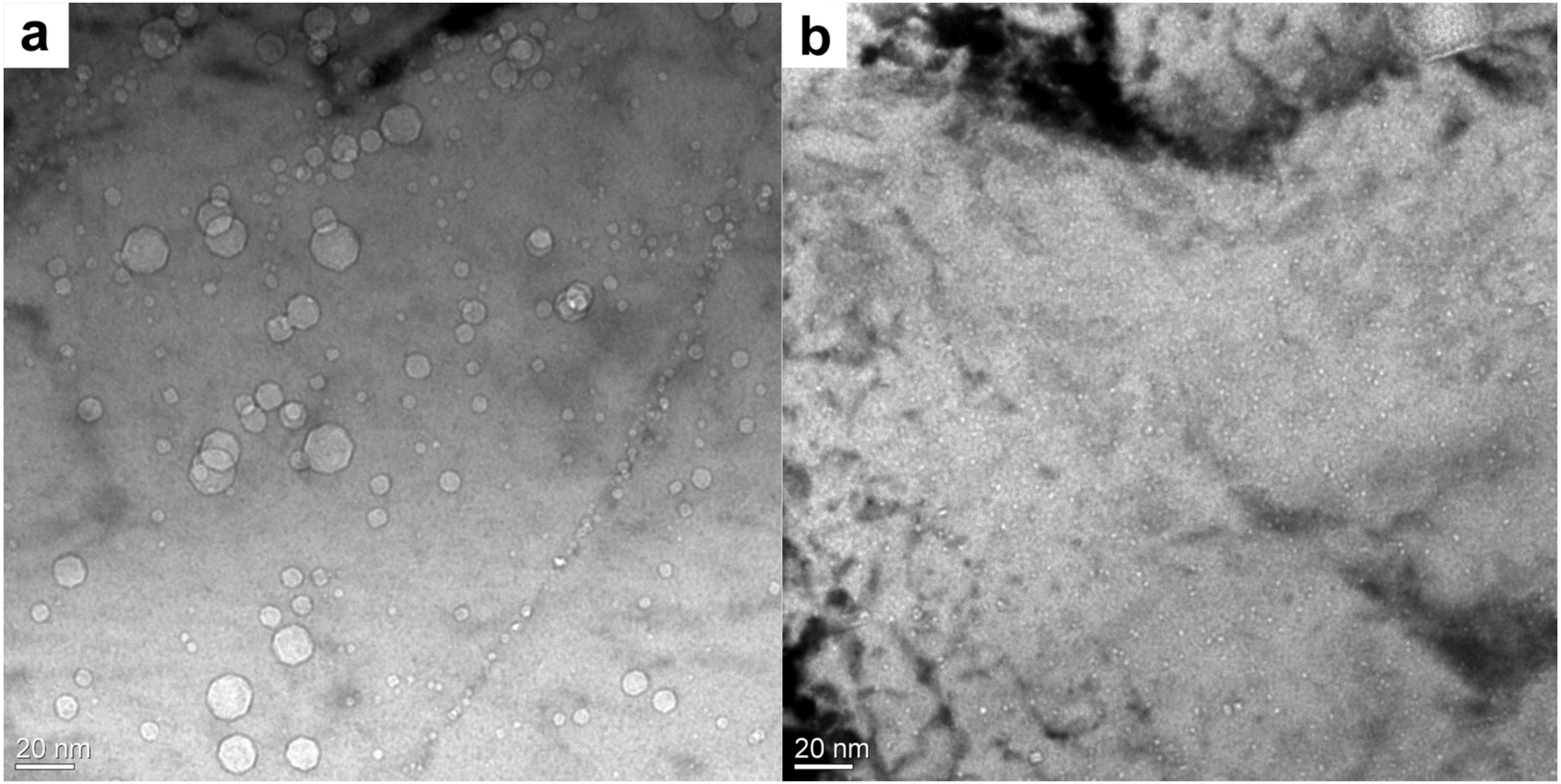


20% Cold Work

Cavity swelling is suppressed at ultra high sink strengths

1400 appm He and 25 dpa at 500°C (56 appm/dpa)

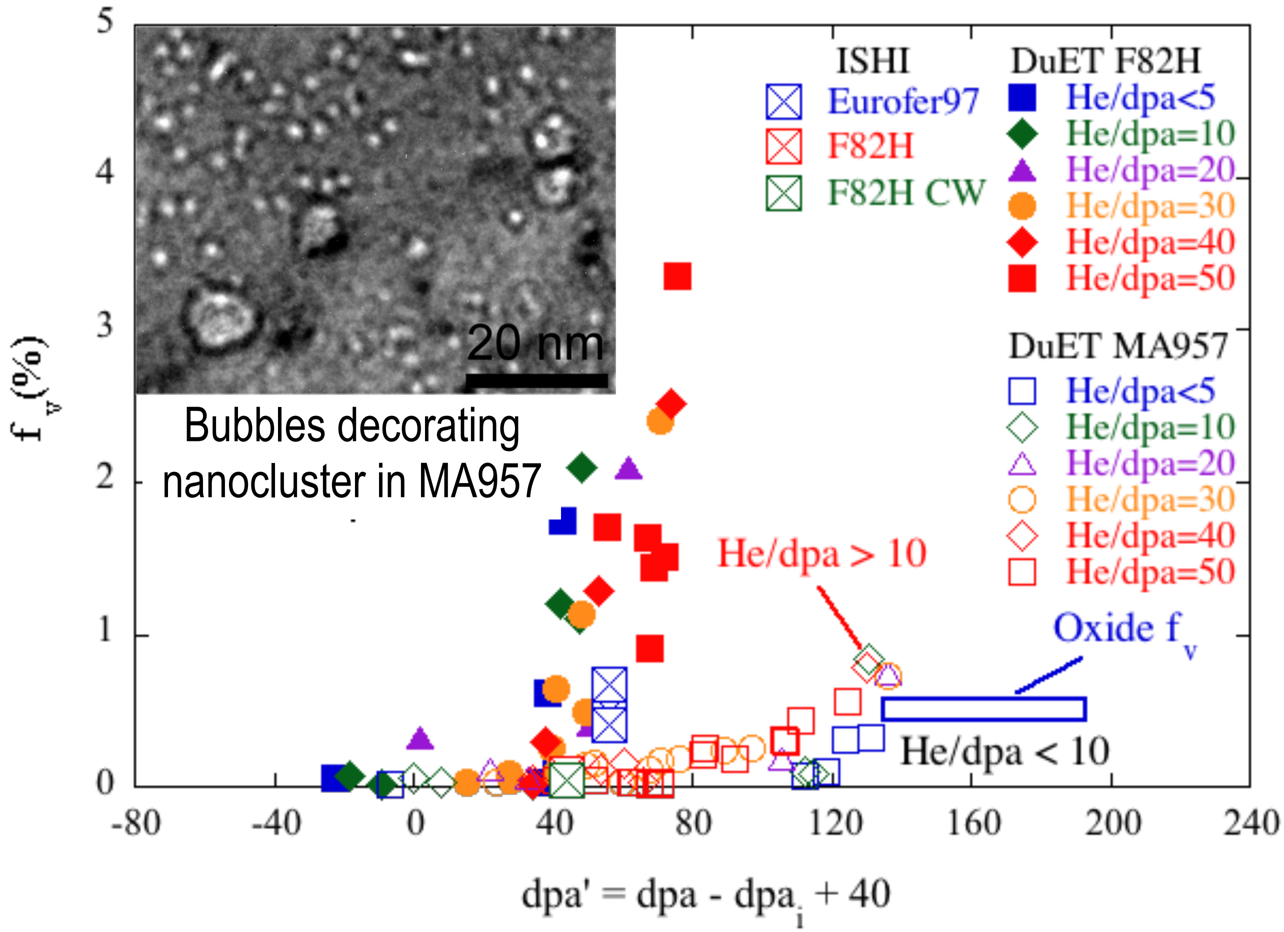
Eurofer97 9%Cr | MA957 (ODS steel)



G.R. Odette et al.

Managing Helium in Nanostructured Ferritic Alloys

- Only bubbles (no voids) in nanostructured ferritic alloys at 500 and 650°C
- Bubbles form on 1 - 4 nm pyrochlore $Y_2Ti_2O_7$ nano-oxides (NO)

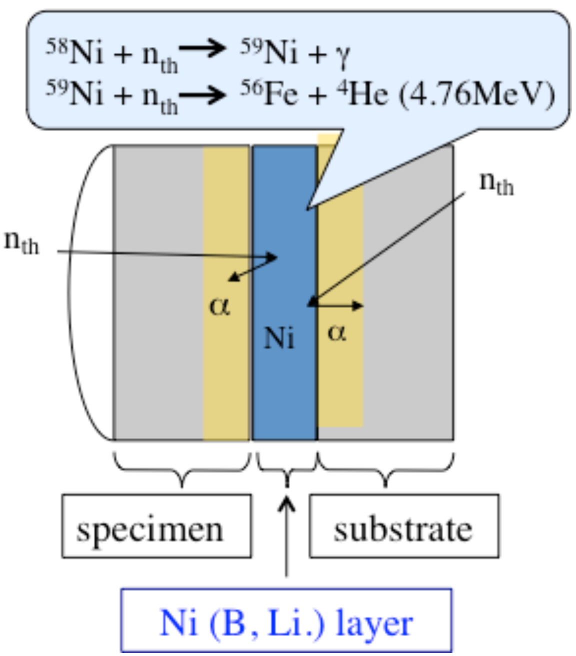


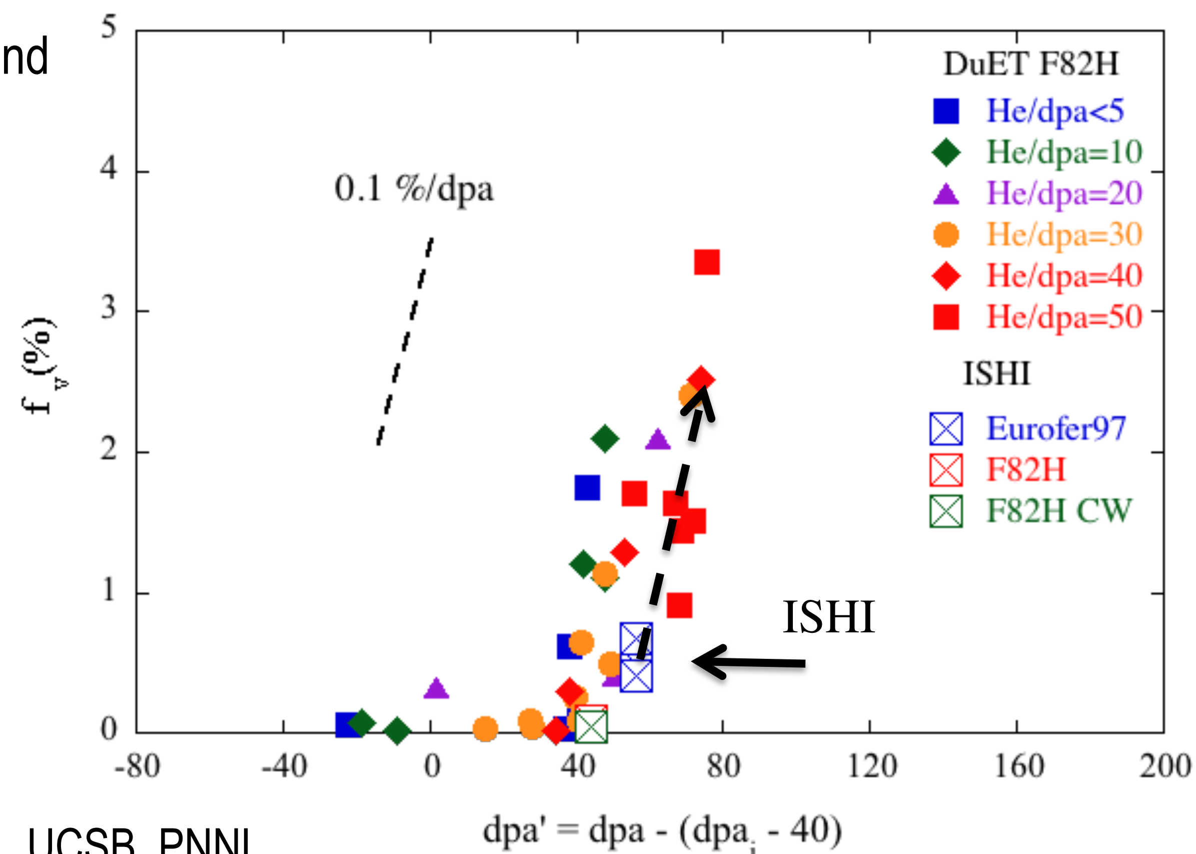
G.R. Odette, JOM 66, 12 (2014) 2427

UCSB, Kyoto, PNNL

In Situ Helium Injection (ISHI) in HFIR

- HFIR ISHI He/dpa ≈ 0 to 50 (HFIR JP26 – 29, also UCSB ATR-I)
- Plot ISHI f_v data on the same DII dose scale: dpa' = dpa – [dpa_i(He/dpa) – 40]
- ISHI 9 & 21 dpa 500°C @ He/dpa ≈ 50 and dpa_i ≈ 5
- ISHI data fall near DII trend band

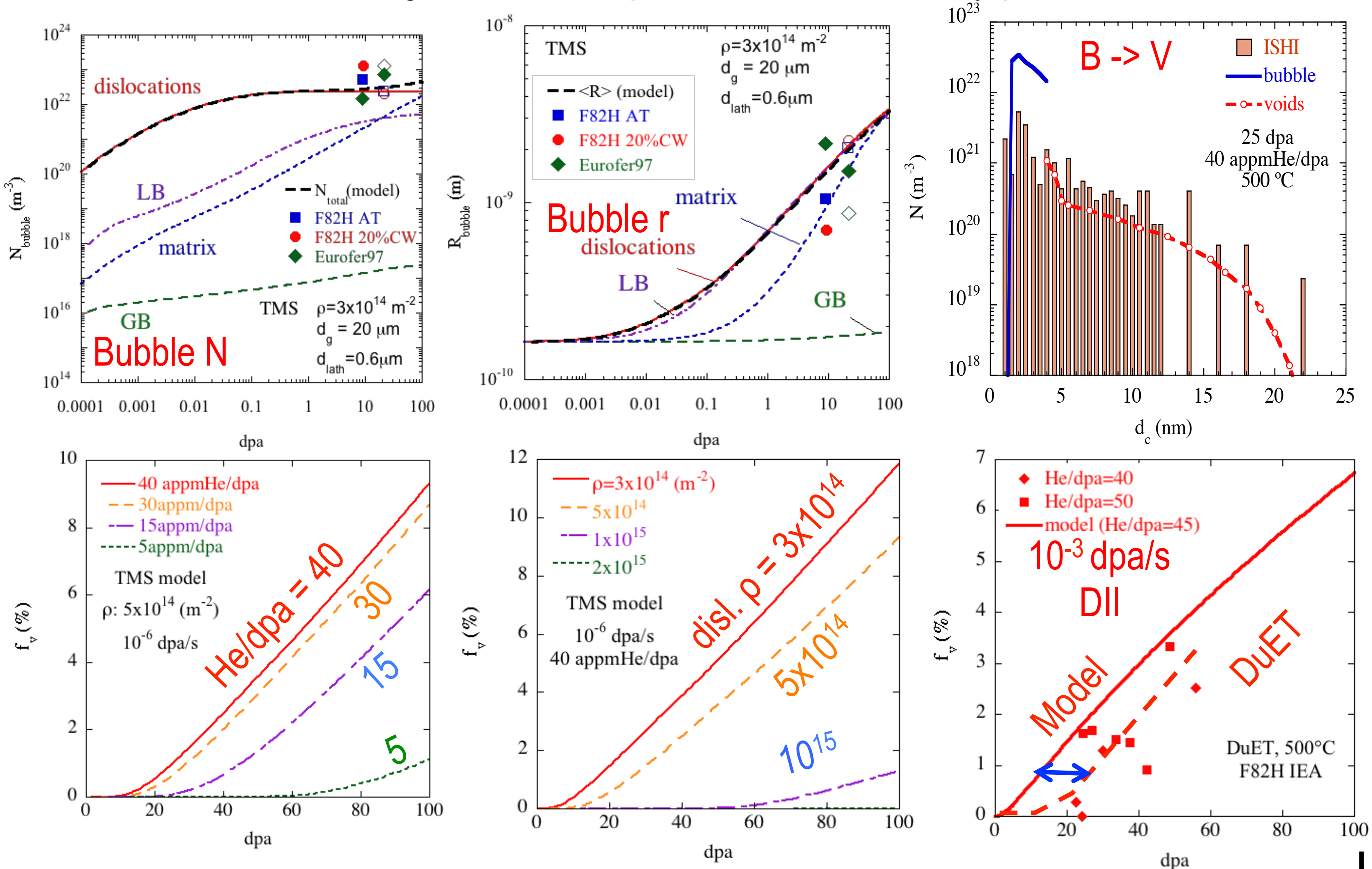




UCSB, PNNL

He Transport, Fate & Consequences Master Model Predictions for Tempered Martensitic Steel

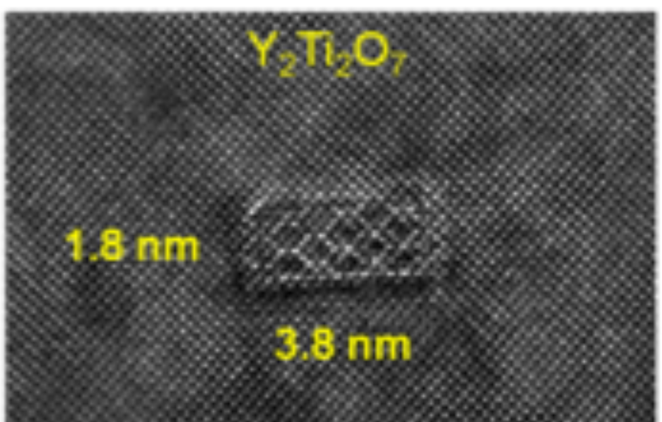
Atomistic/microstructure based rate theory model accounting for He partitioning, bubble to void conversions and integrated with experiment – reasonably predicts most trends



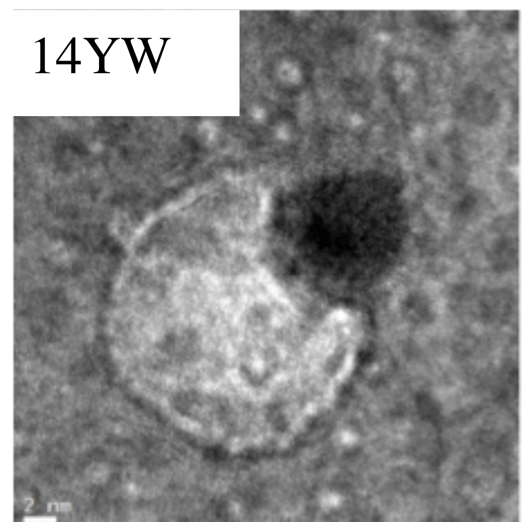
UCSB

NFA He Transport & Fate: Matrix ➠ Oxide ➠ Bubble ➠ Void (Large Oxide Particles Only)

- DFT (Y. Jiang et al.) He in $Y_2Ti_2O_7$ and Y_2TiO_5
- He energy far lower in nm-scale oxides versus the Fe matrix ≈ 2.25 eV versus ≈ 0.94 eV – initially deeply trapped
- Energy even lower in gas bubble above a minimum size
- Sequence: Matrix ➠ Oxide ➠ Bubble of little consequence for small bubbles
- Large oxides/bubbles: Matrix ➠ Oxide ➠ Bubble ➠ Void Swelling
- “Good” particles are numerous and small; undesirable particles are large, lower density



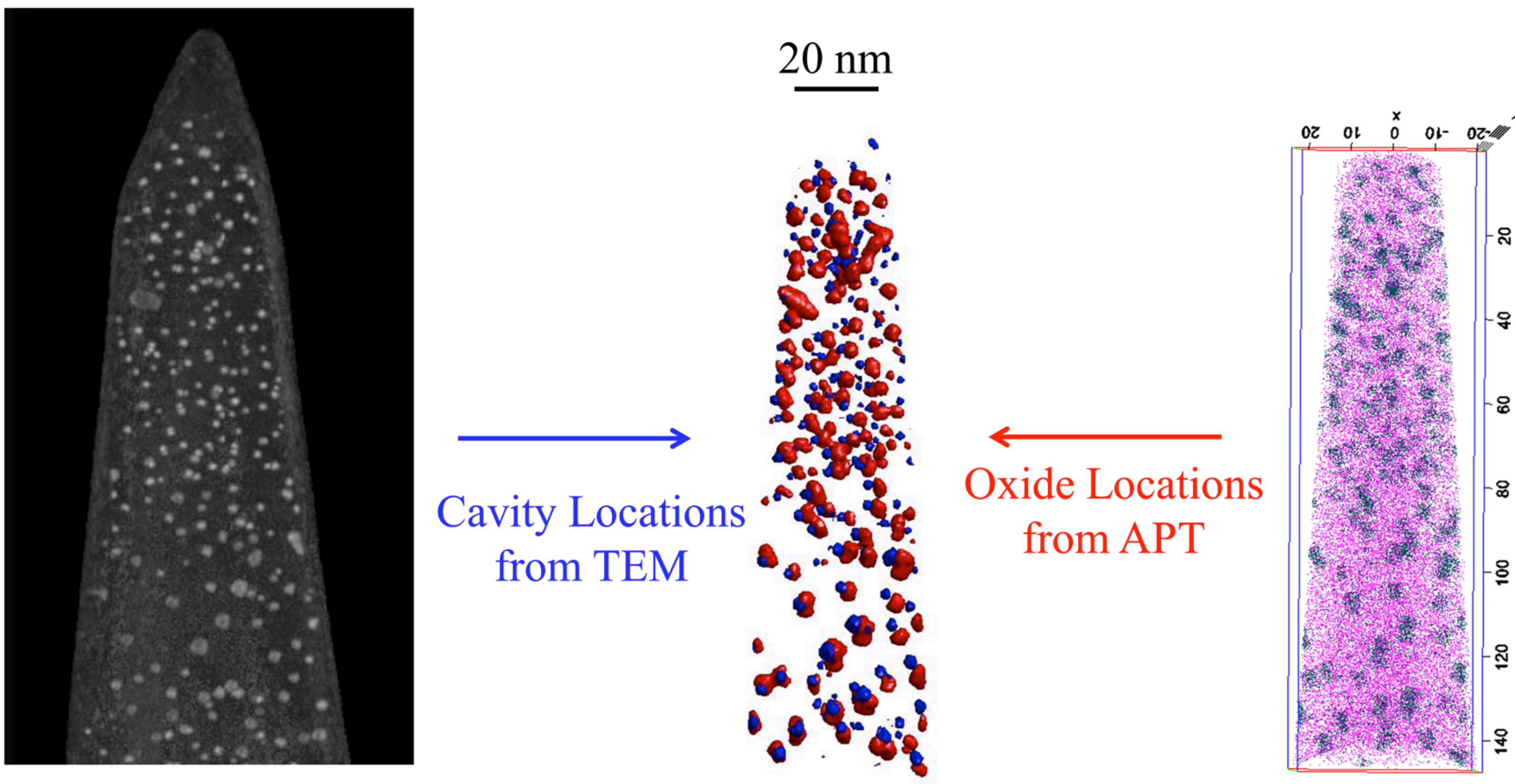
UCSB, PNNL, Central South University, China



G.R. Odette, JOM 66, 12 (2014) 2427

3D Nanoscale Correlative Microscopy and Atom Probe Tomography indicate nanoclusters are effective trapping sites for He cavities

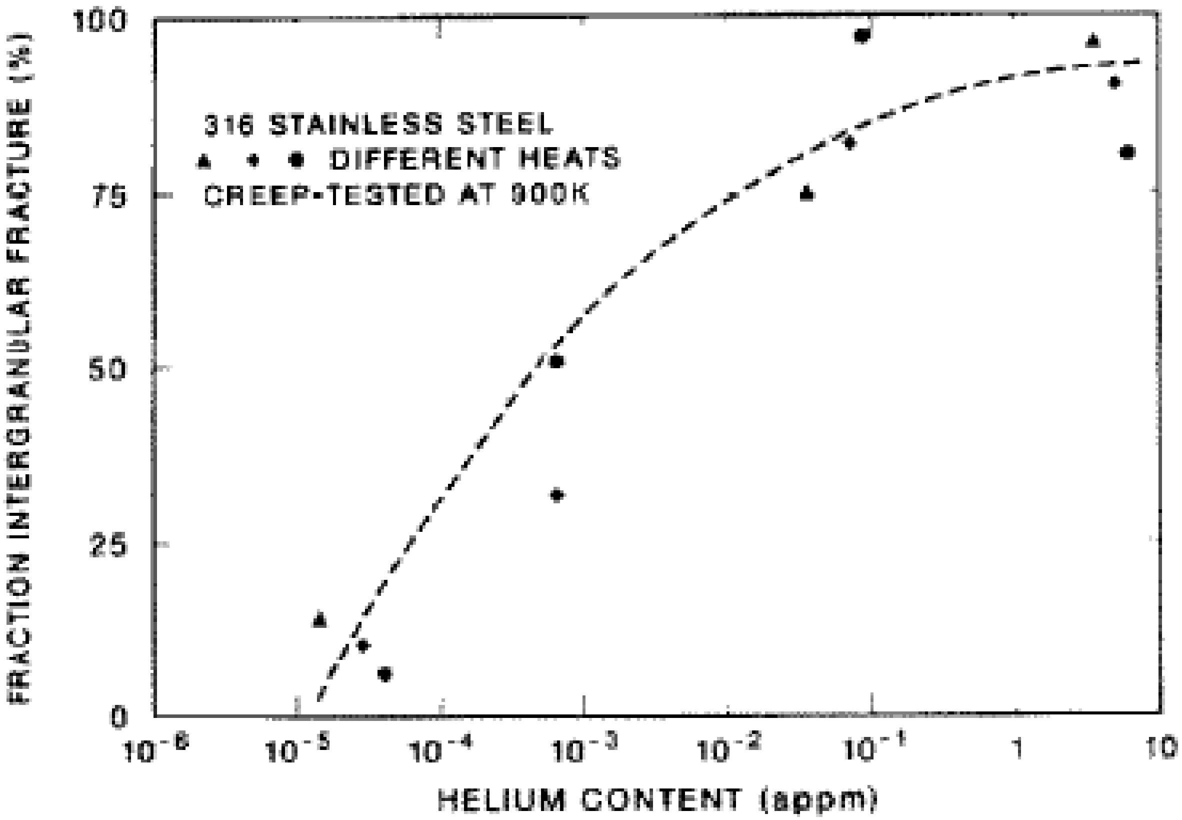
TEM **Combined** **Atom Probe**



UCSB

Similar conclusions regarding effectiveness of nanoclusters for He cavity sequestration also recently reported by C.D. Parish et al, J. Nucl. Mat., in press

Helium Embrittlement in 316 SS emerges for C_{He}>0.01 appm during slow strain rate testing at 625^{o}C

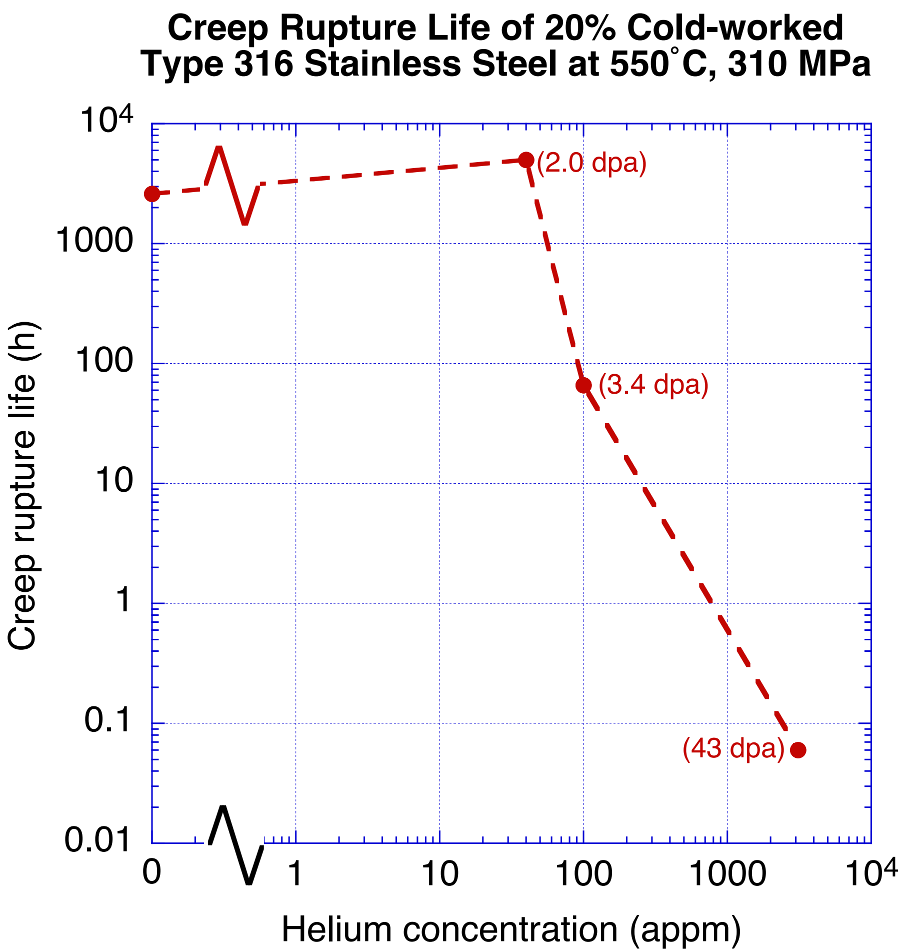


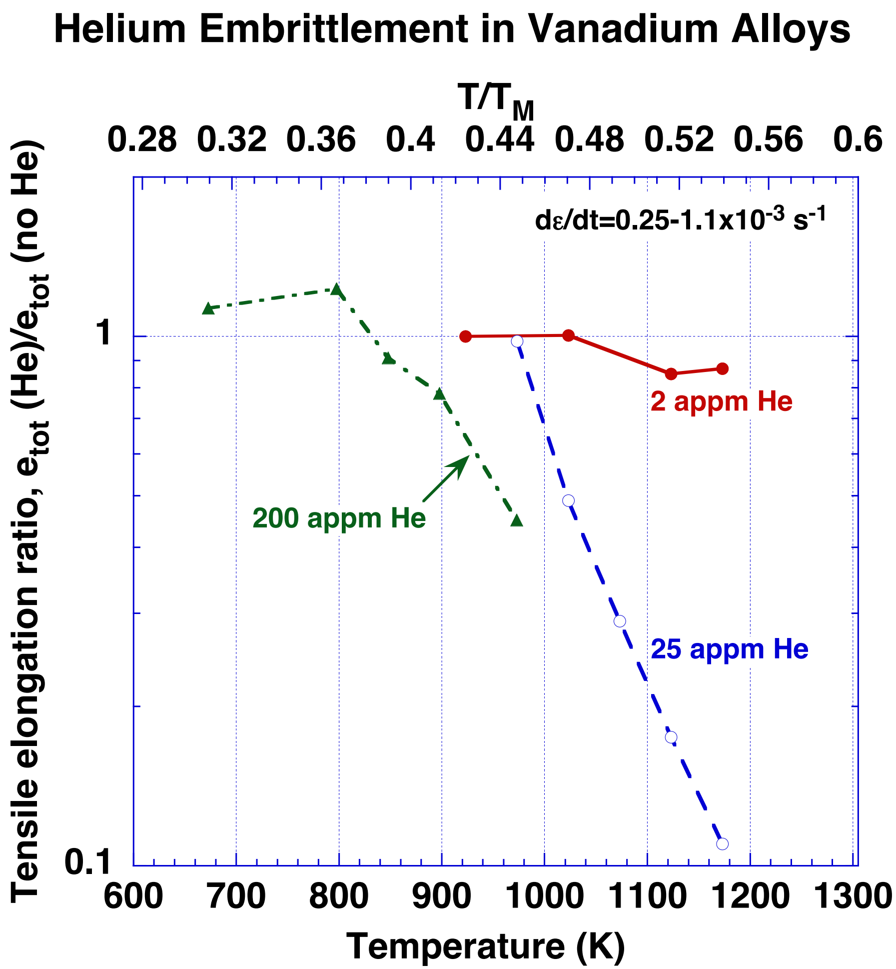
Initial creep stress: 191 MPa

Embrittlement via Intergranular fracture is dependent on helium content, temperature, and strain rate

B. van der Schaaf and P. Marshall, in Dimensional stability and mechanical behaviour of irradiated metals and alloys, vol. 1 (BNES,1983) p. 143

Helium embrittlement of grain boundaries occurs at high temperatures for helium concentrations above ~100 appm



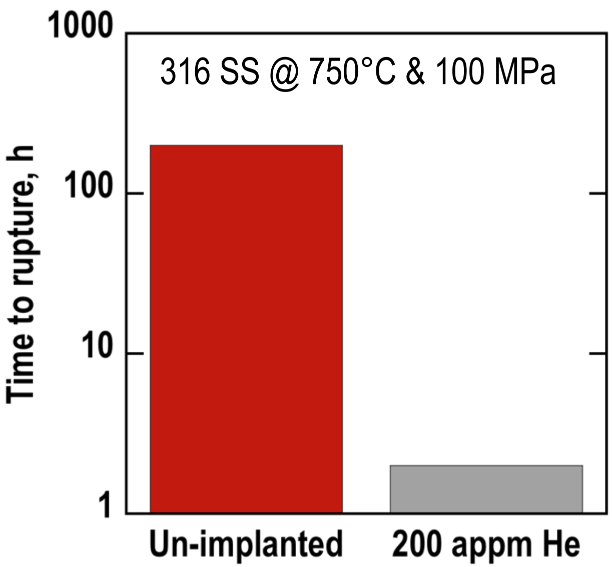


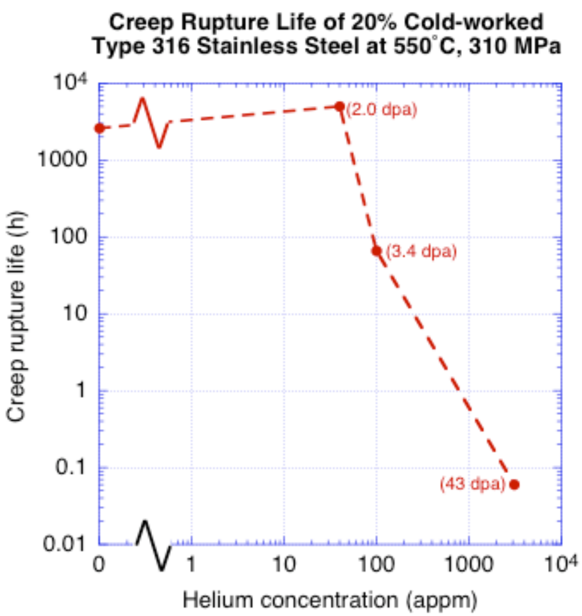
E.E. Bloom & F.W. Wiffen, J. Nucl. Mater. 58 (1975) 171

He trapping at nanoscale precipitates within grains is key for inhibiting He embrittlement

However…….. The formation and microstructural stability of these precipitates is strongly affected by irradiation parameters, in particular the He/dpa ratio

High temperature He grain boundary embrittlement can be severe: High density of nanoscale He trapping sites may be needed to mitigate the effect

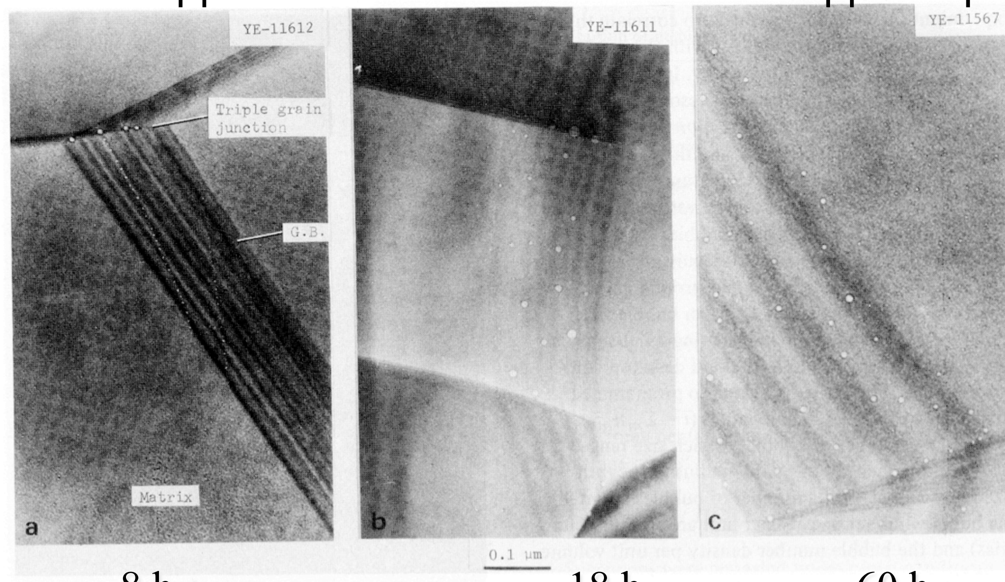




Effect of applied stress on evolution of 160 appm implanted He

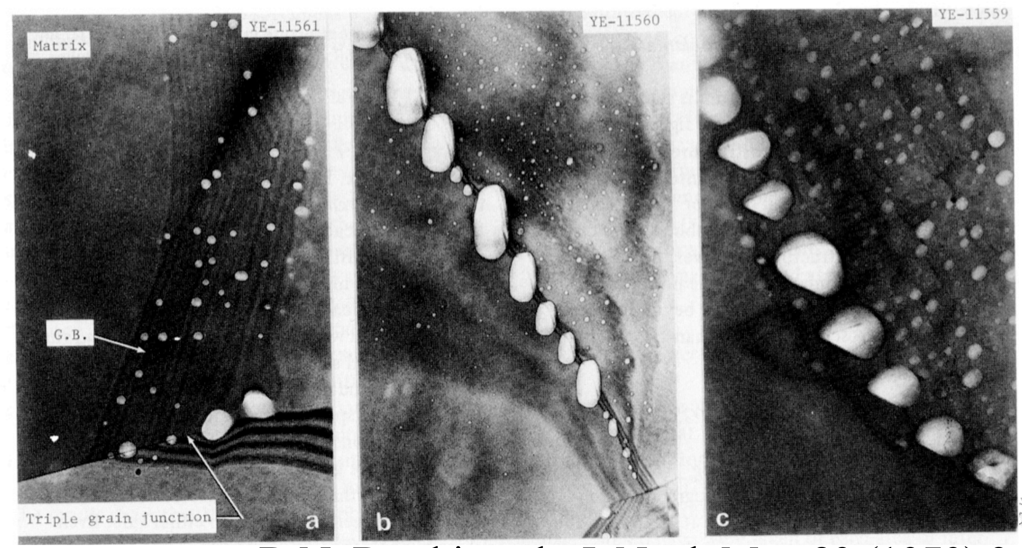
750°C

Fe-17Cr-17Ni



0 MPa

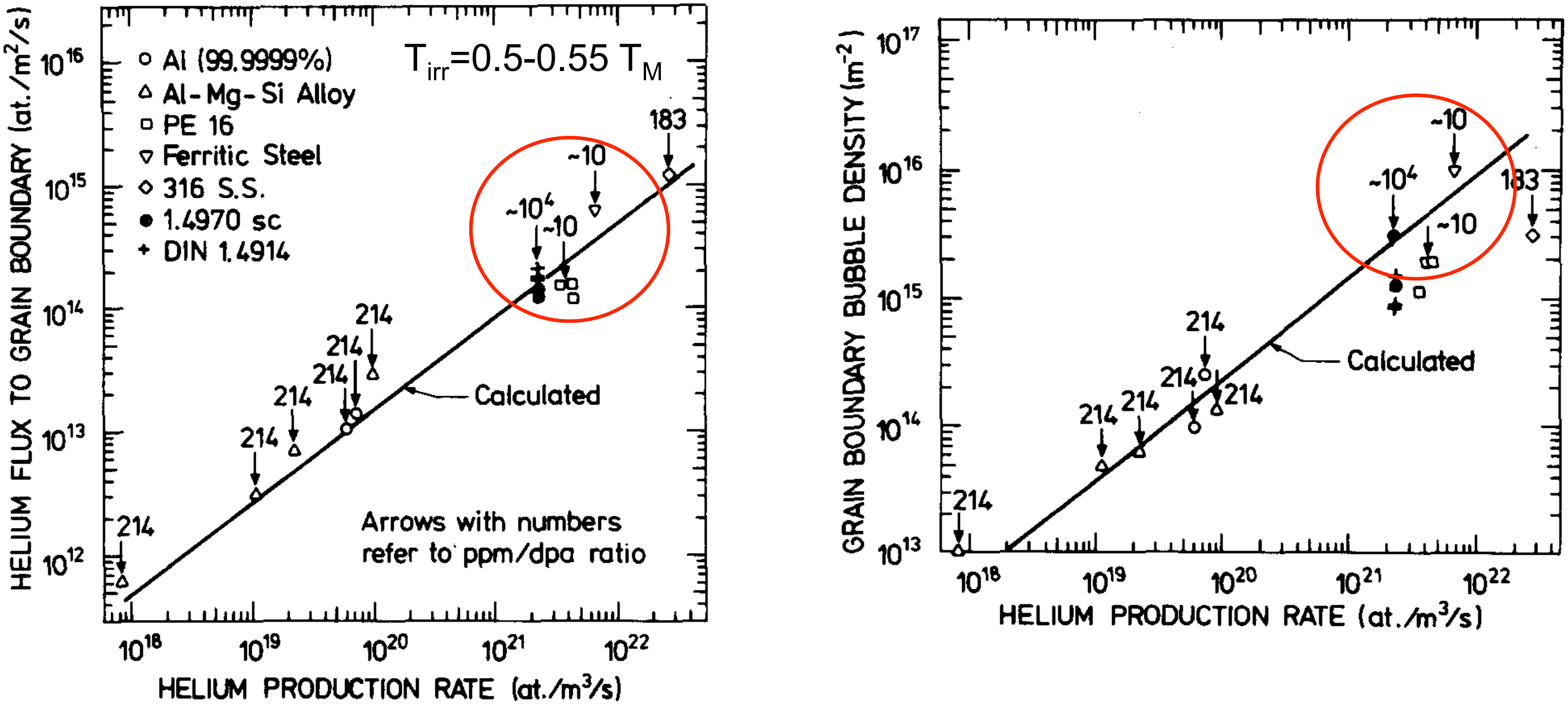
Fig. 2. Growth of helium bubbles in unstressed Fe–17 Cr–17 Ni specimens after annealing at 1023 K for (s) 2.88×10^4 s, (b) 6.48×10^4 s and (c) 21.60×10^4 s.



19.6 MPa

D.N. Braski et al., J. Nucl. Mat. 83 (1979) 265

It is tenuous to perform accelerated-rate studies of helium embrittlement phenomena at high temperatures



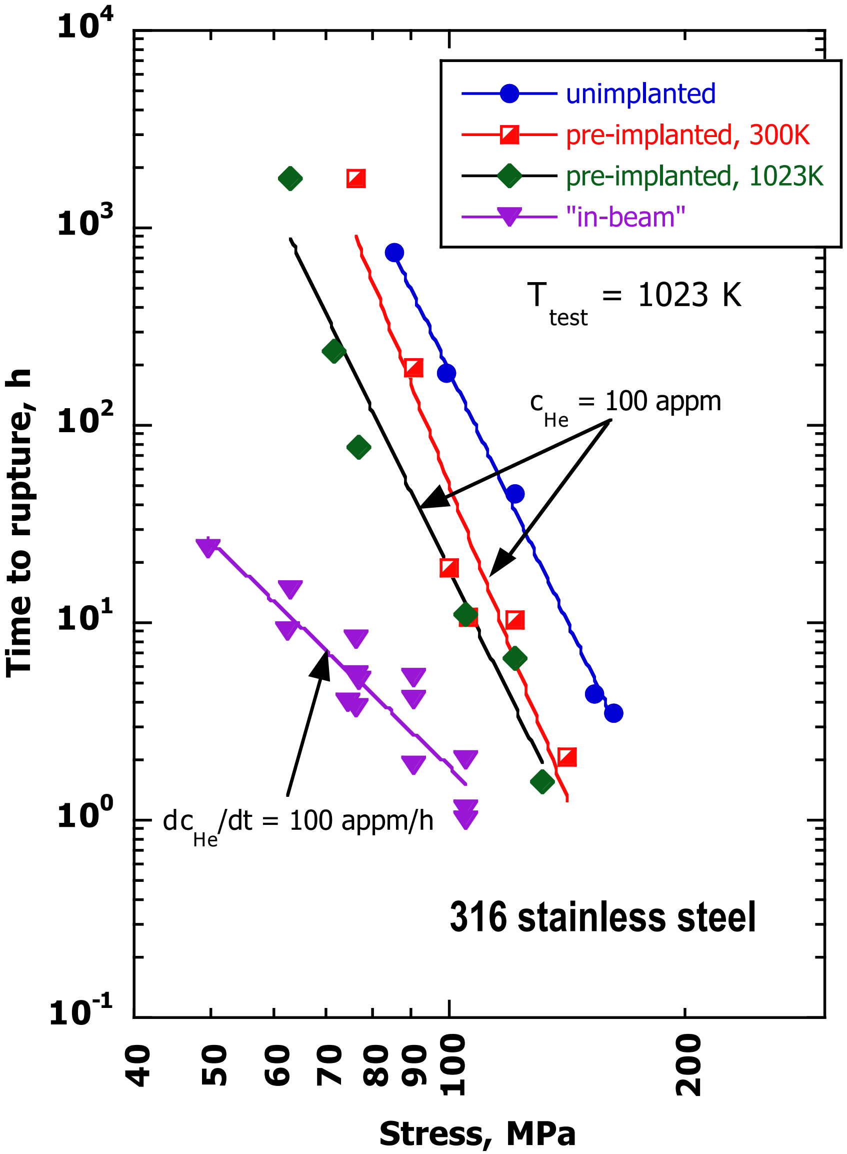
Numbers next to data refer to appm He/dpa ratios

Helium migration to grain boundaries at elevated temperatures is largely controlled by thermal diffusion, not by radiation damage

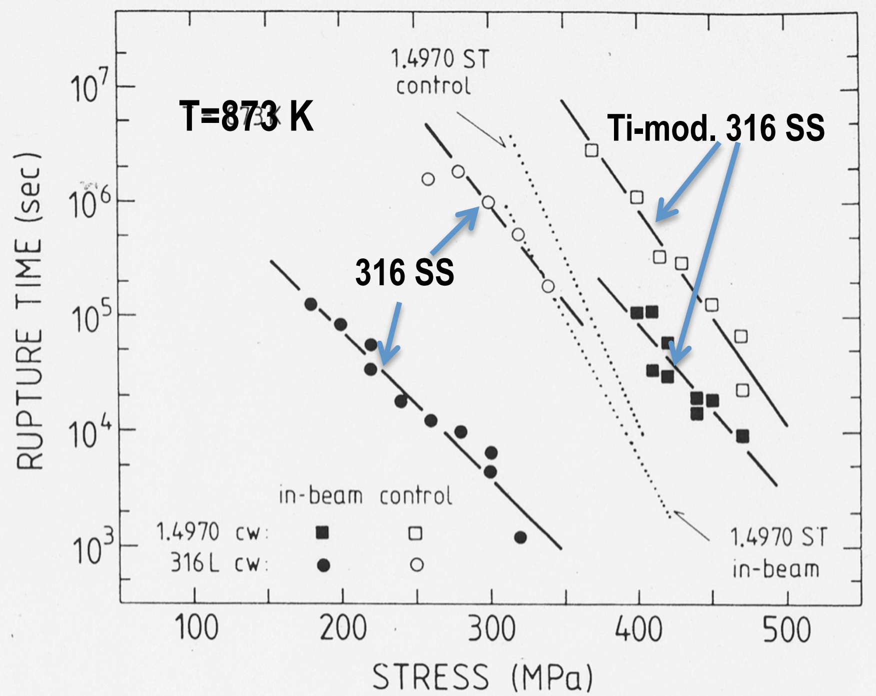
He flux and grain boundary cavity density depend on absolute He production rate (He/s), not He/dpa

B.N. Singh & A.J.E. Foreman, J. Nucl. Mater. 179-181 (1991) 990

High temperature helium embrittlement of austenitic SS



Schroeder and Batfalsky, J. Nucl. Mater. 117(1983)287



Yamamoto & Schroeder J.Nucl. Mat. 155-157 (1988)

Fundamental process involves migration of He to grain boundaries

- **Matrix cavity "overnucleation" for He preimplantation case**
  - **Once matrix cavities have formed, it is difficult for migration to g.b.'s to occur**
- **Co-implantation at high temperature is more favorable for diffusion to g.b.'s of implanted single He or small He clusters**
  - **Single He or small He-vacancy clusters have higher mobility than large bubbles**
- **TiC matrix ppts effectively trap He in matrix**

Effect of strain rate on high temperature He embrittlement in irradiated austenitic stainless steel

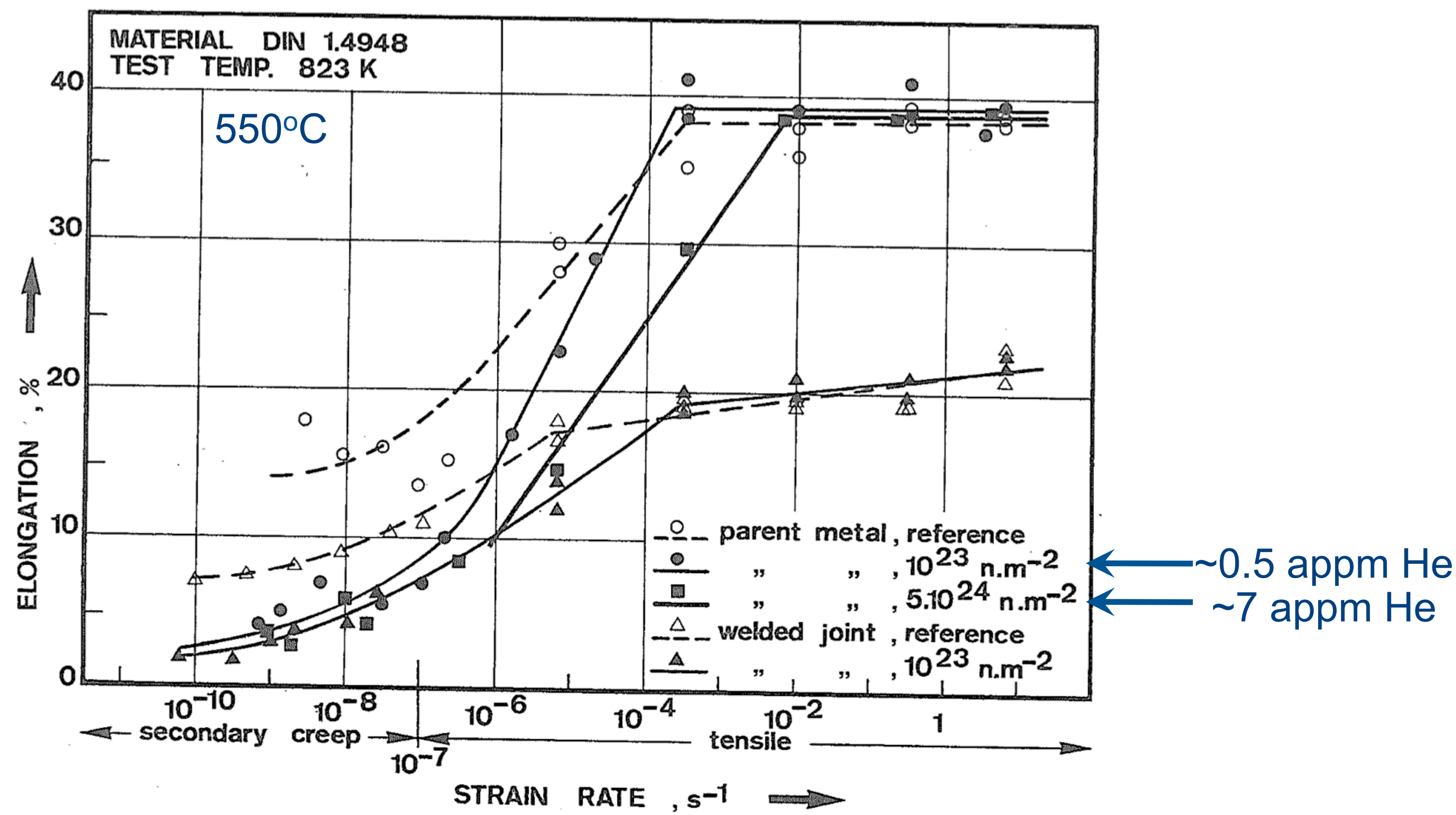


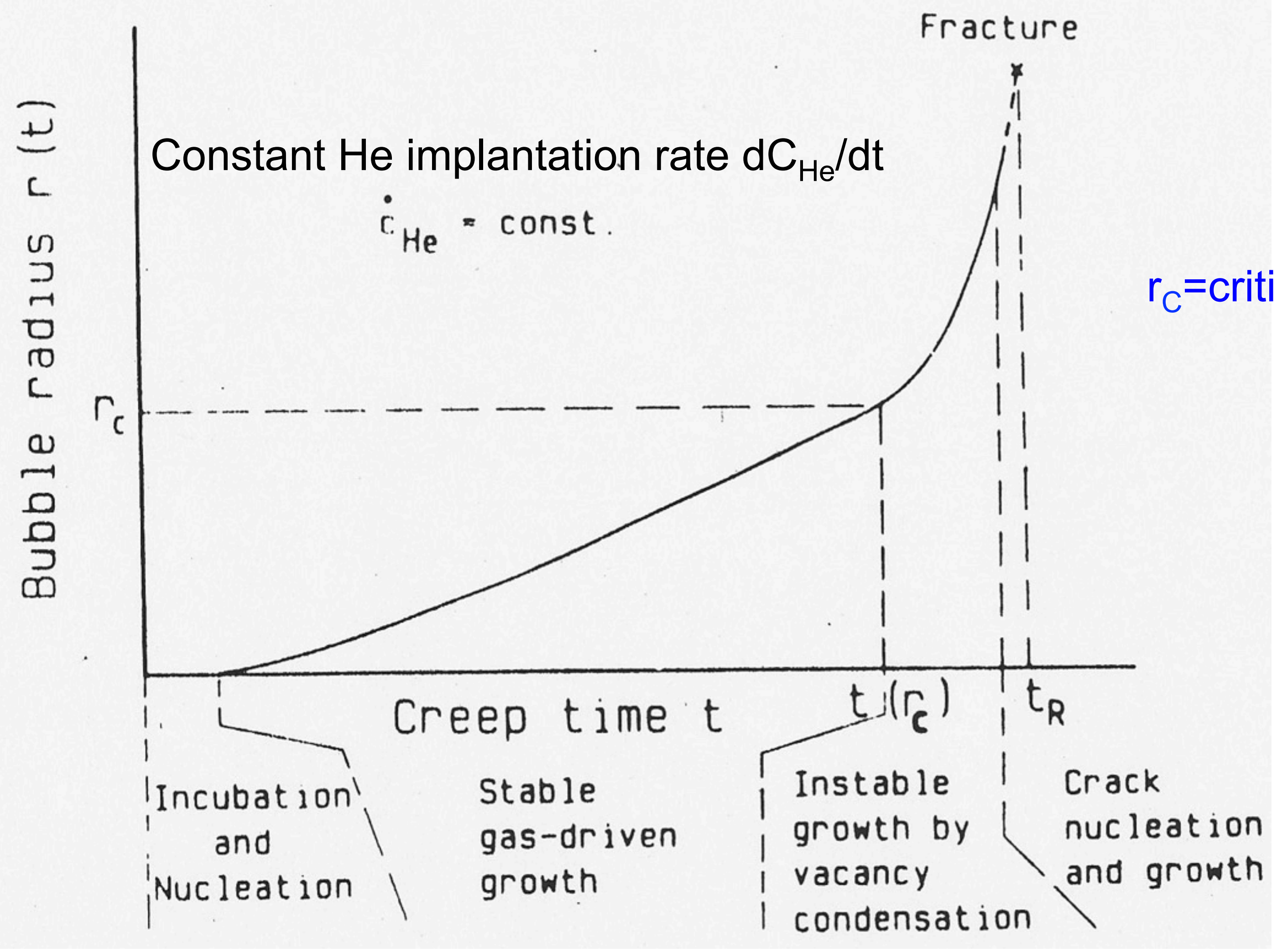
Fig. 4. Total elongation after creep and tensile test in dependence of the secondary creep rate and the tensile strain rate.

Pronounced reduction in ductility observed at 550°C for irradiated 304 SS containing ~0.5-7 appm He at slow strain rates (~10^{-9}/s), but not at typical tensile strain rates (~10^{-4}/s)

B. van der Schaaf, et al. in Radiation Effects in Breeder Reactor Structural Materials, Bleiberg & Bennett, eds. (TMS-AIME, 1977) p.307

G.B. helium cavities grow slowly until they achieve critical size, whereupon growth is more rapid

Helium bubble radius vs. implantation time for in-beam creep tests to fracture



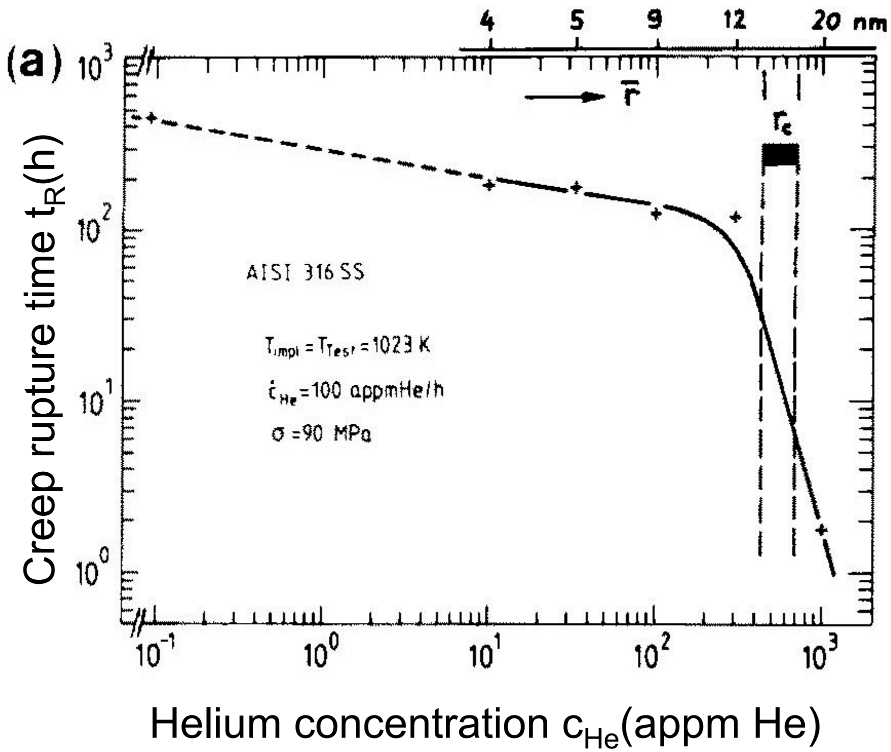
r_C=critical radius

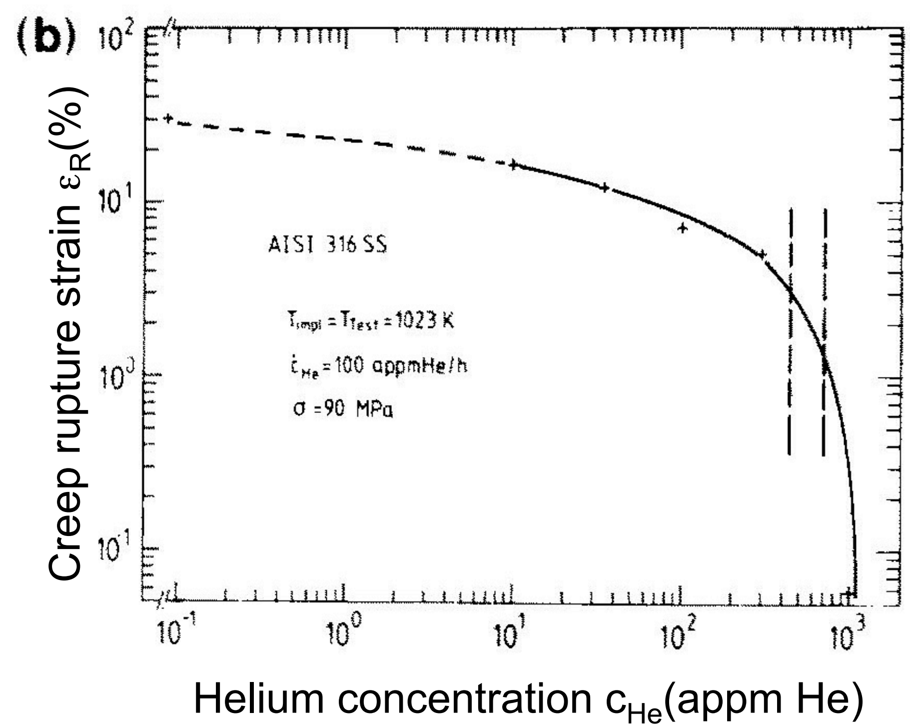
Schroeder & Stamm
ASTM STP 1046 (1989) p. 244

High temperature helium embrittlement

Since high temperature He embrittlement is a diffusion-activated phenomena, the effect becomes more pronounced with increasing:

He concentration
Test temperature
Testing time (i.e., decreasing strain rate)





H. Ullmaier, J. Nucl. Mater. 133&134(1985)100

Qualitative comparison between austenitic and ferritic/ martensitic steel

Austenitic steel generally exhibits high thermal creep strength (high vacancy migration E)
Austenitic steel generally has a lower threshold to convert subcritical gb bubbles to rapidly growing cavities

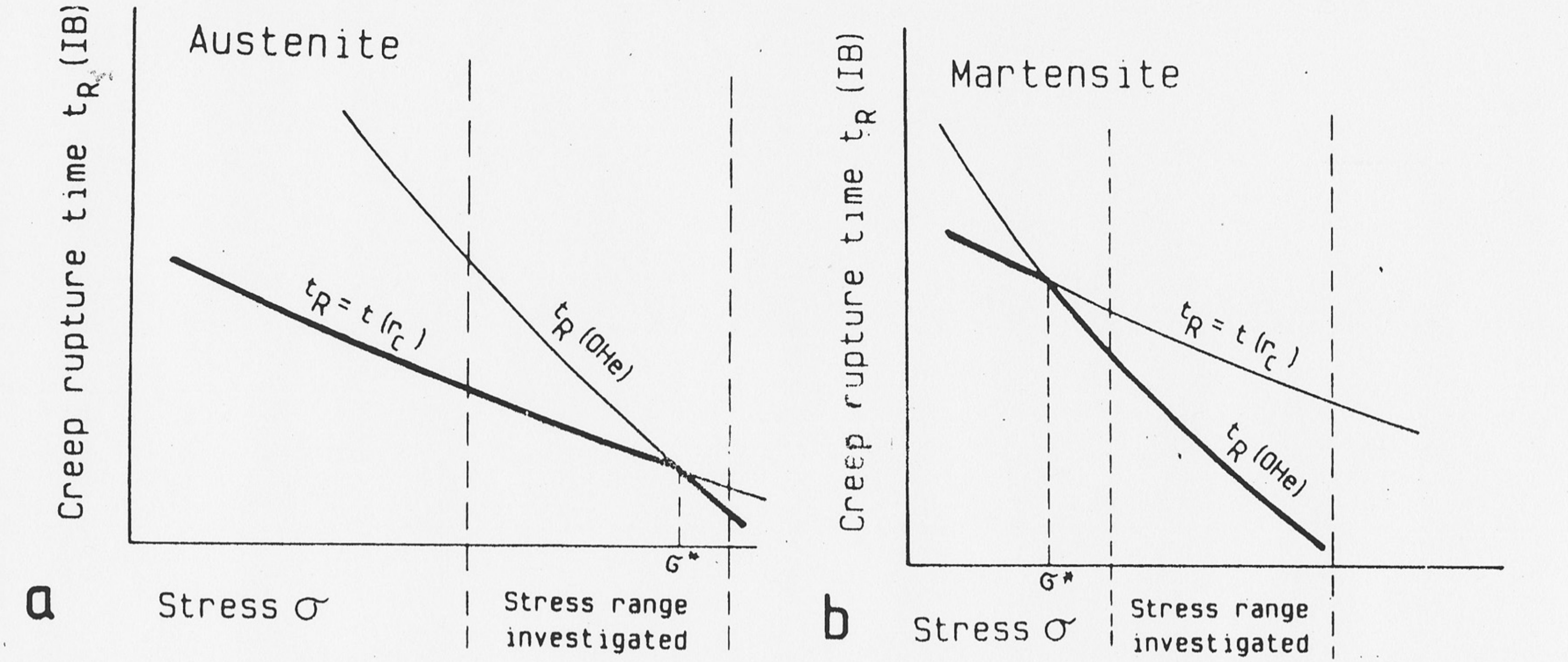
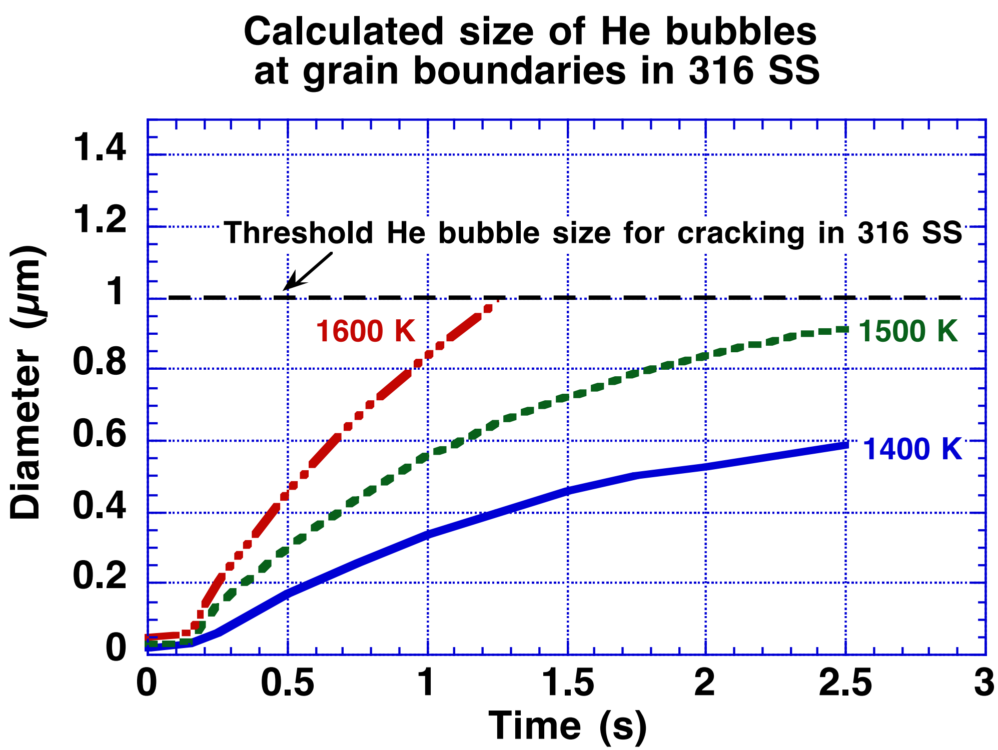


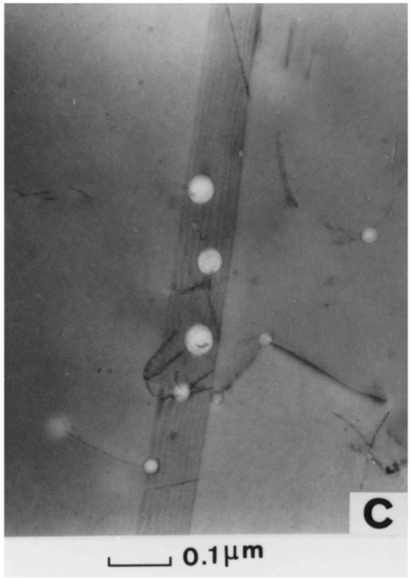
FIG. 7—*Comparison of fracture time* t_R *as a function of applied stress* σ *between unimplanted controls* (t_R(OHe)) *and stable gas-driven growth as a lifetime controlling mechanism* ($t_R = t(r_c)$) *(schematic). Indicated is the stress range investigated and the transition stress* σ*, *at which the fracture mechanism changes. The bold line indicates the minimum fracture time that is measured in an in-beam test.* (a) *Austenite and* (b) *Martensite.*

Schroeder & Stamm
ASTM STP 1046 (1989) p.244

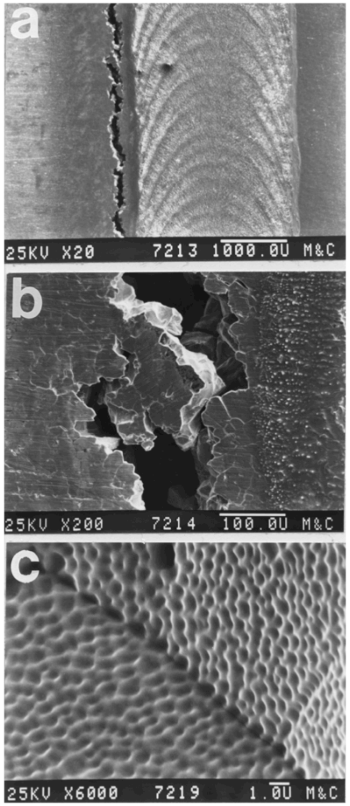
Joining of He-containing metals is problemmatic

- Irradiated materials with He contents above ~1 appm cannot be fusion-welded due to cracking associated with He bubble growth; the lower temperatures associated with a solid-state joining process such as friction stir welding may allow repair joining of irradiated materials





H.T. Lin et al., Metall. Trans. 21A (1990) 2585



Helium in Irradiated Materials

- **Progress**
  - **Swelling-critical number of gas atoms, n_g***
    - **Enables swelling at conditions where critical radius is otherwise too large to attain by fluctuations in point defect fluxes.**
    - **Produces bimodal cavity size distributions. Key to designing experiments where reasons for the differences in swelling for high and low nickel Fe-Ni-Cr ternary were uncovered.**
    - **Led to key principle for designing swelling resistant alloys in the high swelling Fe-Ni-Cr composition range--Introduce high # of precipitates to provide large interfacial area for profuse bubble nucleation, so that no bubble can accumulate n_g*.**

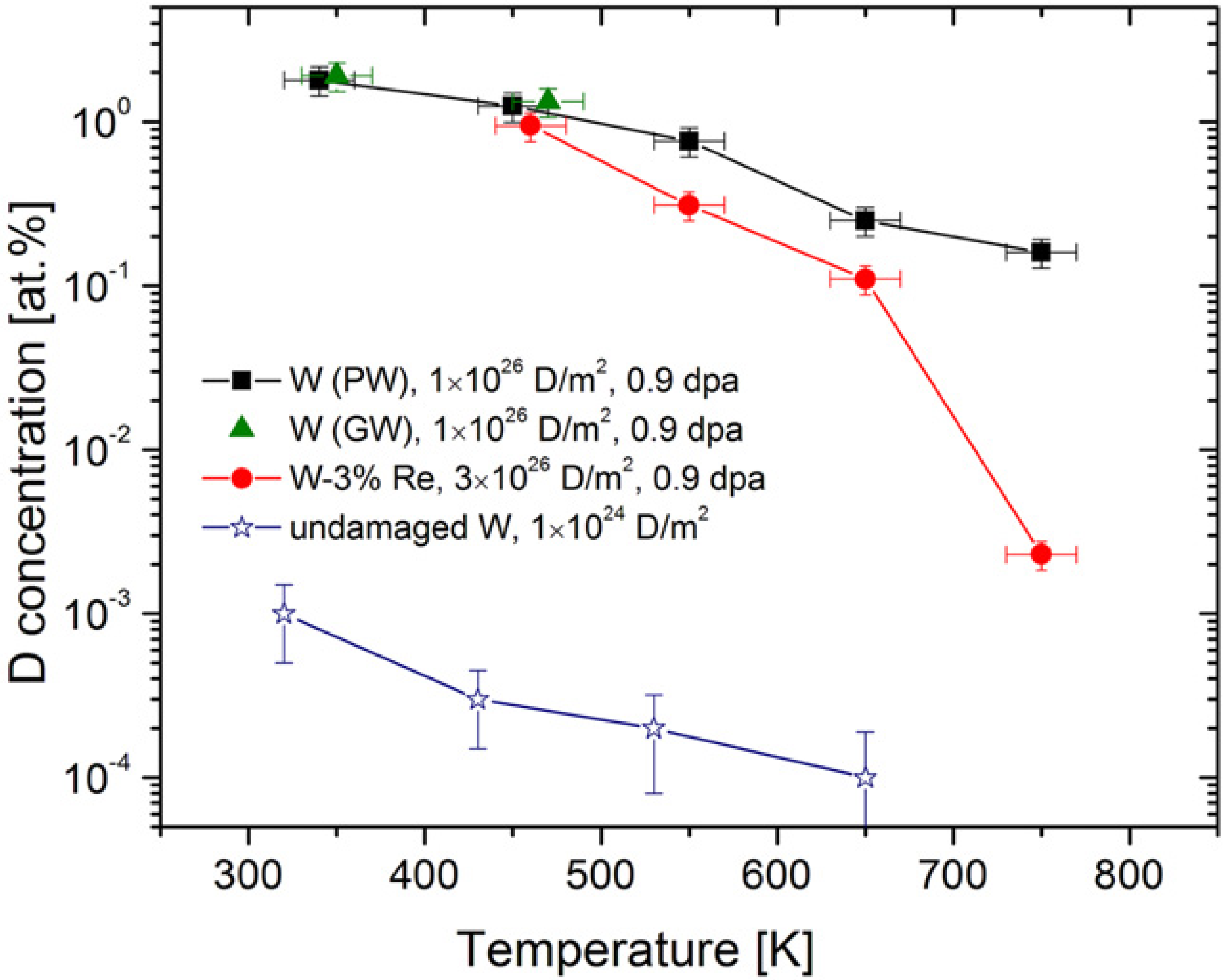
L.K. Mansur, ~2000

Helium in Irradiated Materials

- **Progress**
  - **Grain boundary embrittlement**
    - **Pure helium embrittlement under stress at high temperatures has been understood by similar concepts, cavity critical radius and critical number of gas atoms, as for swelling under irradiation.**
  - **Helium diffusion**
    - **Understood as combination of substitutional and dissociative mechanisms. In the former there is a large population diffusing slowly and in the latter there is a very small population diffusing very quickly. Three main mechanisms briefly convert He from substitutional to interstitial status: thermal release, interstitial replacement and direct displacement. Their relative importance varies strongly with T and dose rate.**

L.K. Mansur, ~2000

Irradiation damage can significantly enhance the retention of H isotopes in metals



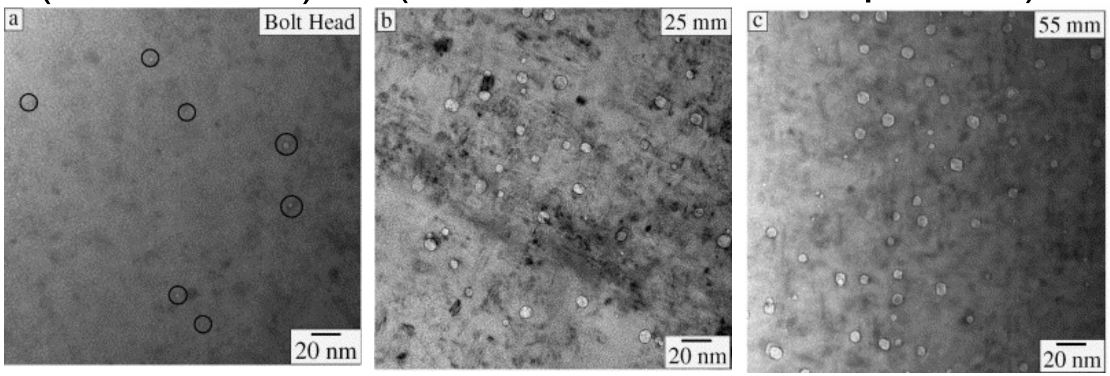
B. Tyburska-Püschel & V. Alimov, Nucl. Fusion 53 (2013) 123021

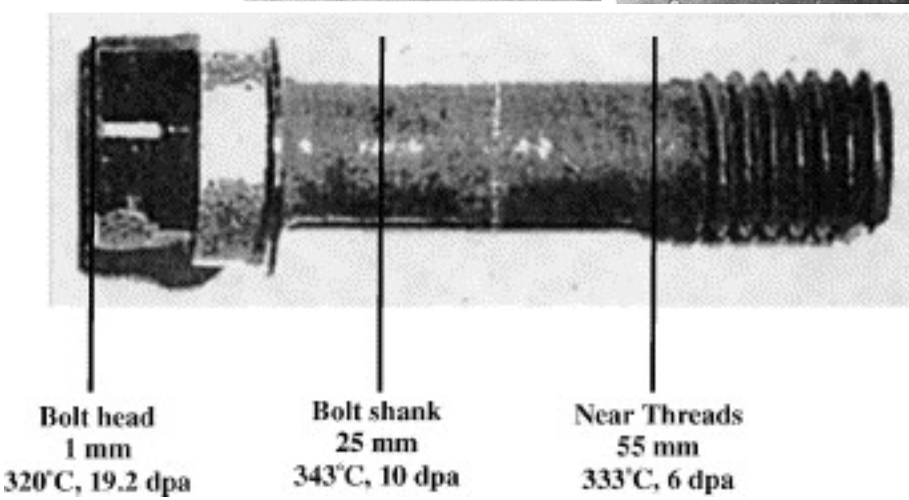
H retention increases dramatically in the presence of cavity formation

3 to 5x increase in retained hydrogen when cavities are present, even with 2-3x reduction in neutron fluence exposure

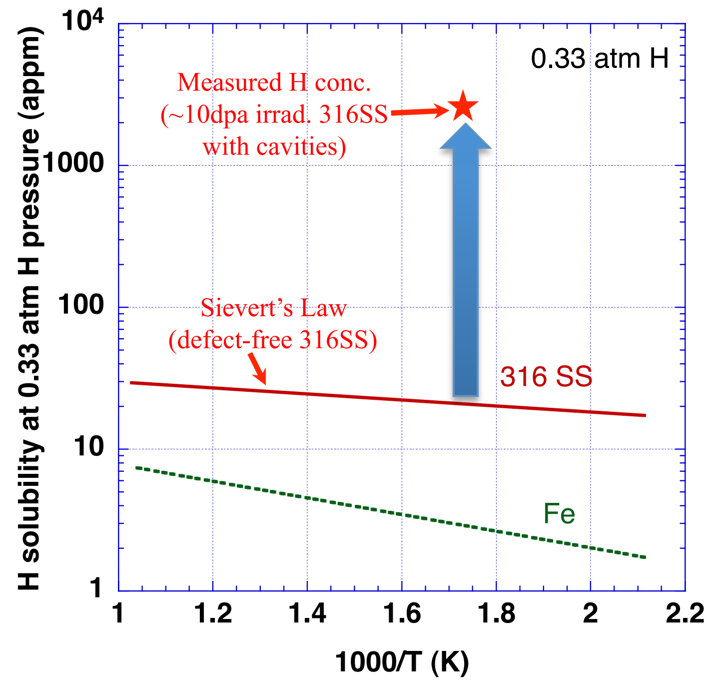
500-700 appm H (few cavities)

1700-3700 appm H (rad.-induced cavities present)





Retained H level is ~100x higher than expected from Sievert's law solubilities



Baffle-former bolt removed from Tihange-1 (Belgium) pressurized water reactor

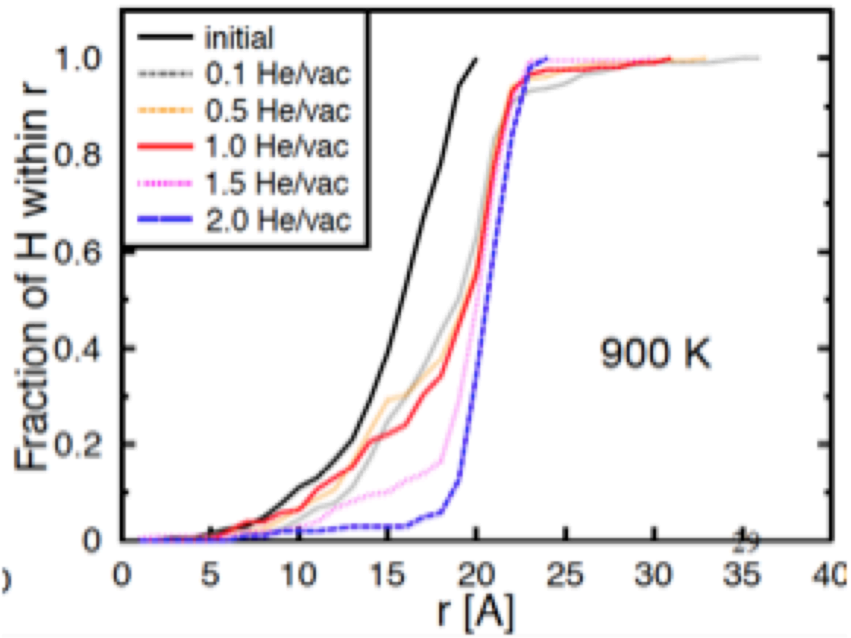
Type 316 austenitic stainless steel

F.A. Garner et al., J. Nucl. Mater. 356 (2006) 122

H trapping in neutron irradiated tungsten

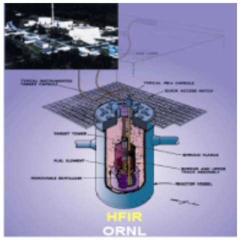
- Hot wall operation introduces several new phenomena
  - enhanced D/T retention after neutron irradiation (due to trapping at defect complexes)

Modeling

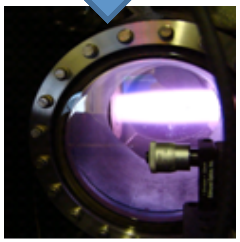


Calculated fraction of hydrogen that is trapped in the vicinity of a 2 nm radius He bubble in tungsten at 900 K (B.D. Wirth).

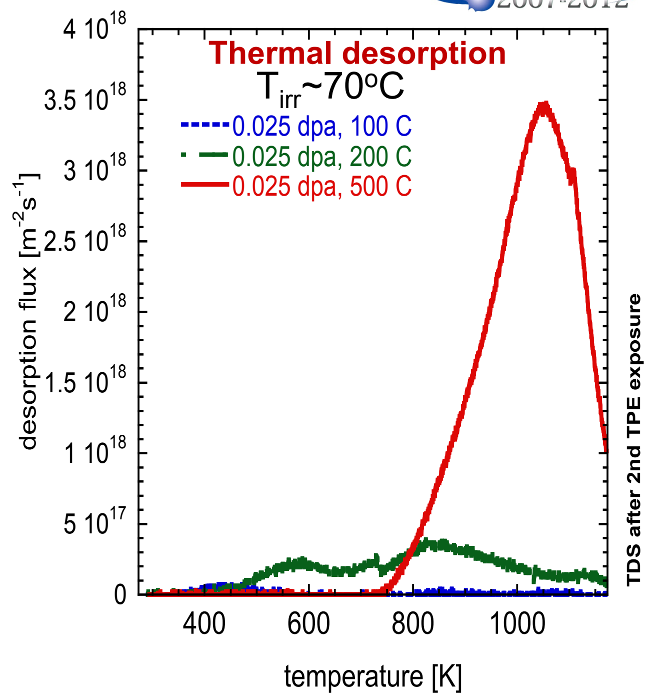
Experiment



Neutron irradiation in HFIR



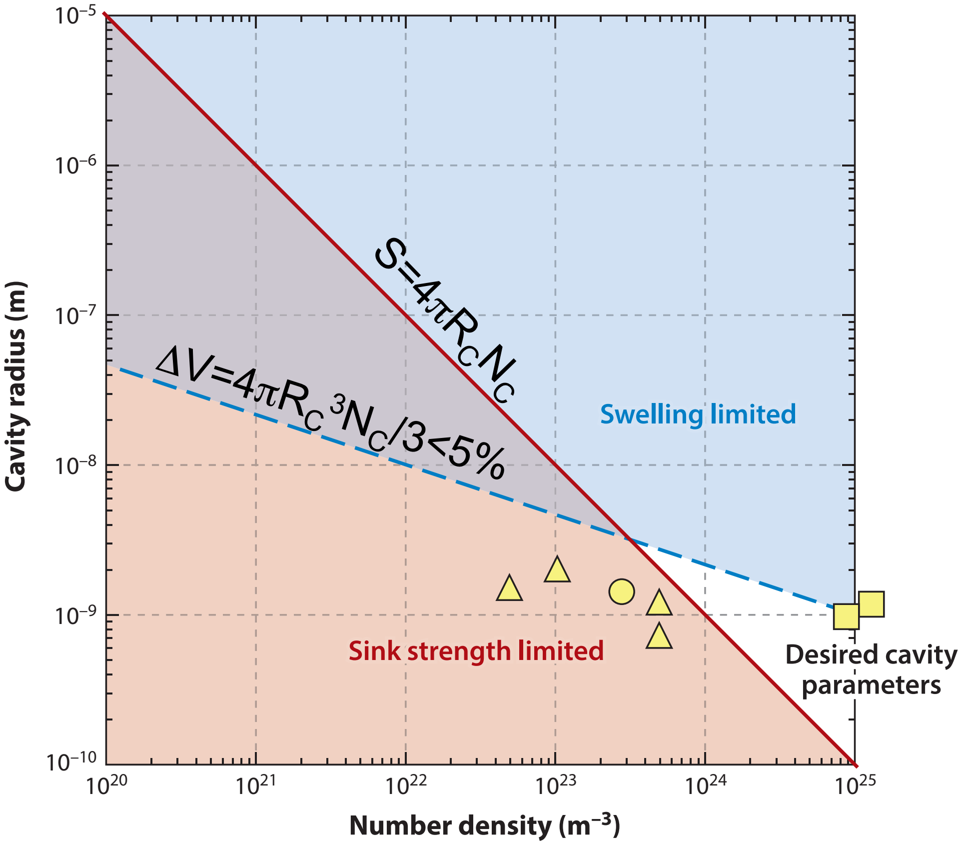
D plasma exposure in TPE

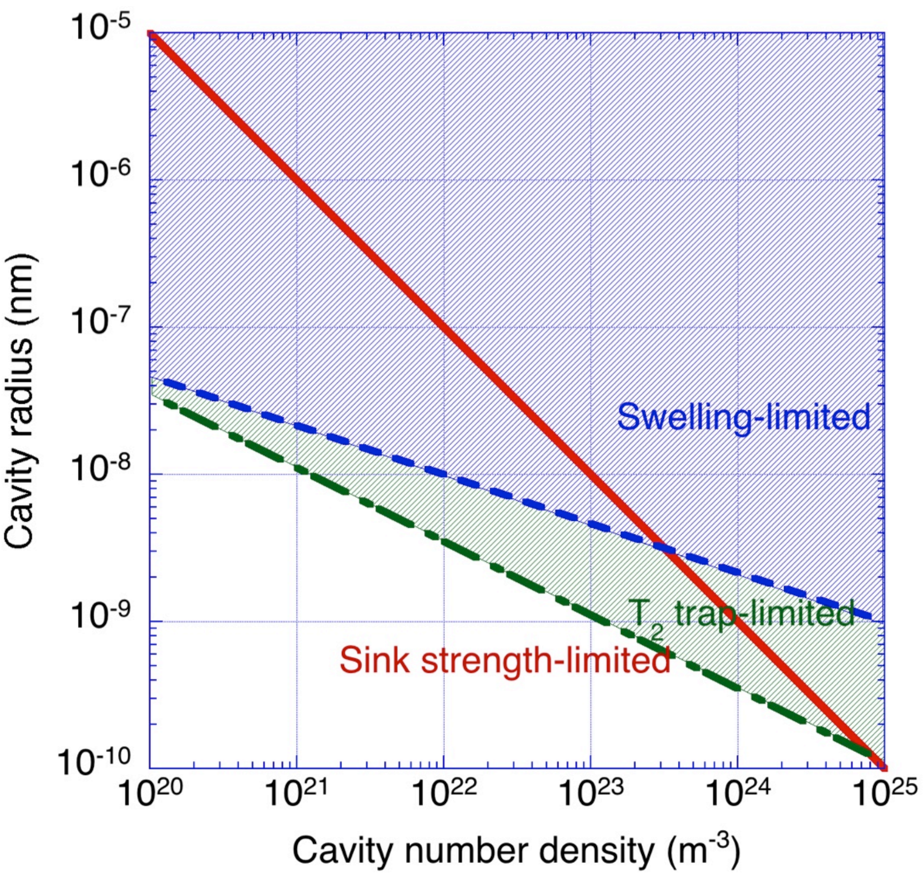


Hatano et al. FTP 4-1 IAEA Fusion Energy Conf San Diego 2012

Desorption experiments on W neutron-irradiated at high temperature are needed

Does the mainstream approach for designing radiation resistance cause unacceptable tritium sequestration in DT fusion energy structures?





Zinkle and Snead, Ann. Rev. Mater. Res. 44 (2014) 241

Conclusions

- **Low temperature phenomena: Hardening and embrittlement**
  - Major effects observed in ferritic steels for C_{He}>500 appm
- **Medium temperature phenomena: Cavity swelling**
  - Major effects observed in austenitic steels for C_{He}>100 appm; ferritic steels for C_{He}>500 appm?
- **High temperature phenomena: High temperature He embrittlement of grain boundaries**
  - Major effects observed in austenitic steels for C_{He}>1-100 appm; ferritic steels C_{He}>500 appm?
- **Influence of H is less pronounced (per atom) than He**
  - H microstructural influence is mainly via chemical/chemisorption effects, vs. insoluble cavity precipitation (vacancy trapping) for He
  - H trapping in cavities at intermediate temperatures can be an important safety issue for DT fusion energy systems