Consequences of Irradiation on Behavior of Structural Materials

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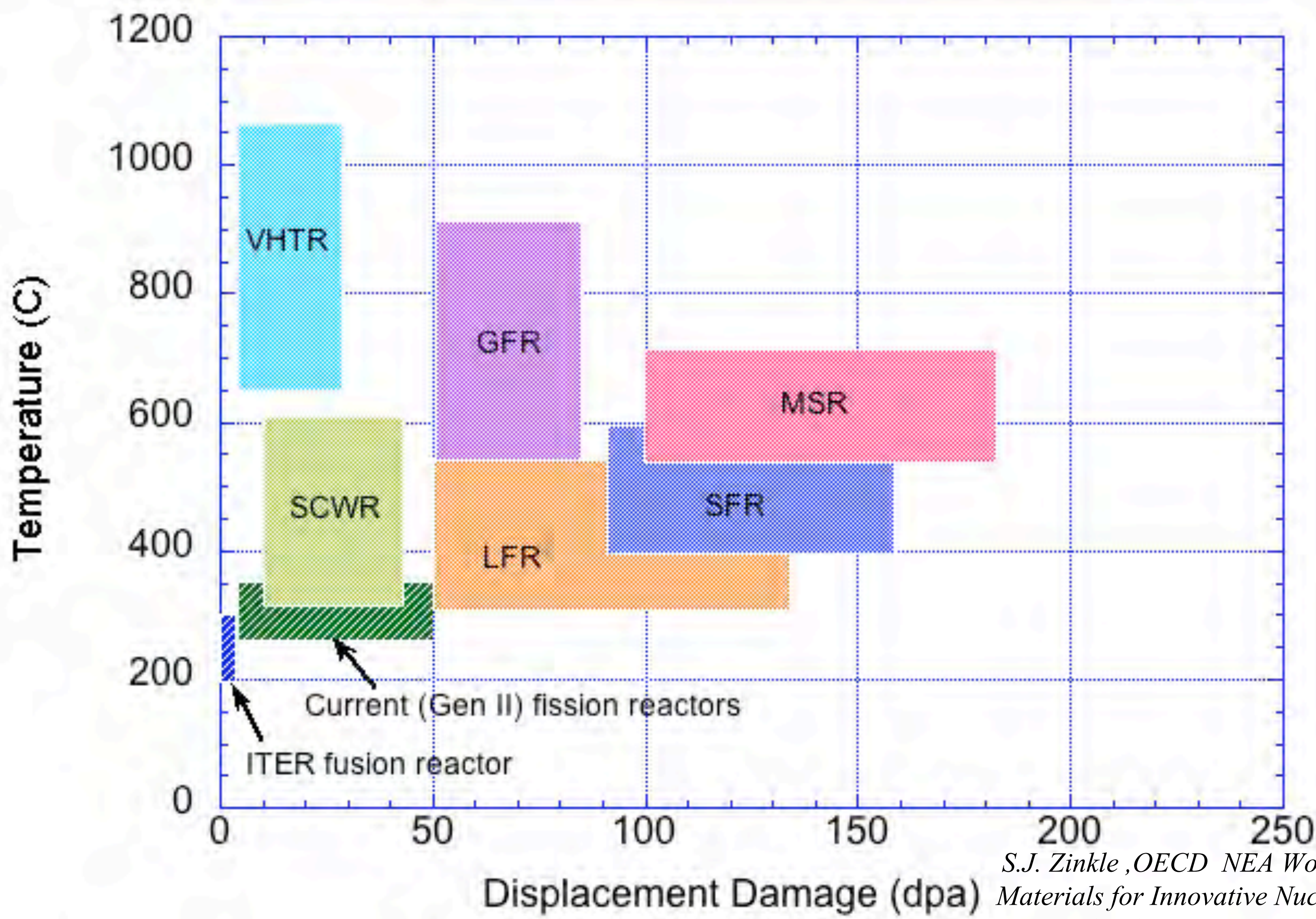
Outline

- Effect of low temperature (<0.3 T_M) irradiation on the mechanical properties of austenitic and ferritic/martensitic steels
  - Dose and temperature dependence
- Deformation mechanism maps for irradiated materials
- Overview of irradiation creep
- High temperature He embrittlement of grain boundaries
- Prospects for developing new high-performance structural materials for Gen IV reactor applications

Overview of Gen IV and current (Gen II+) Structural Materials Environments



S.J. Zinkle ,OECD NEA Workshop on Structural Materials for Innovative Nuclear Energy Systems, Karlsruhe, Germany, June 2007, in press

All Gen IV concepts introduce substantial increases in dose and temperature

Room Temperature Radiation Hardening in 9Cr FM

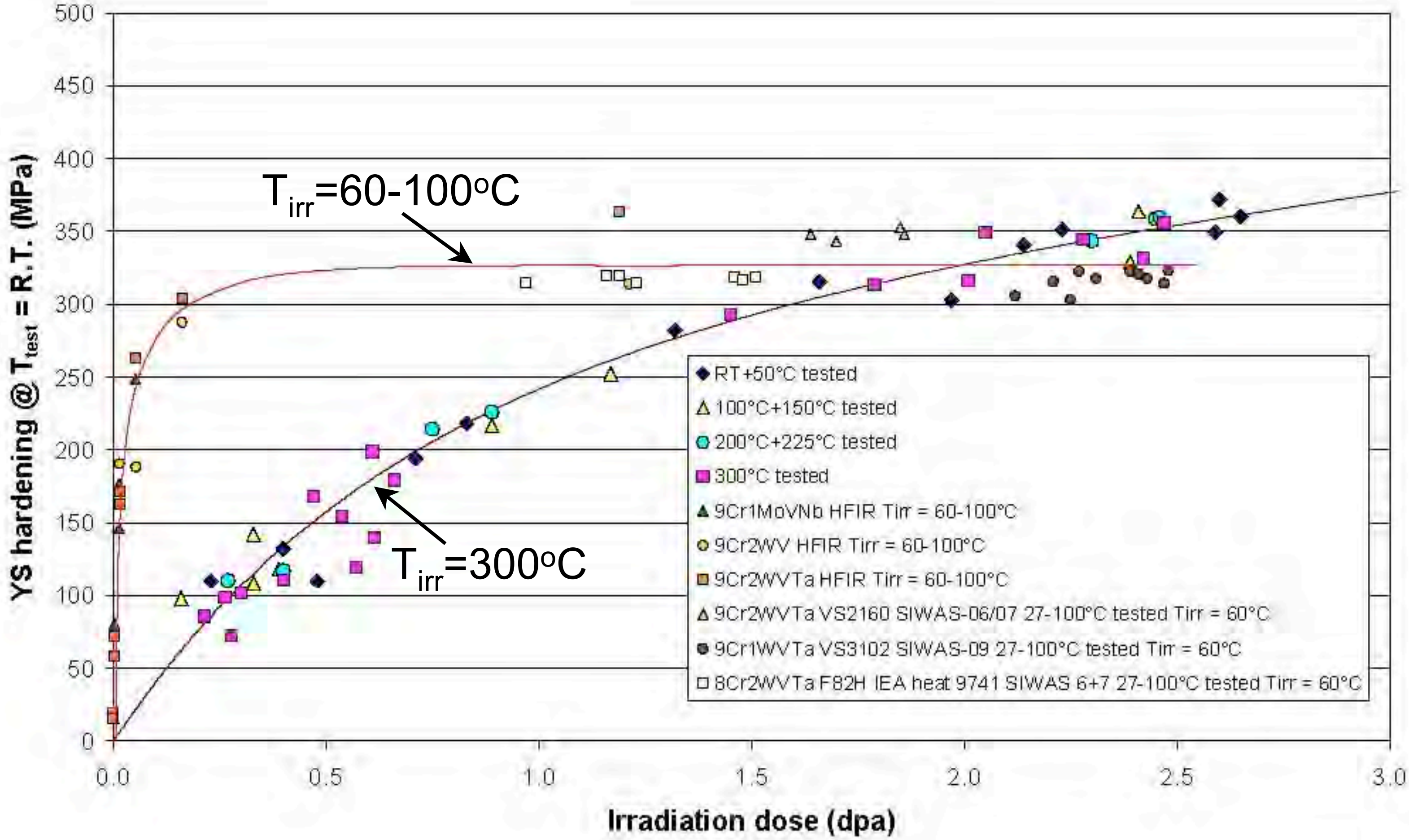


figure 86 Yield stress hardening at 60-80 °C (est.) and 300 °C irradiation temperatures at RT test temperature. NRG data from current report and previous NRG report [4]. HFIR data from [26]

J. Rensman, 2005

Room Temperature Radiation Hardening in 9Cr FM Steels

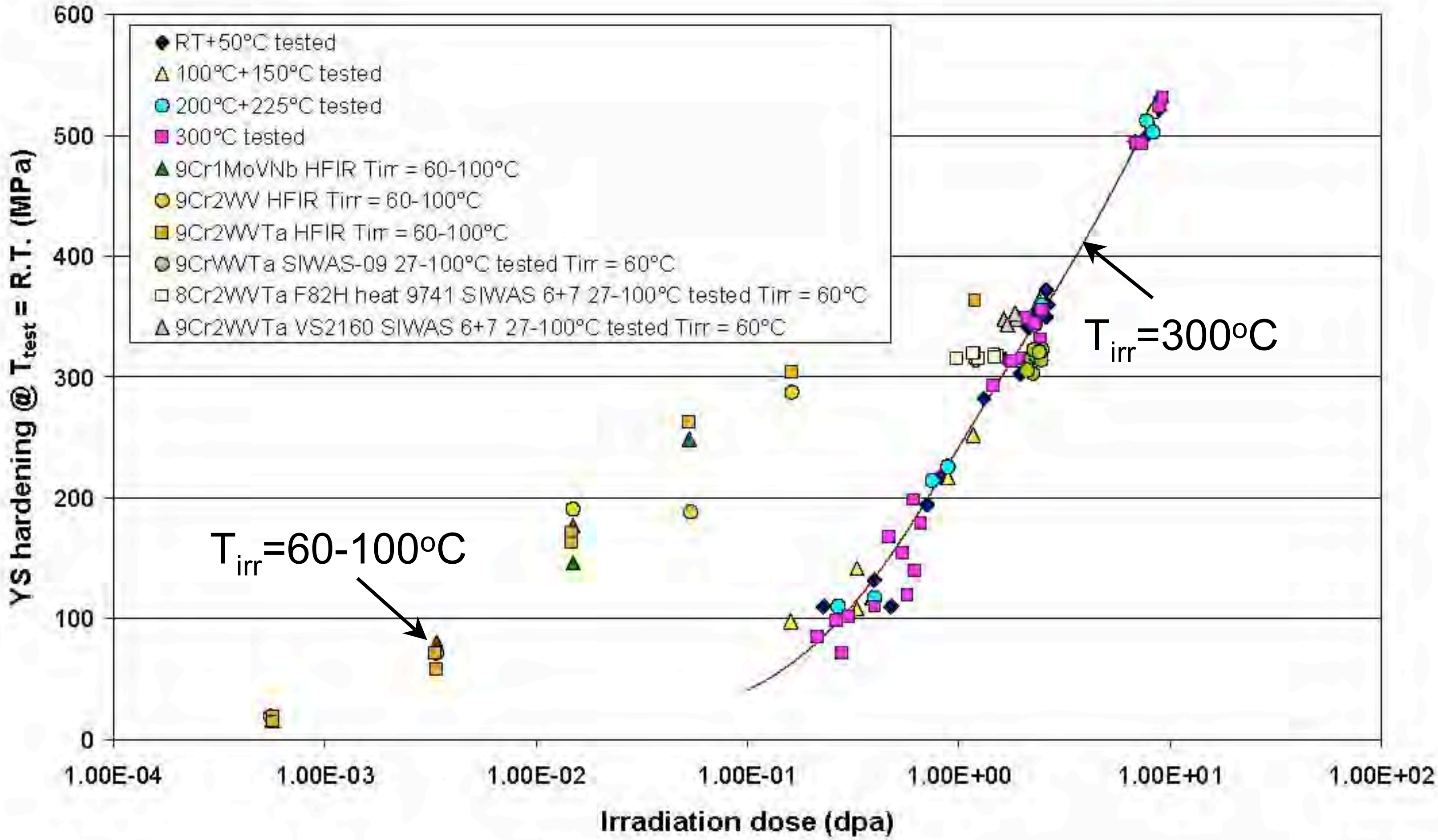
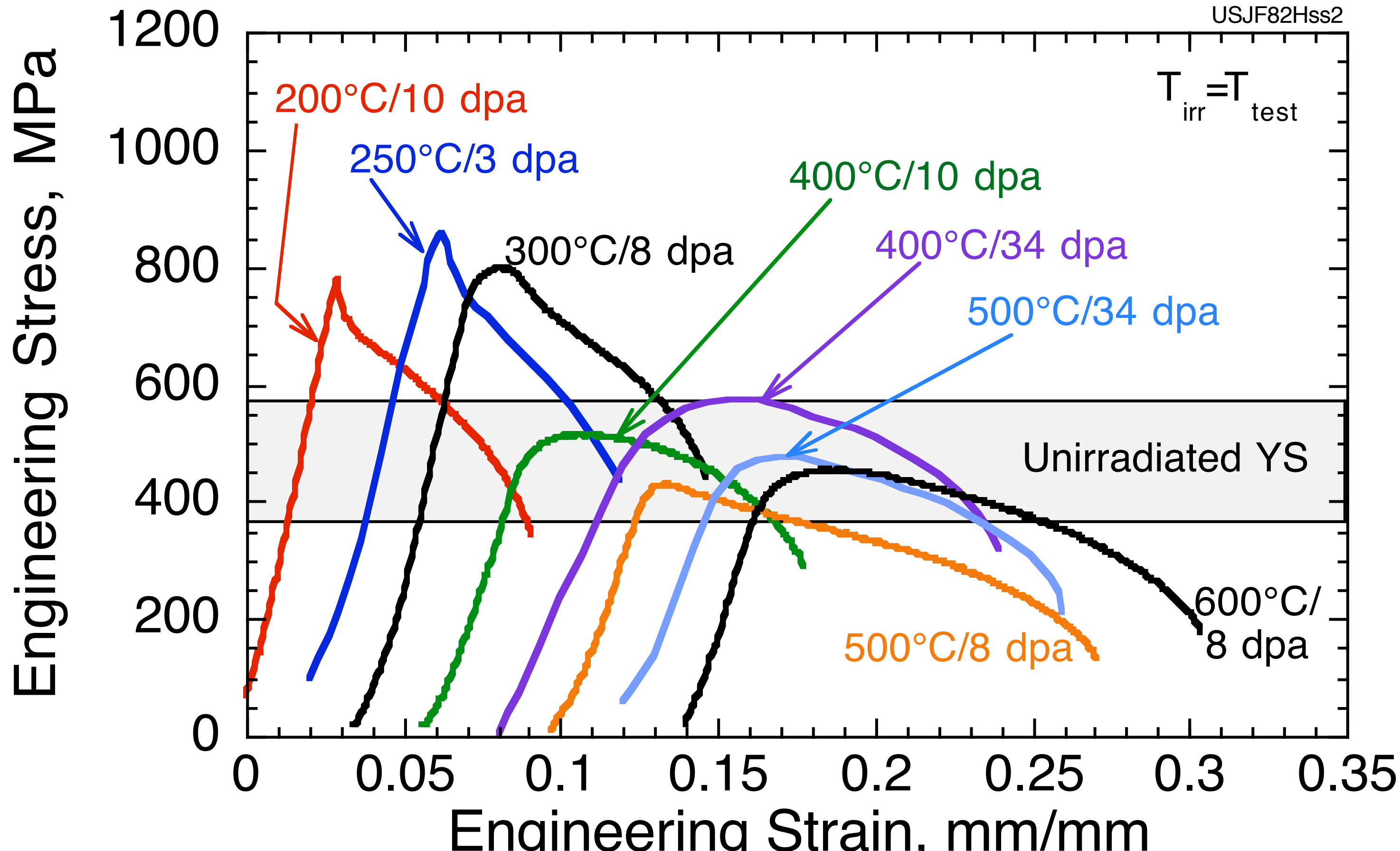


figure 70 Low temperature irradiation hardening of various 9Cr steels vs. dose at a log scale. NRG data from current report and [4], HFIR data from [26]

J. Rensman, 2005

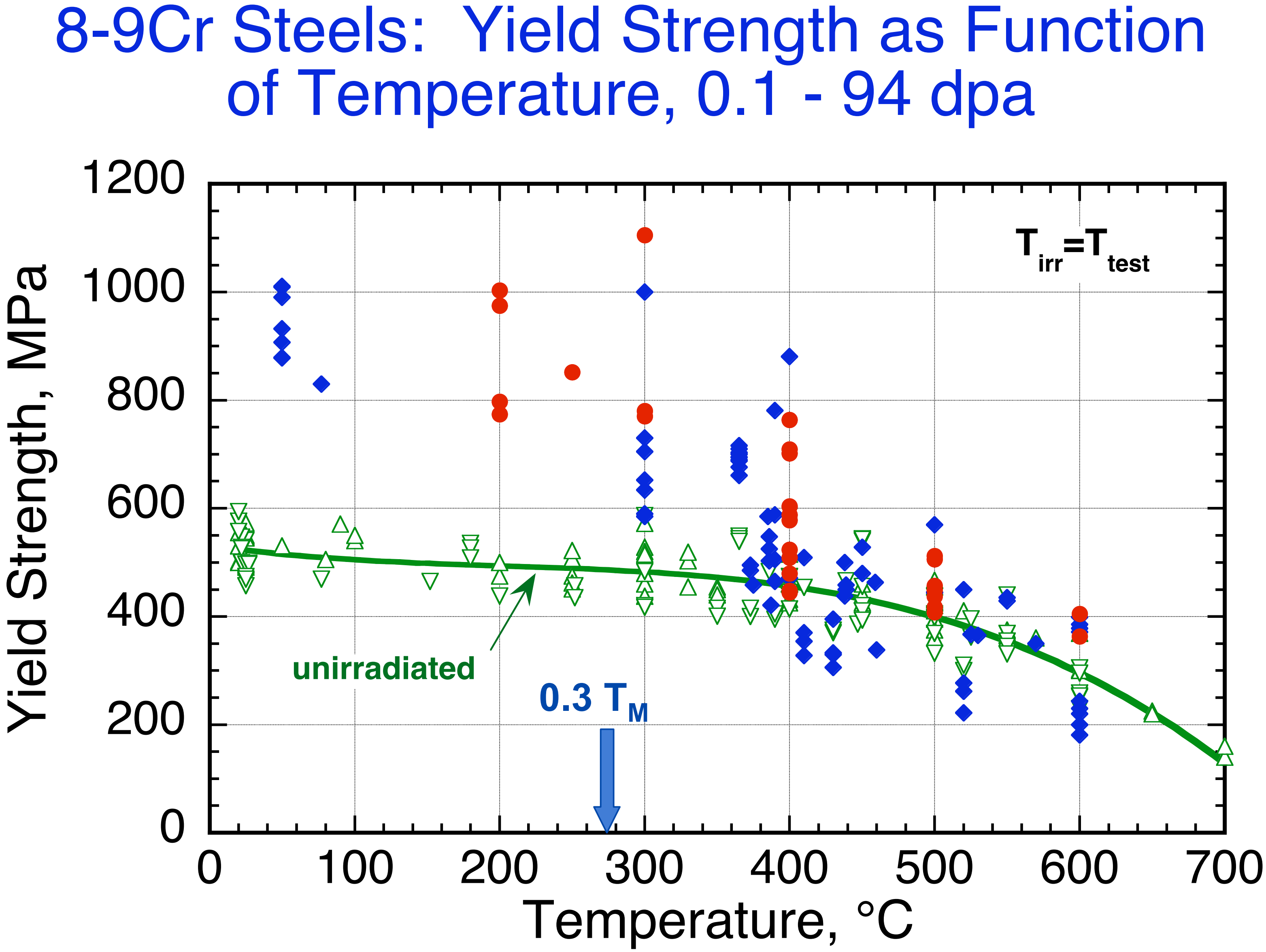
Low Temperature Radiation Hardening is Important in Ferritic/martensitic steel up to ~400˚C

Representative USDOE/JAERI F82H Data: 200-600°C, 3-34 dpa

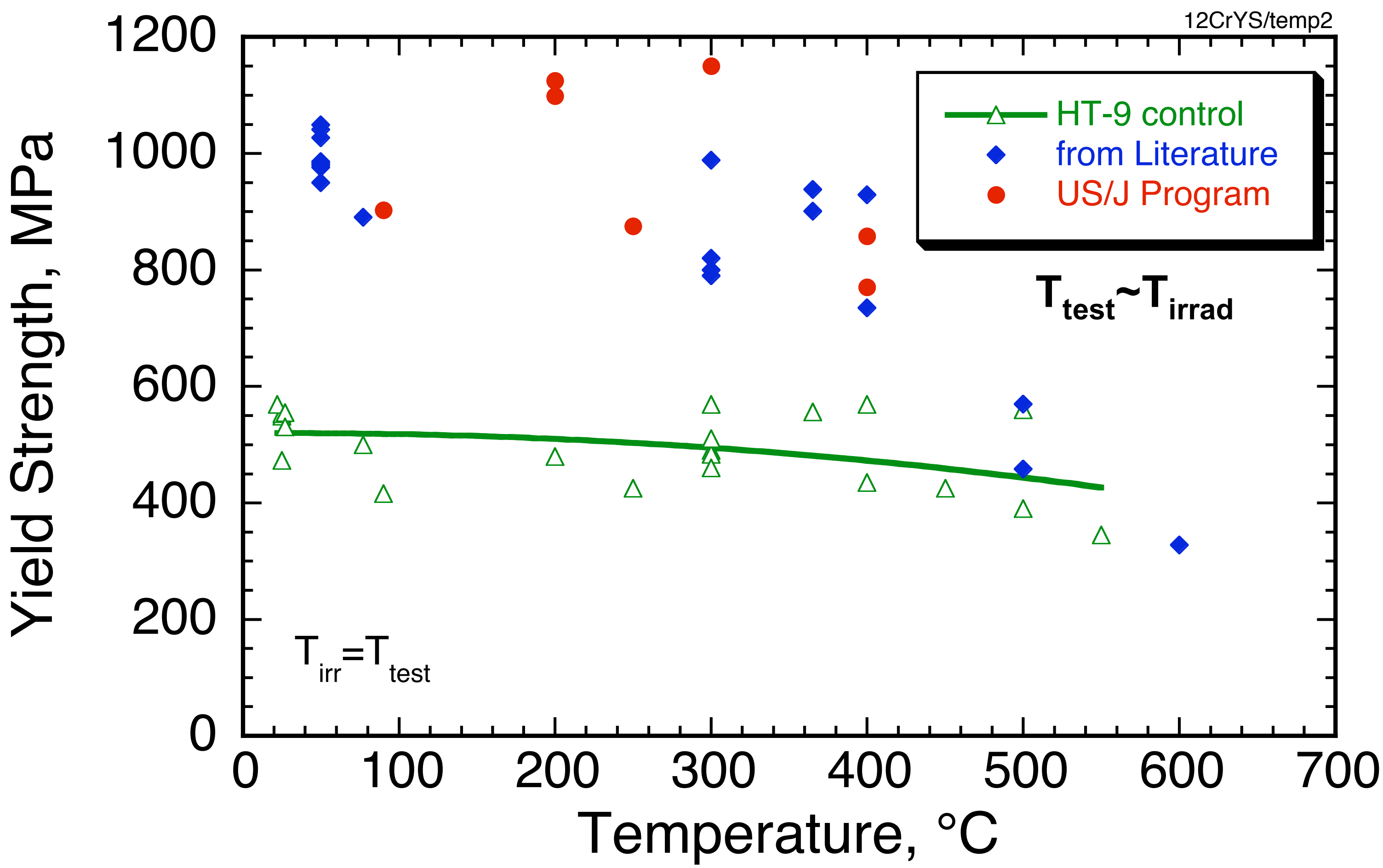


J.P. Robertson et al.,
(DOE/JAERI collaboration)

Radiation hardening in Fe-(8-9%Cr) steels

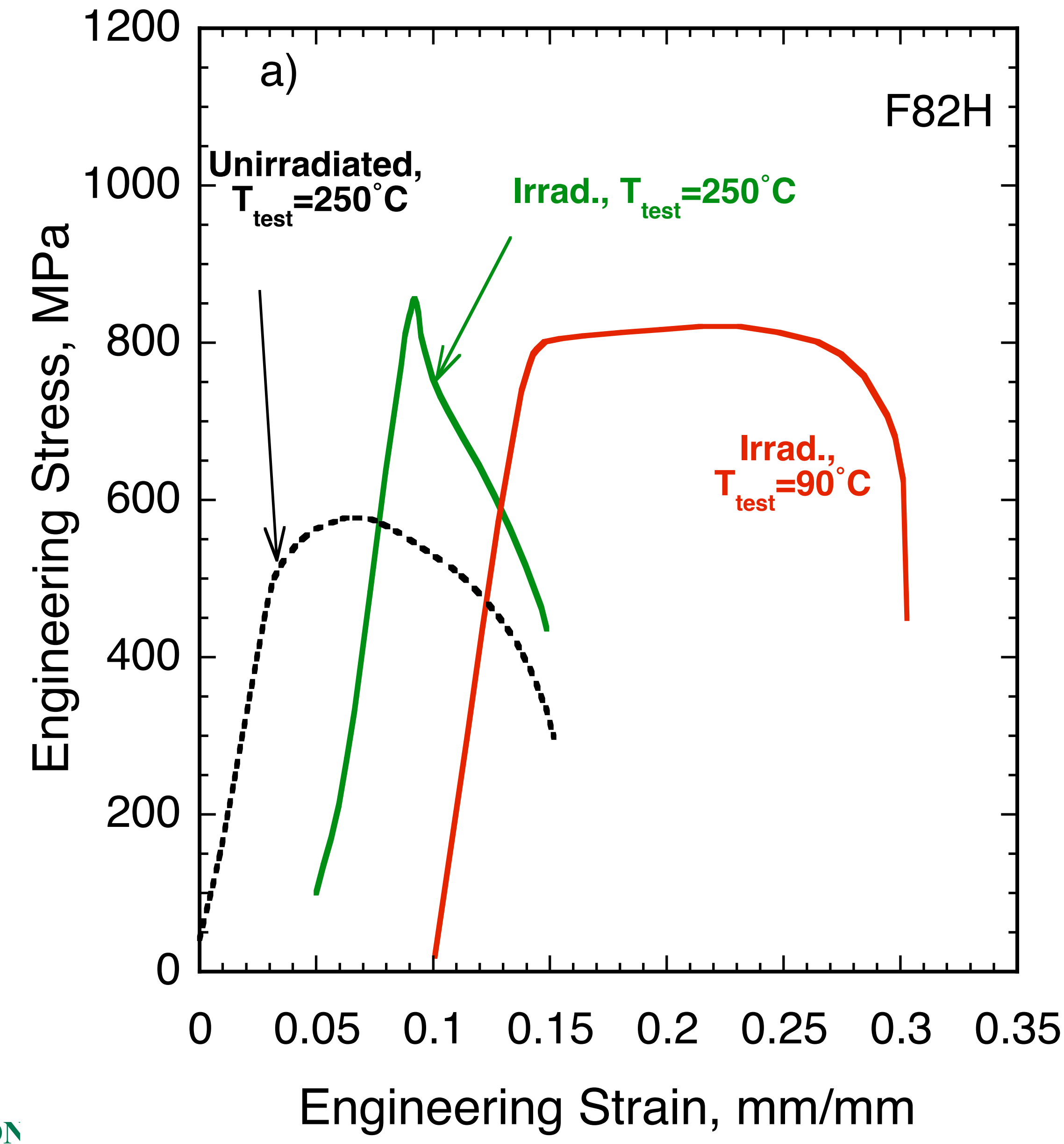


HT-9: Yield Strength as Function of Temperature



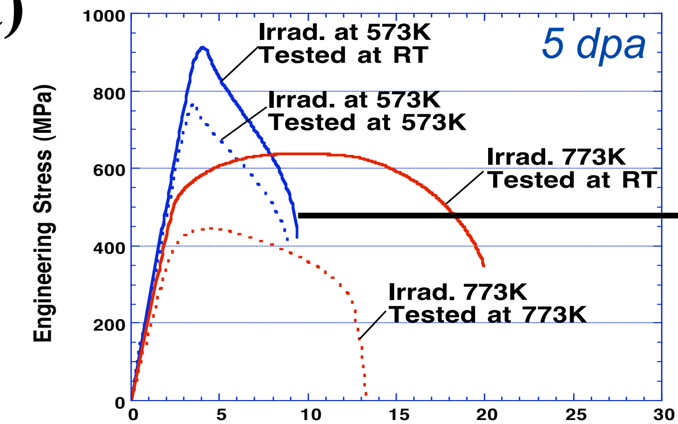
Radiation embrittlement is of concern in HT-9 for irradiation temperatures up to ~400°C

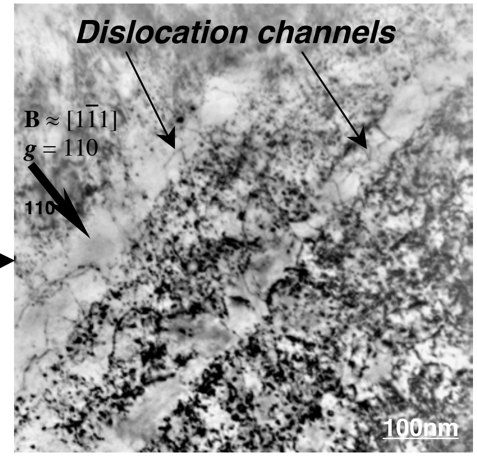
Radiation hardening in Fe-(8-9%Cr) steels after 3 dpa

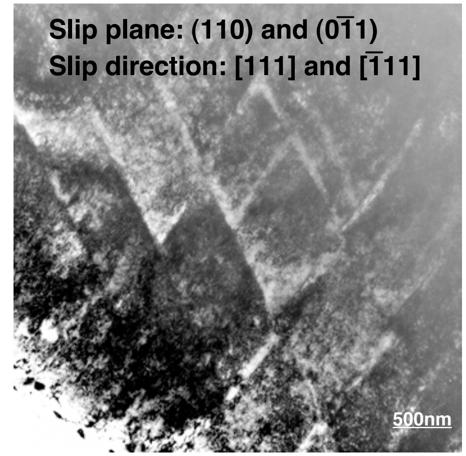


Deformation microstructures in neutron-irradiated Fe-8Cr-2WVTa ferritic/martensitic steel (F82H)

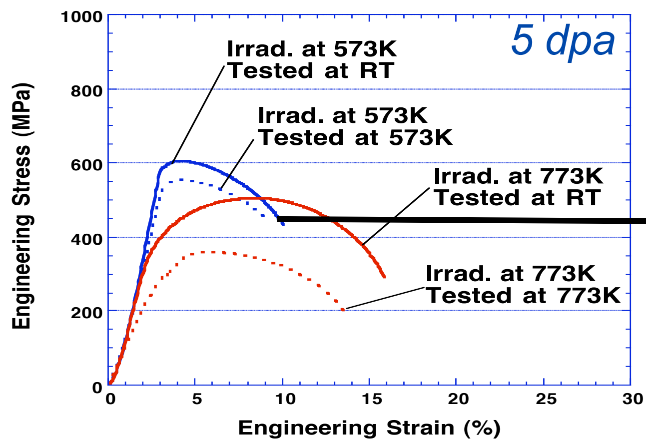
(a)

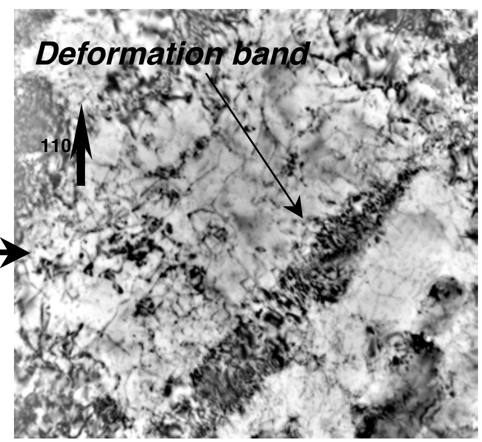






(b)





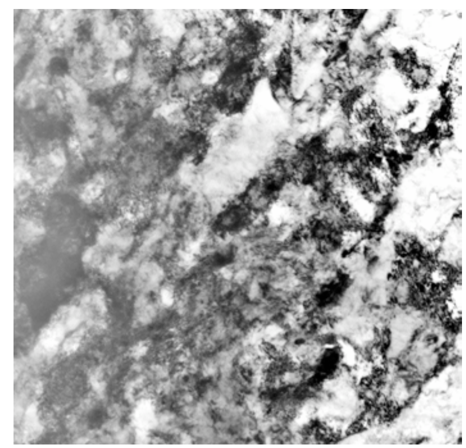
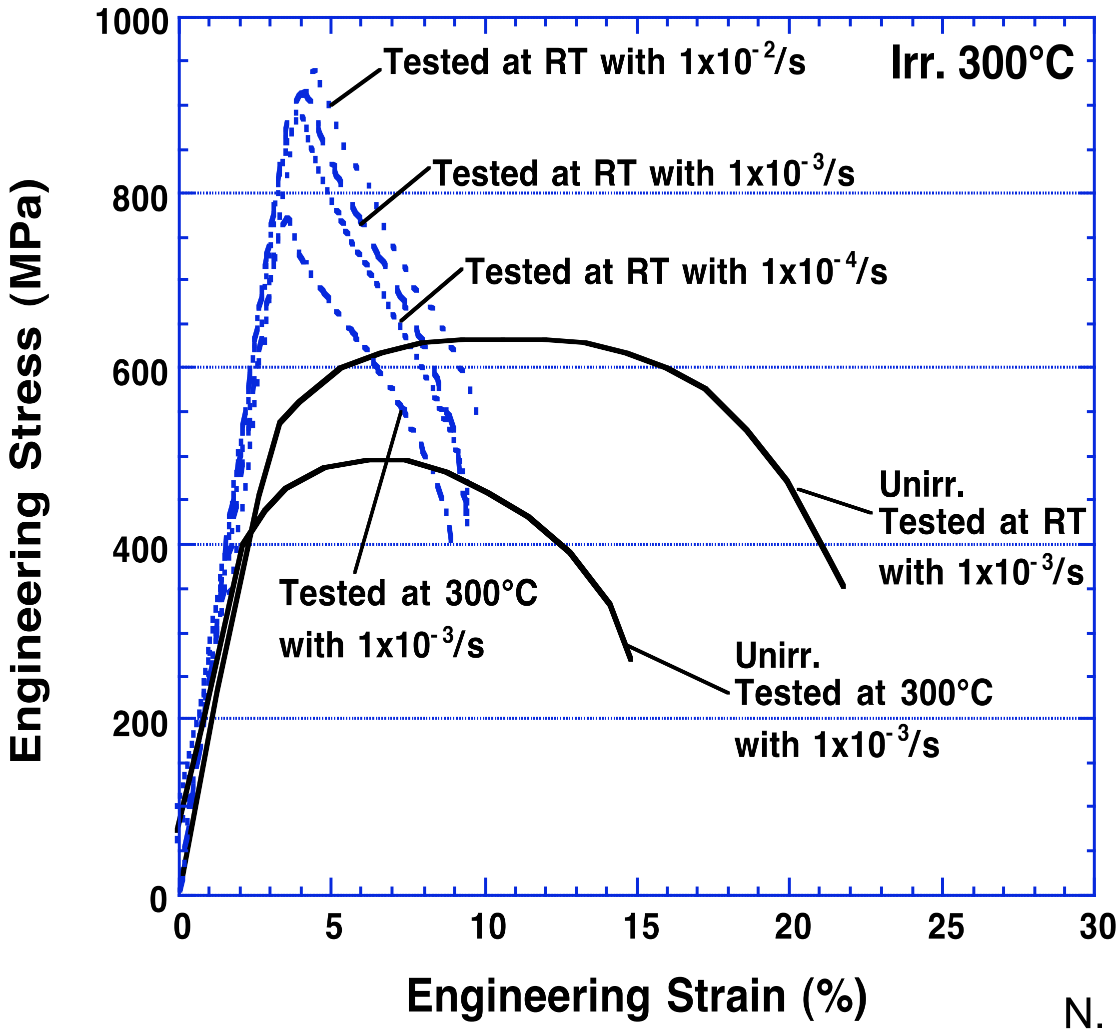


Fig. 1 Stress-strain curves of F82H BM (a) and TIG (b) irradiated at 573K and 773K in tests at RT

Irradiated weld metal (lower radiation hardening) did not exhibit dislocation channeling after deformation

N. Hashimoto et al., Fus.Sci.Tech. 44 (2003)

Tensile curves for F82H ferritic/martensitic steel irradiated at 300˚C to 5 dpa



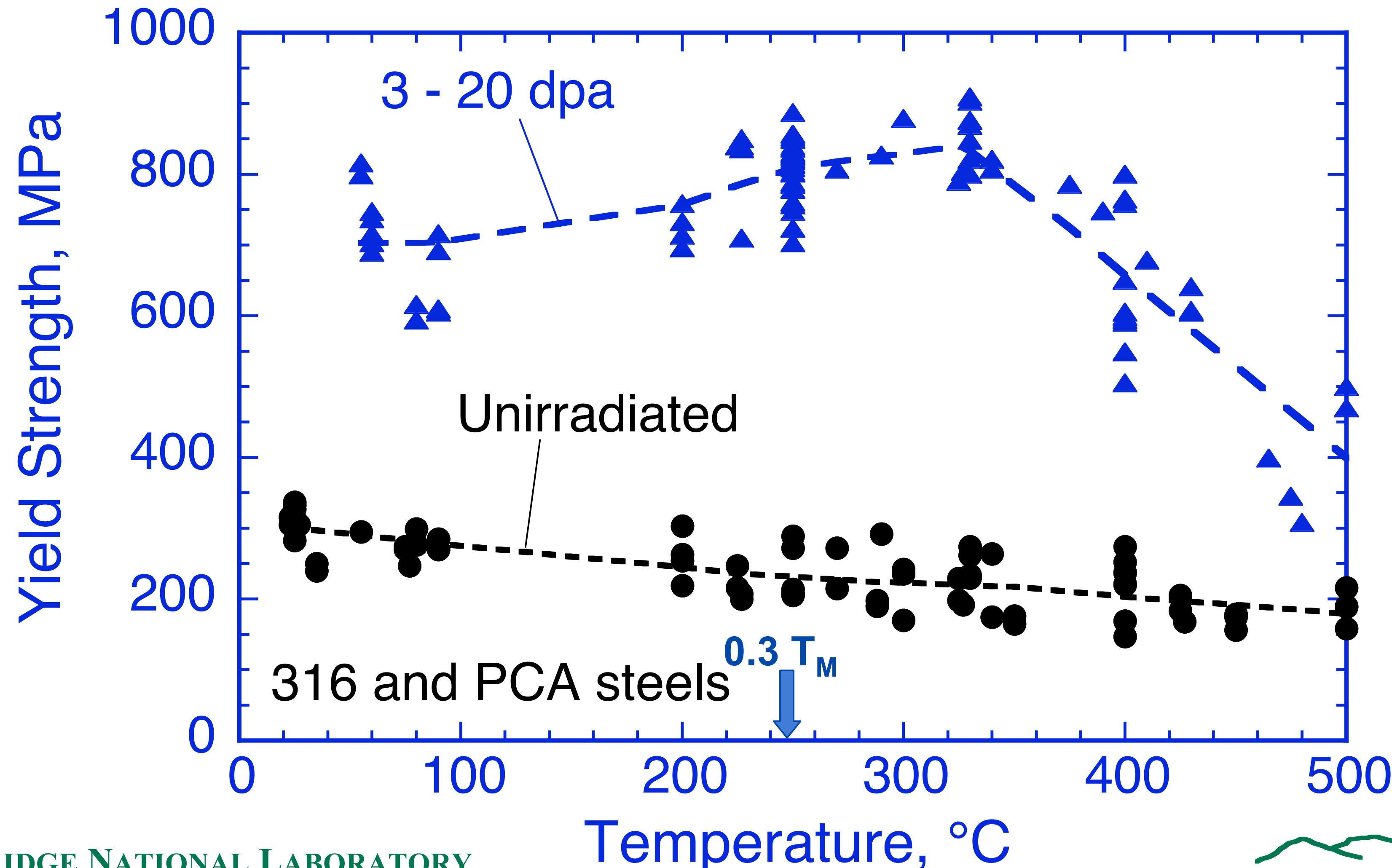
$$m = (1/\sigma)(\partial \sigma / \partial \ln\varepsilon)$$

Strain hardening exponent @RT:
m = 0.041 (unirradiated)
m = 0.028 (irrad. 300˚C)

N. Hashimoto et al., MRS vol. 650 (2001)

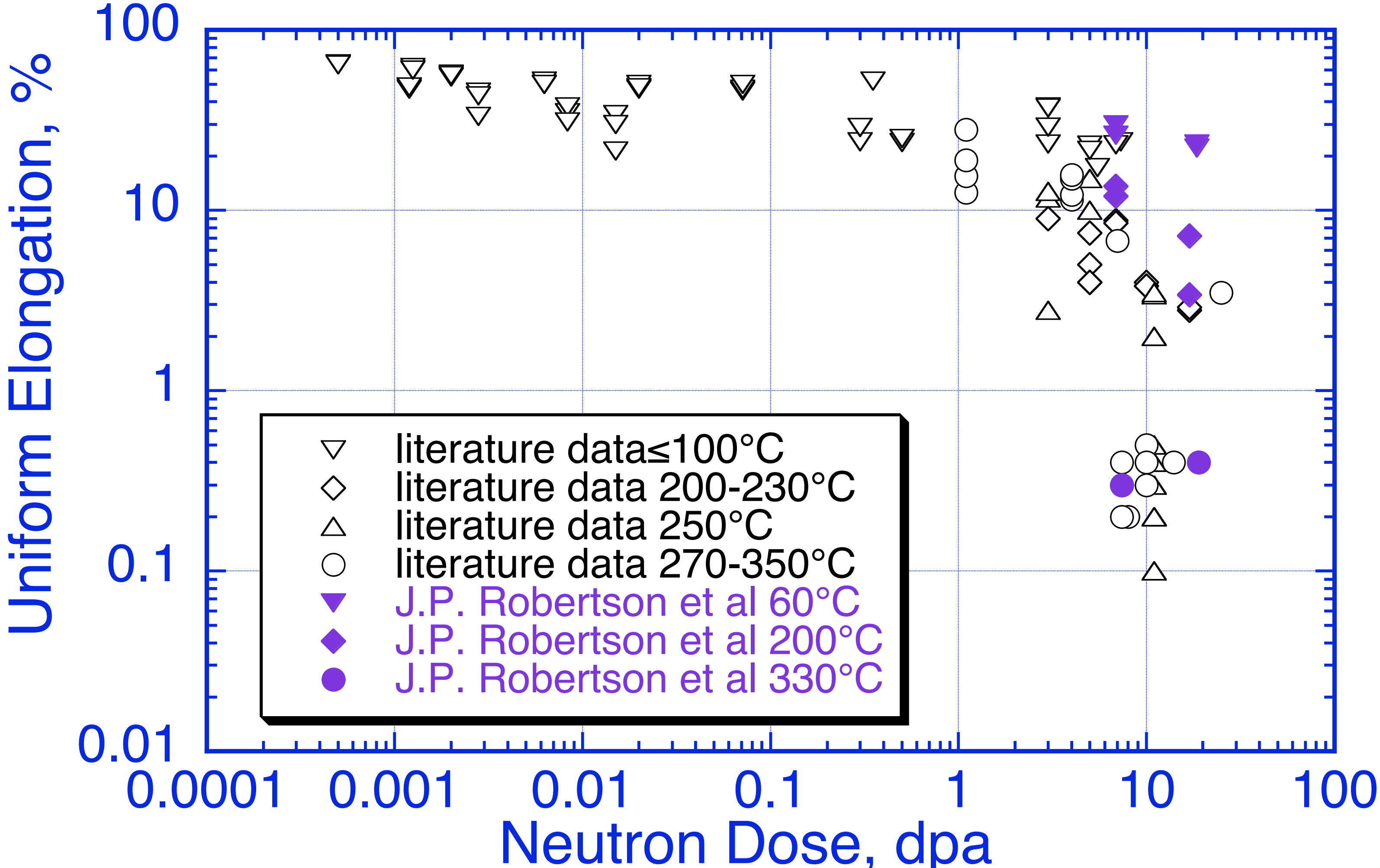
Irradiation of Austenitic Stainless Steel in Mixed Spectrum Reactors produces significant hardening up to 500˚C, with peak hardening at ~300˚C

Hardening at 290-400˚C is dominated by He bubbles

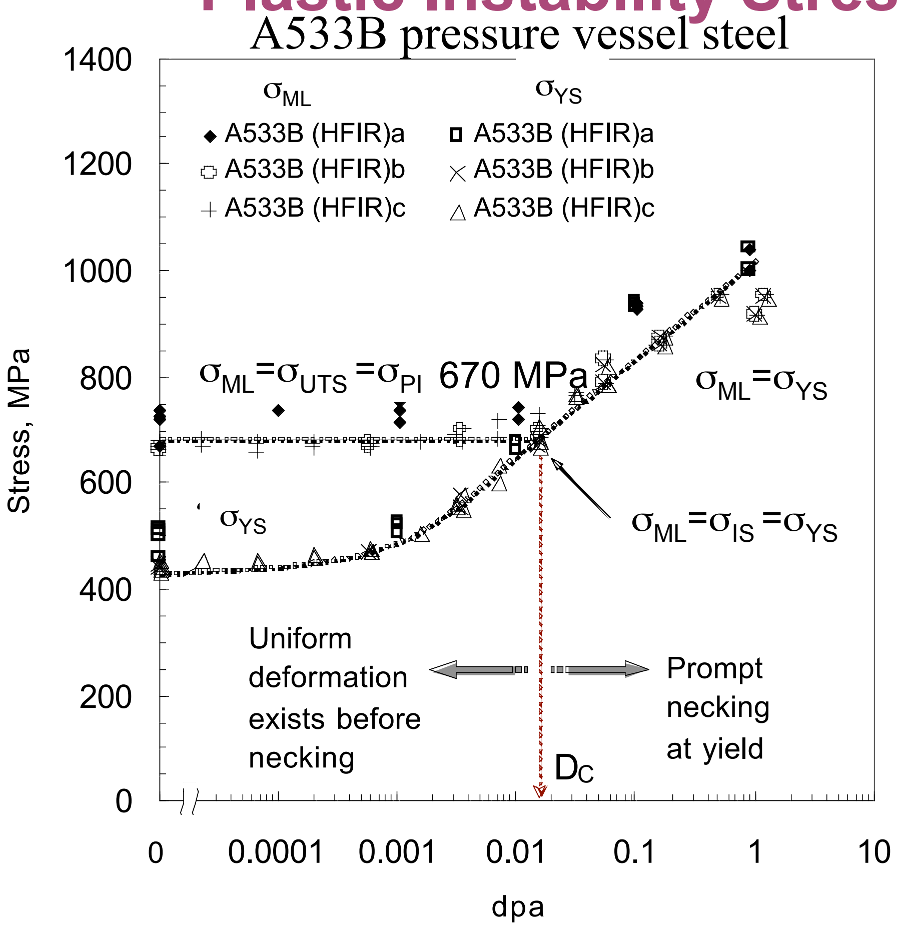


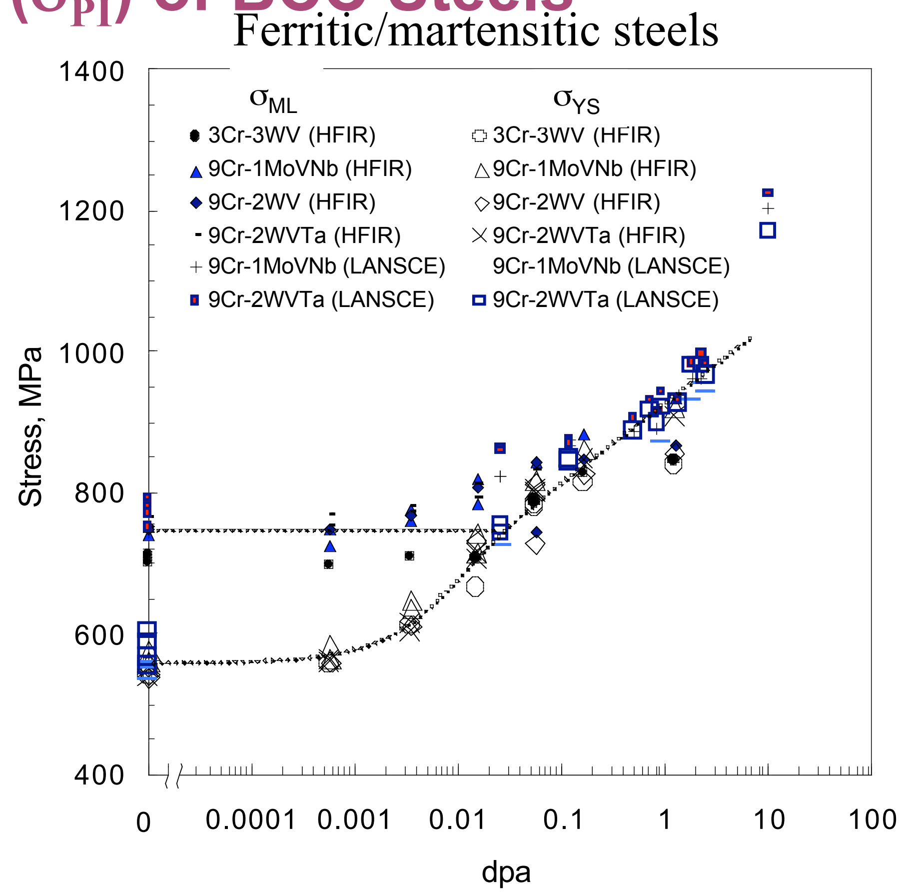
Austenitic Stainless Steel exhibits low uniform elongation after irradiation at temperatures up to ~400˚C

Reduction in uniform elongation requires higher doses than in simple metals (e.g. Cu, Ni)



Plastic Instability Stress (σ_{PI}) of BCC Steels



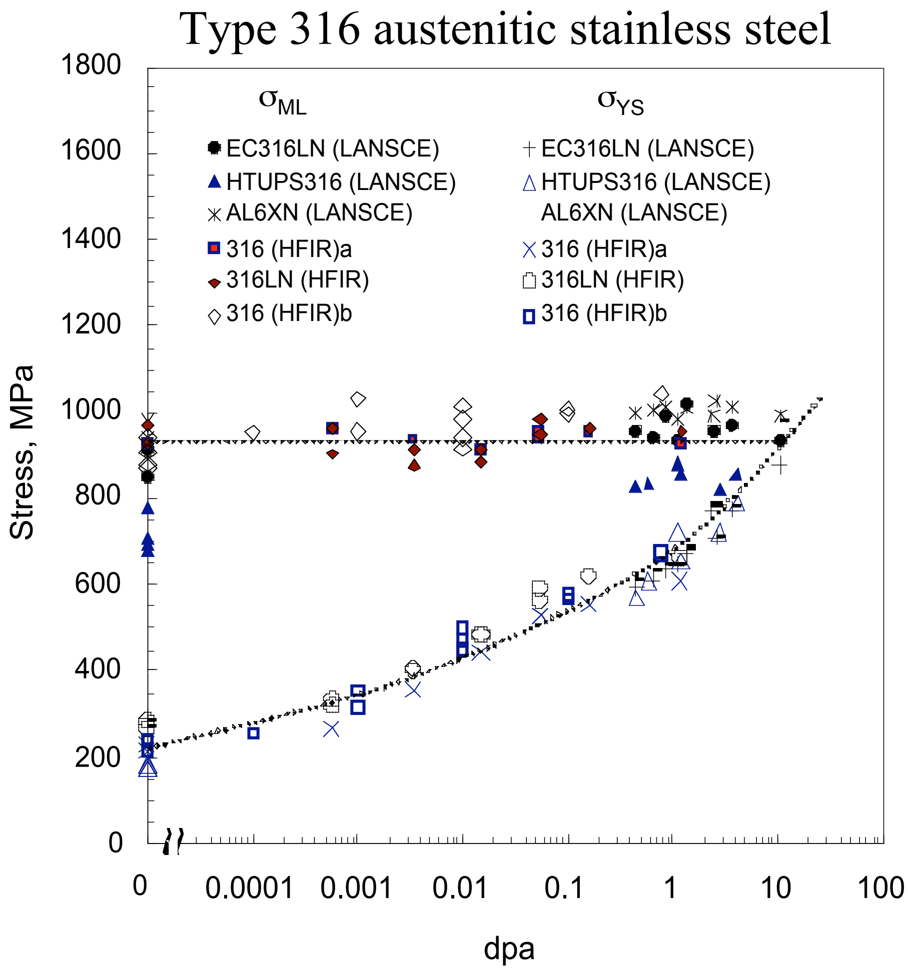


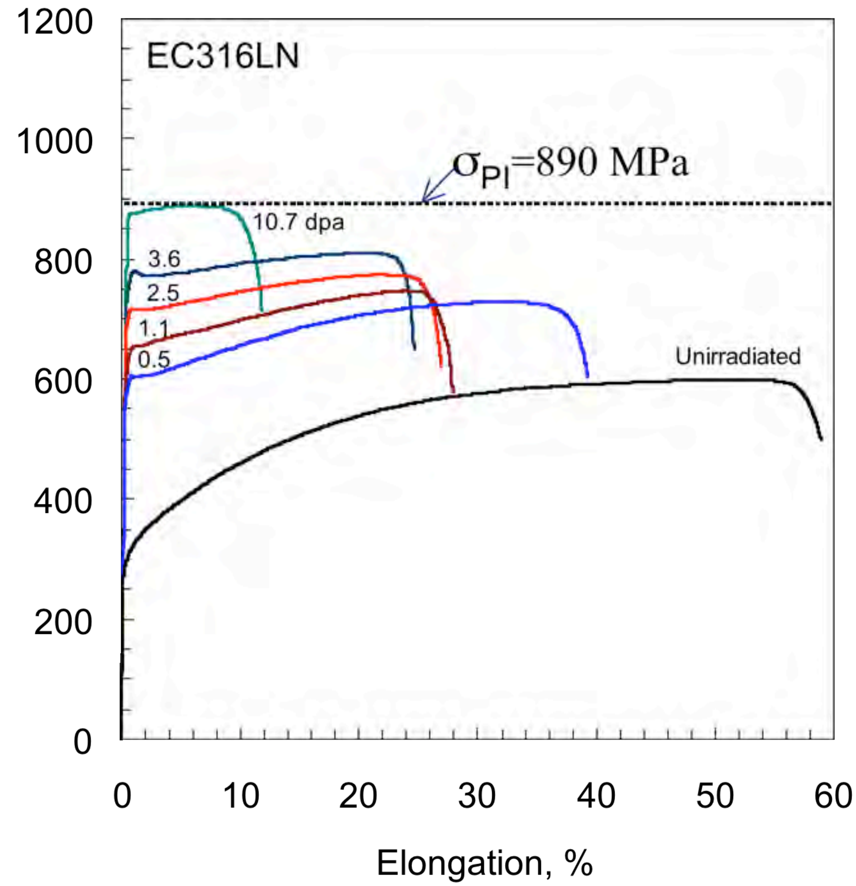
σ_{ML}=true stress at maximum load

- Plastic Instability Stress (σ_{PI}) = the true stress version of Ultimate Tensile Stress
- Plastic Instability Stress is independent of dose when yield stress < σ_{PI}.
- Yield stress can be > σ_{PI}, which is defined only when uniform deformation exists.
- σ_{PI} is considered to be a material constant, independent of initial cold-work or radiation-induced defect clusters

T.S. Byun & K. Farrell,
Acta Mater. 52 (2004) 1597

Plastic Instability Stress (σ_{PI}) of austenitic stainless steel irradiated near 70˚C

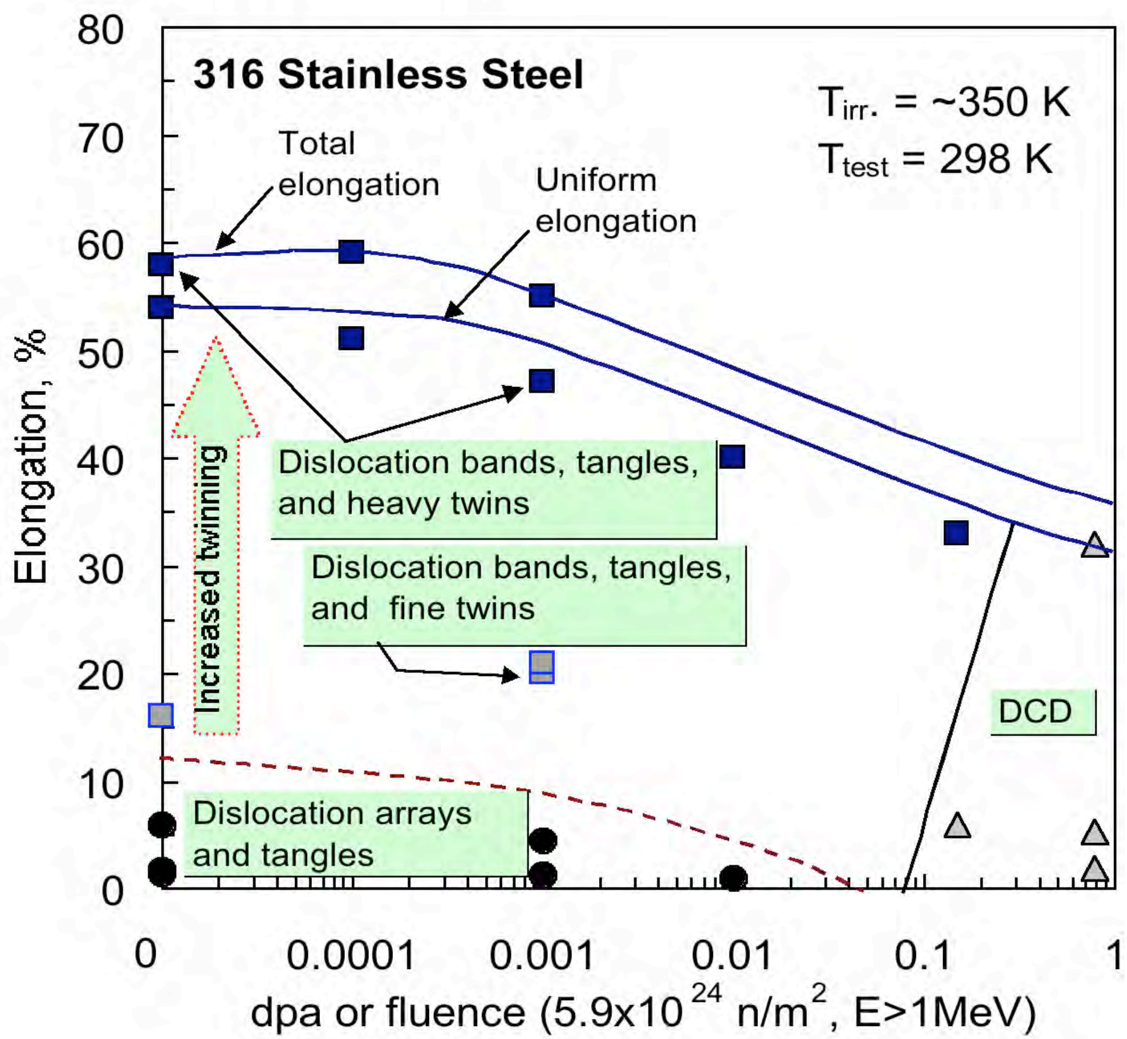




T.S. Byun & K. Farrell,
Acta Mater. 52 (2004) 1597

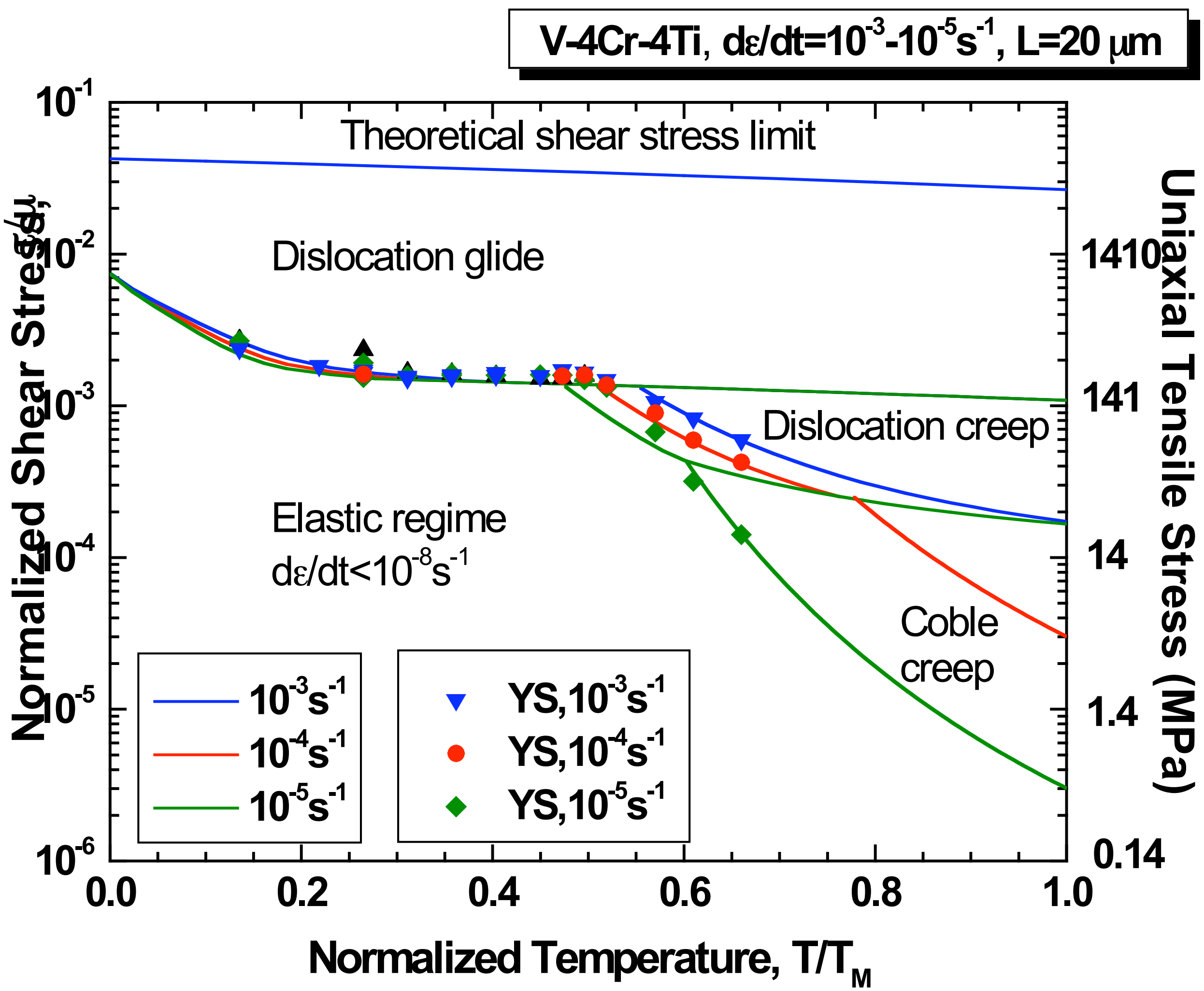
Deformation mode map for 316 SS neutron-irradiated at 65-100^{o}C and tested at 25°C

K. Farrell et al. ORNL/TM-2002/66 (2002)



Deformation Mechanism Map for Unirradiated V-4Cr-4Ti at Strain Rates of 10^{-3}-10^{-5} s^{-1}:

Comparison of experimental data and revised calculated map



M. Li and S.J. Zinkle, J. ASTM Intern. 2(2005)12462

The Operating Window for BCC metals can be Divided into Four Regimes (red values are relevant for Nb1Zr)

I, II: Low Temperature Radiation Embrittlement Regimes

- Fracture toughness (K_J) embrittlement: high radiation hardening causes low resistance to crack propagation (occurs when S_U>500-700 MPa)
  - Regimes which cause K_J<30 MPa-m$^{1/2}$ should be avoided (T_{irr}< ~600 K ?)
- Loss of ductility: localized plastic deformation requires use of more conservative engineering design rules for primary+secondary stress (S_e)

$$S_e = \begin{cases} \frac{1}{3} S_u & \varepsilon_U < 0.02 \\ \frac{1}{3}\left[S_u + \frac{E(\varepsilon_U - 0.02)}{8}\right] & \varepsilon_U > 0.02 \end{cases}$$

(T_{irr}< ~900-1270 K)

where ε_U is uniform elongation, S_U is ultimate tensile strength, E is elastic modulus (additional design rules also need to be considered)

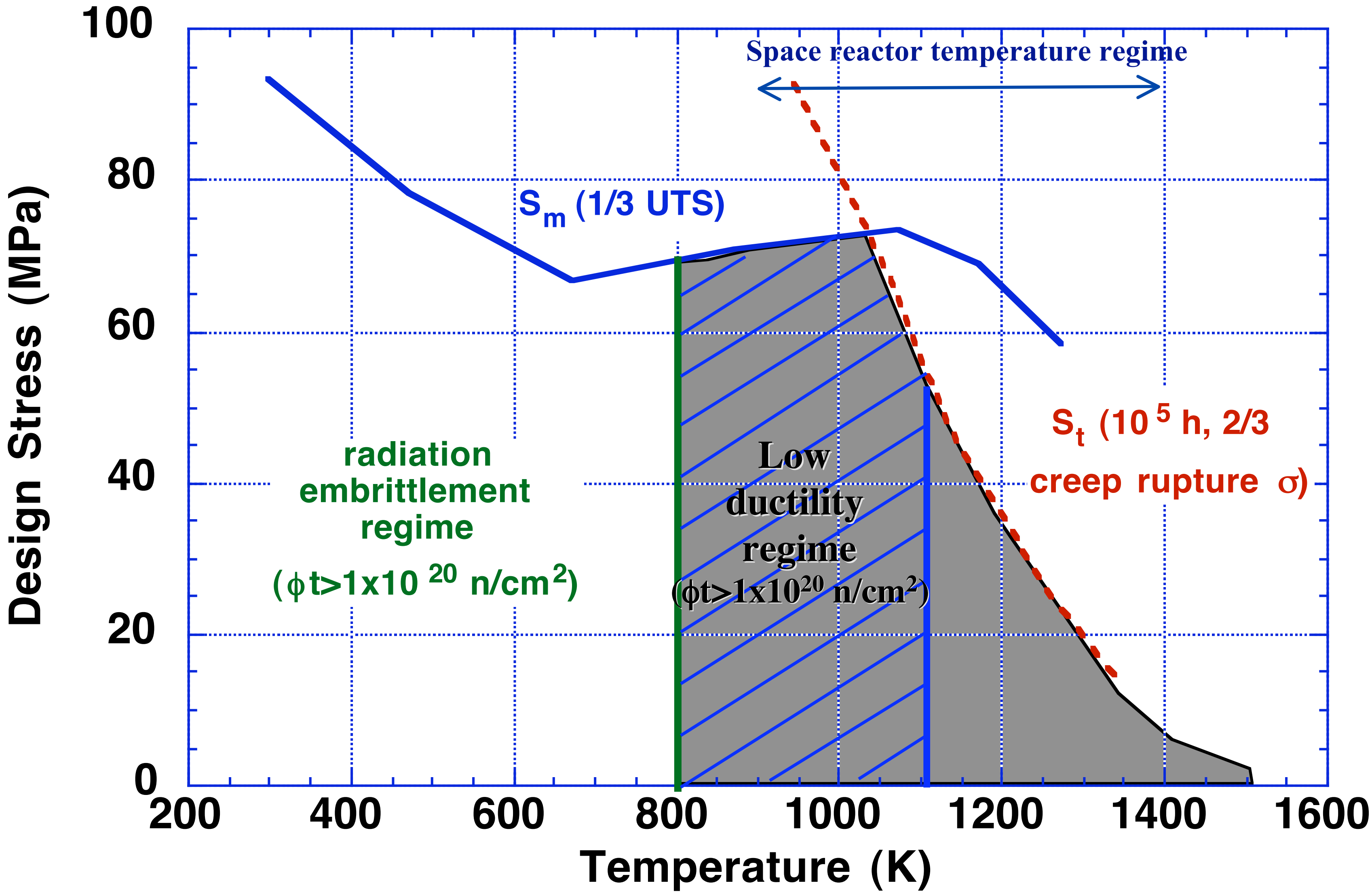
III: Ductile Yield and Ultimate Tensile Strength Regime (e_U>0.02)

- Sets allowable stress at intermediate temperature (very small regime for Nb-1Zr)

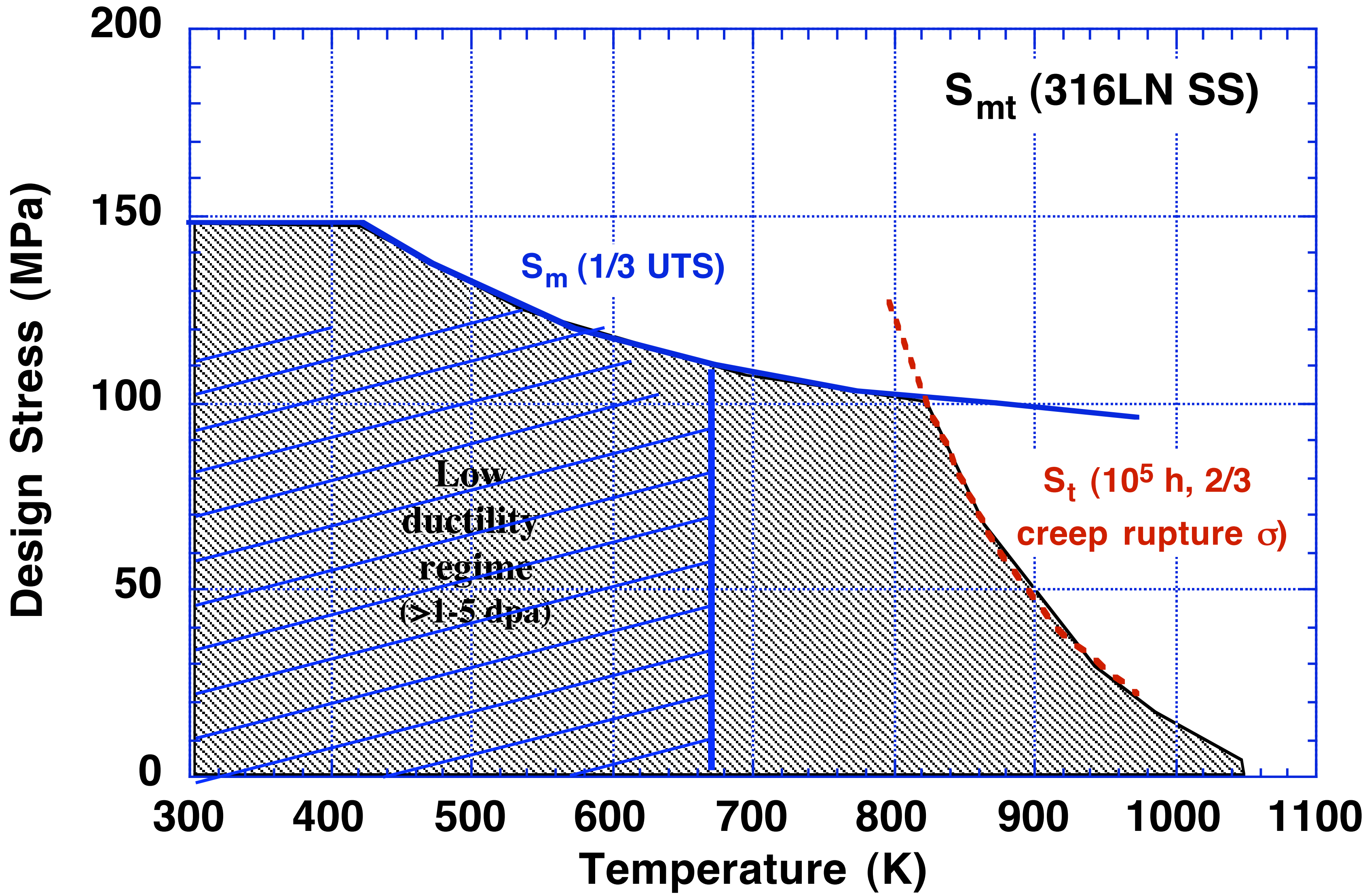
IV: High Temperature Thermal Creep Regime (T>~1050 K)

- Deformation limit depends on engineering application (common metrics are 1% deformation and complete rupture)

Stress-Temperature Design Window for Nb-1Zr

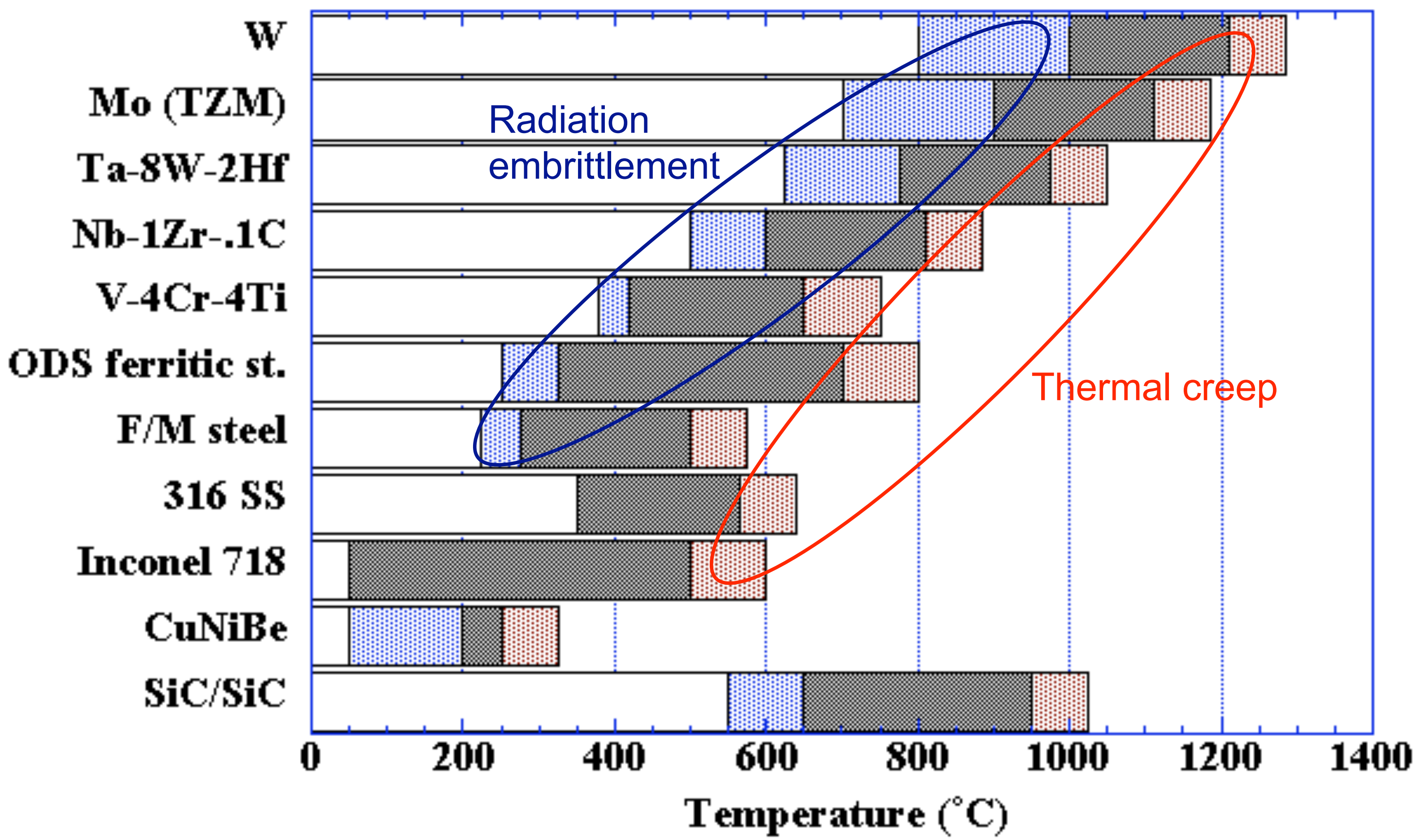


Stress-Temperature Design Window for Unirradiated Type 316 Stainless Steel



Can we break the shackles that limit conventional structural materials to ~300^{o}C temperature window?

Structural Material Operating Temperature Windows: 10-50 dpa



η_{Carnot}=1-T_{reject}/T_{high}

Low temperature radiation embrittlement typically occurs for damage levels ~0.1 dpa (0.01 MW-yr/m^2)

Zinkle and Ghoniem, Fusion Engr. Des. *<u>51-52</u> (2000) 55*

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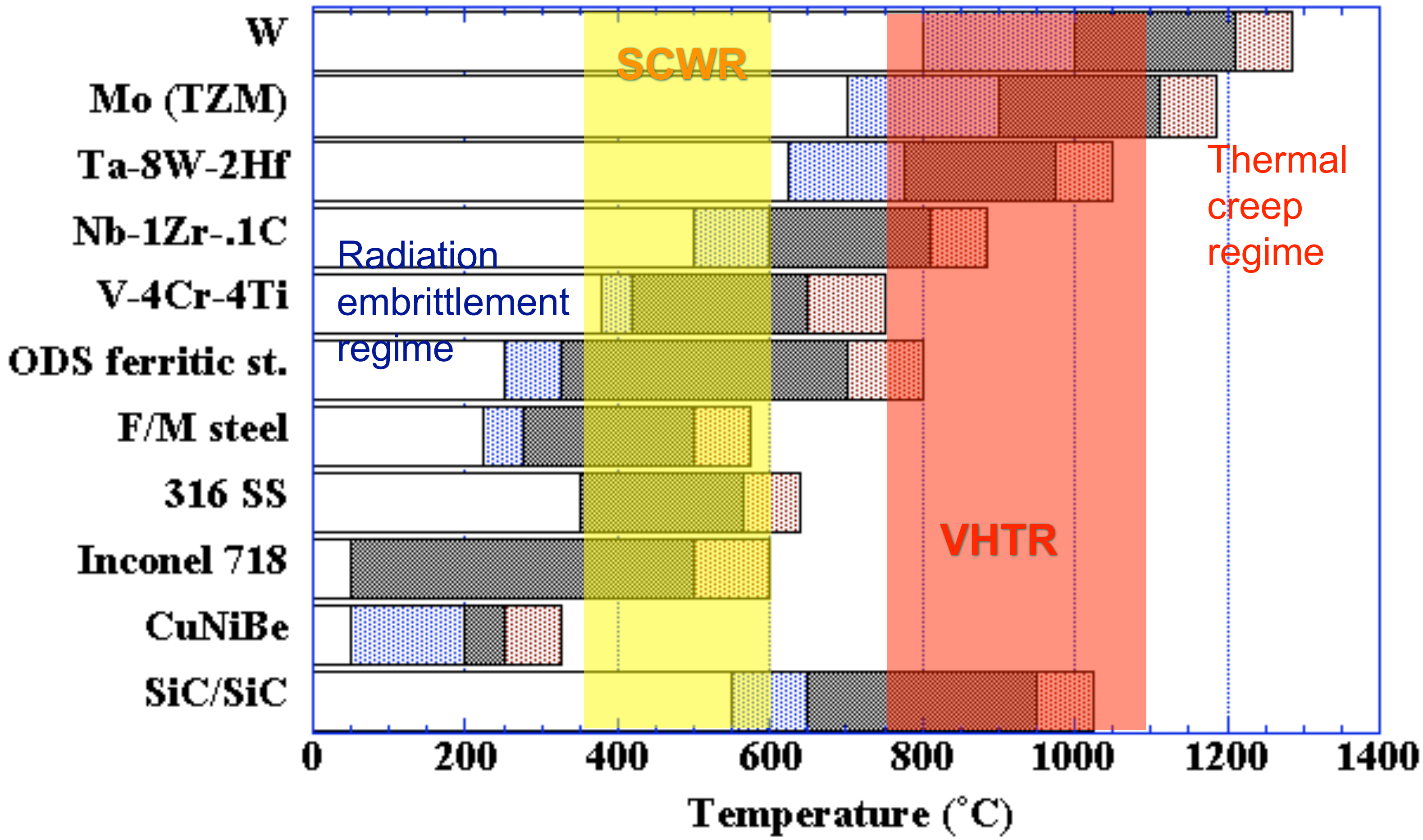

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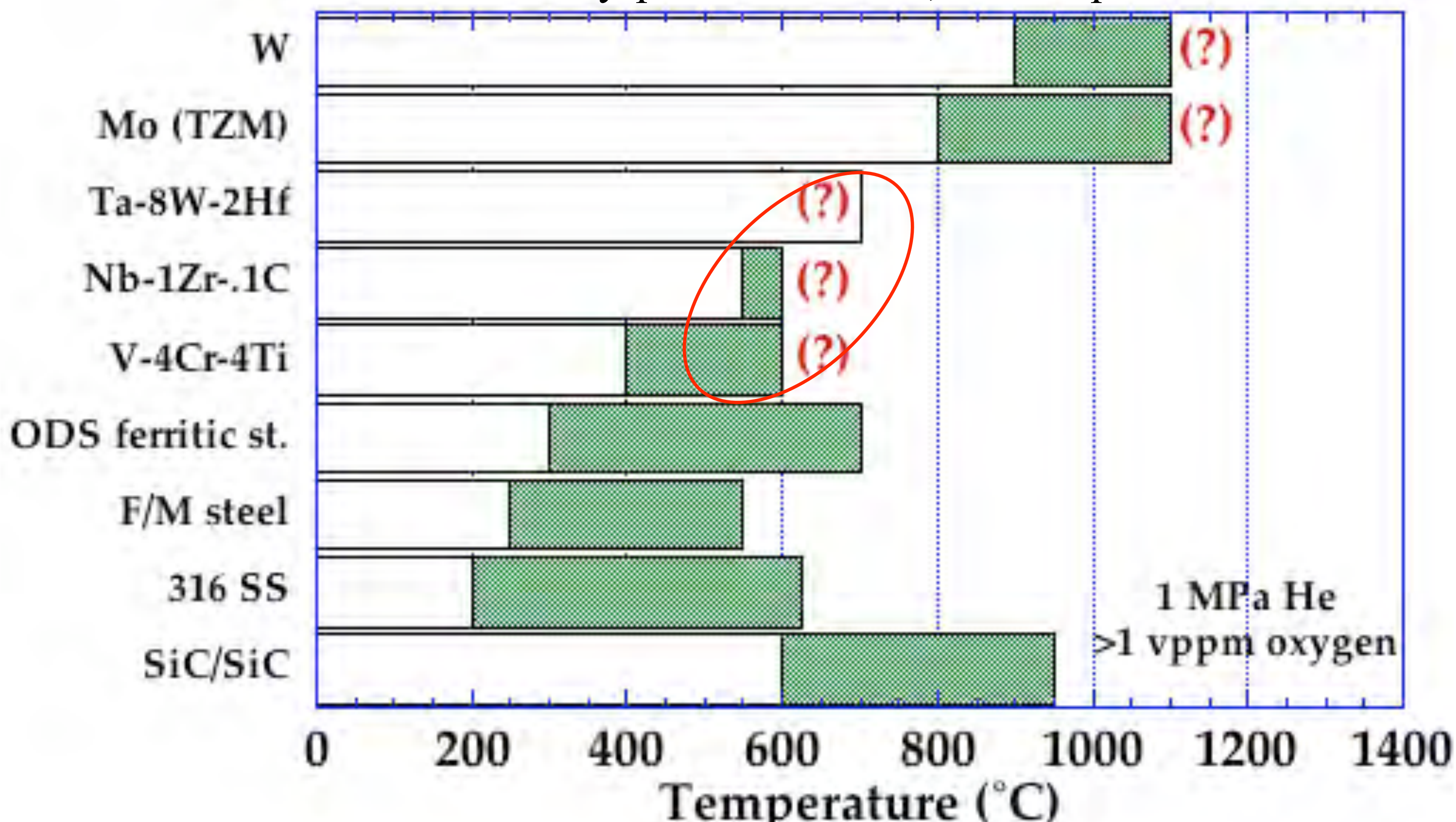
Zinkle and Ghoniem, Fusion Engr. Des. *51-52 (2000) 55*

Consideration of Chemical Compatibility can Result in Dramatic Reductions in Temperature Window

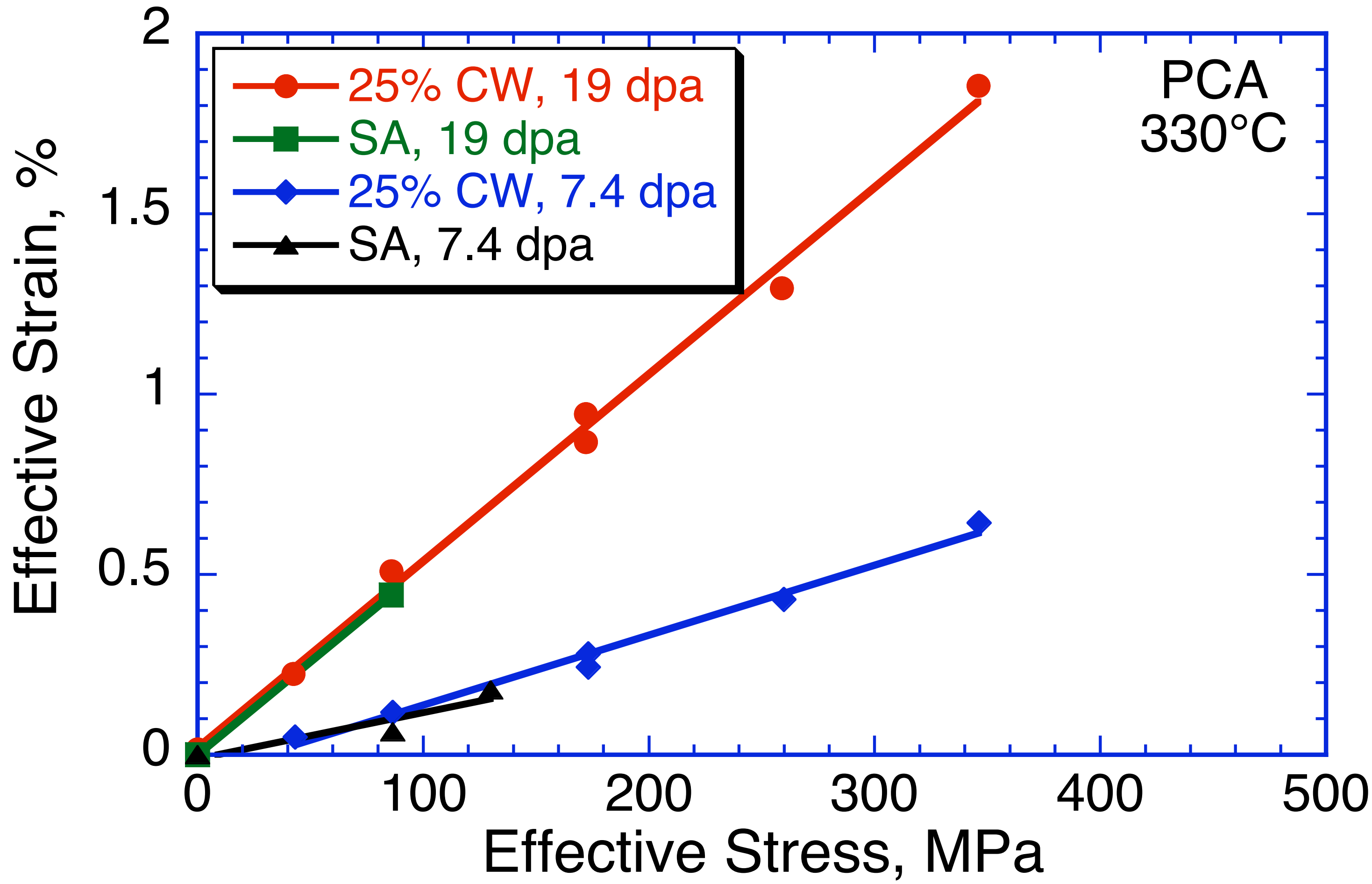
Estimated Structural Material Operating Temperature Windows:
Moderately-pure He coolant, 10-50 dpa



Group V refractory alloys are particularly sensitive to embrittlement from O,C,N solute

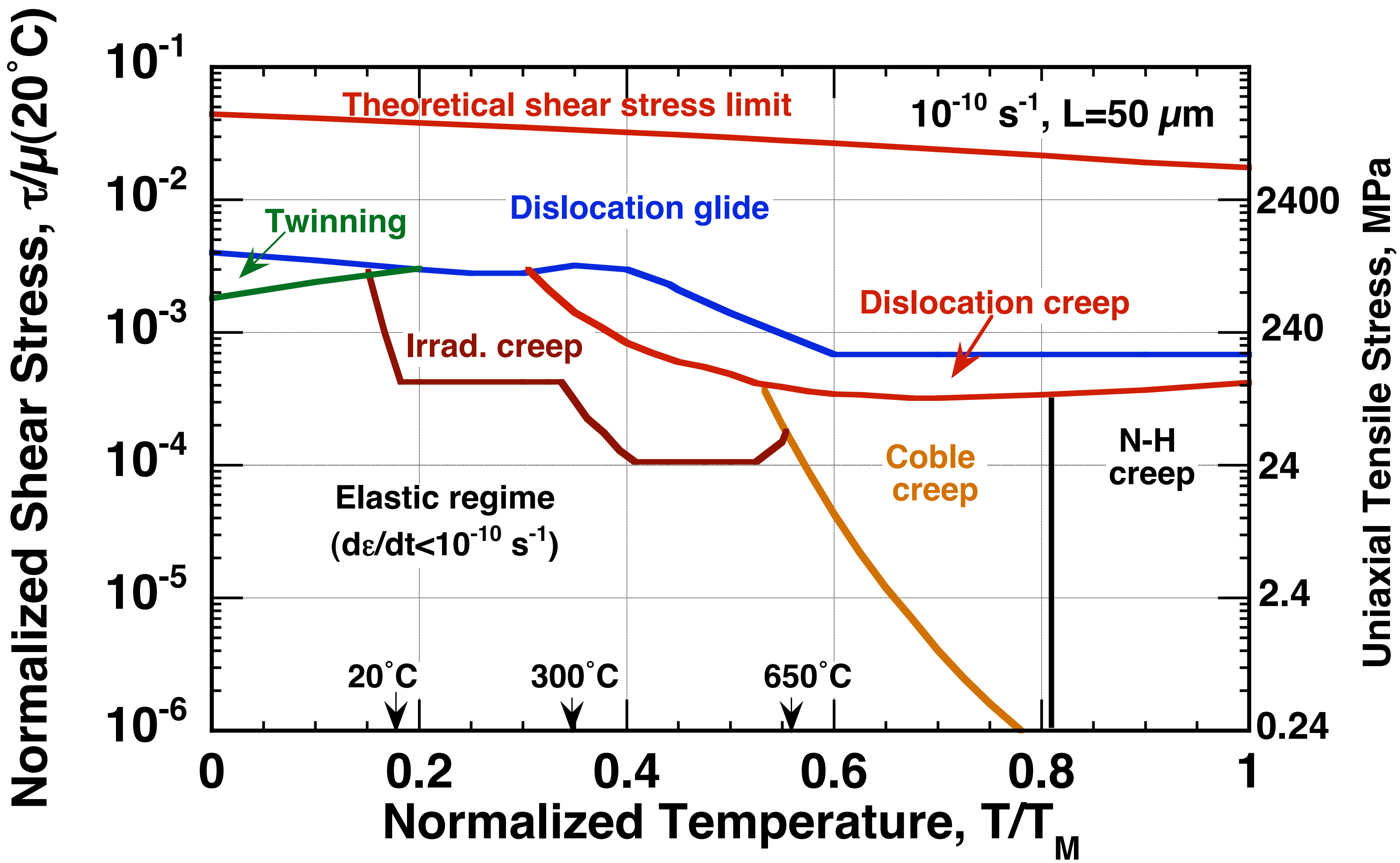
Zinkle and Ghoniem, Fusion Engr. Des. *51-52* *(2000) 55*

Irradiation Creep of Austenitic Stainless Steel will Generally be of Concern only for High Fluence (>20 dpa), High Stress Environments



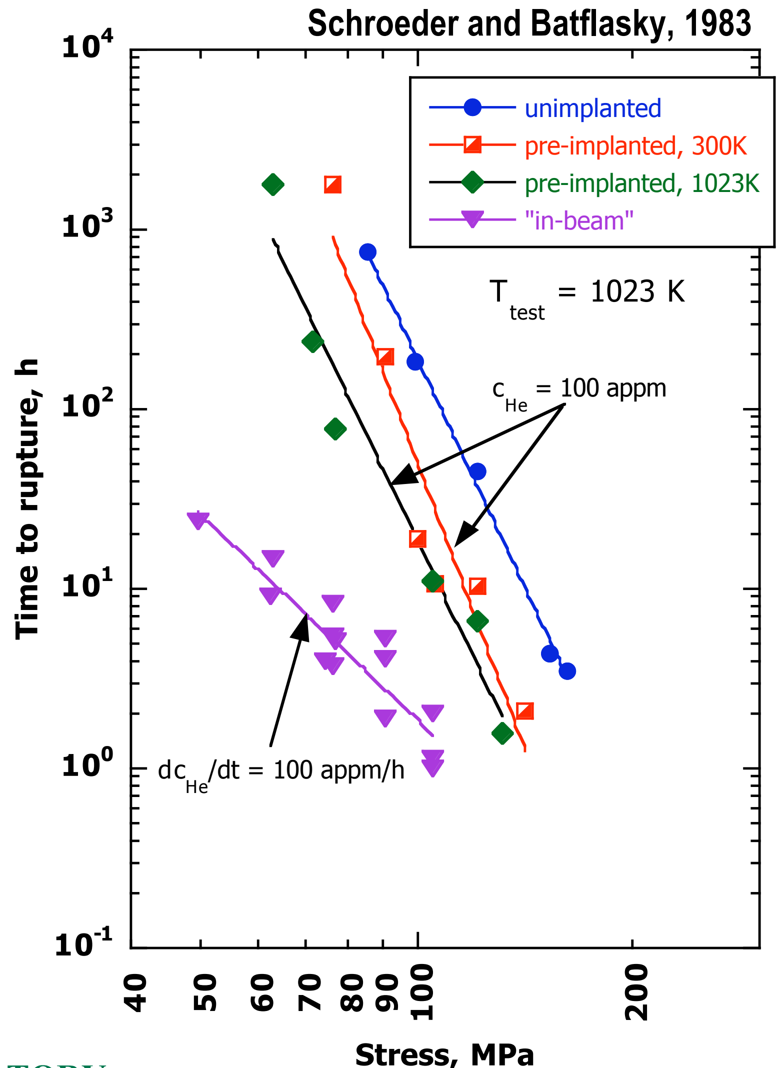
Calculated irradiated Ashby deformation map for Type 316 stainless steel at low strain rates

Damage rate = 10^{-6} dpa/s

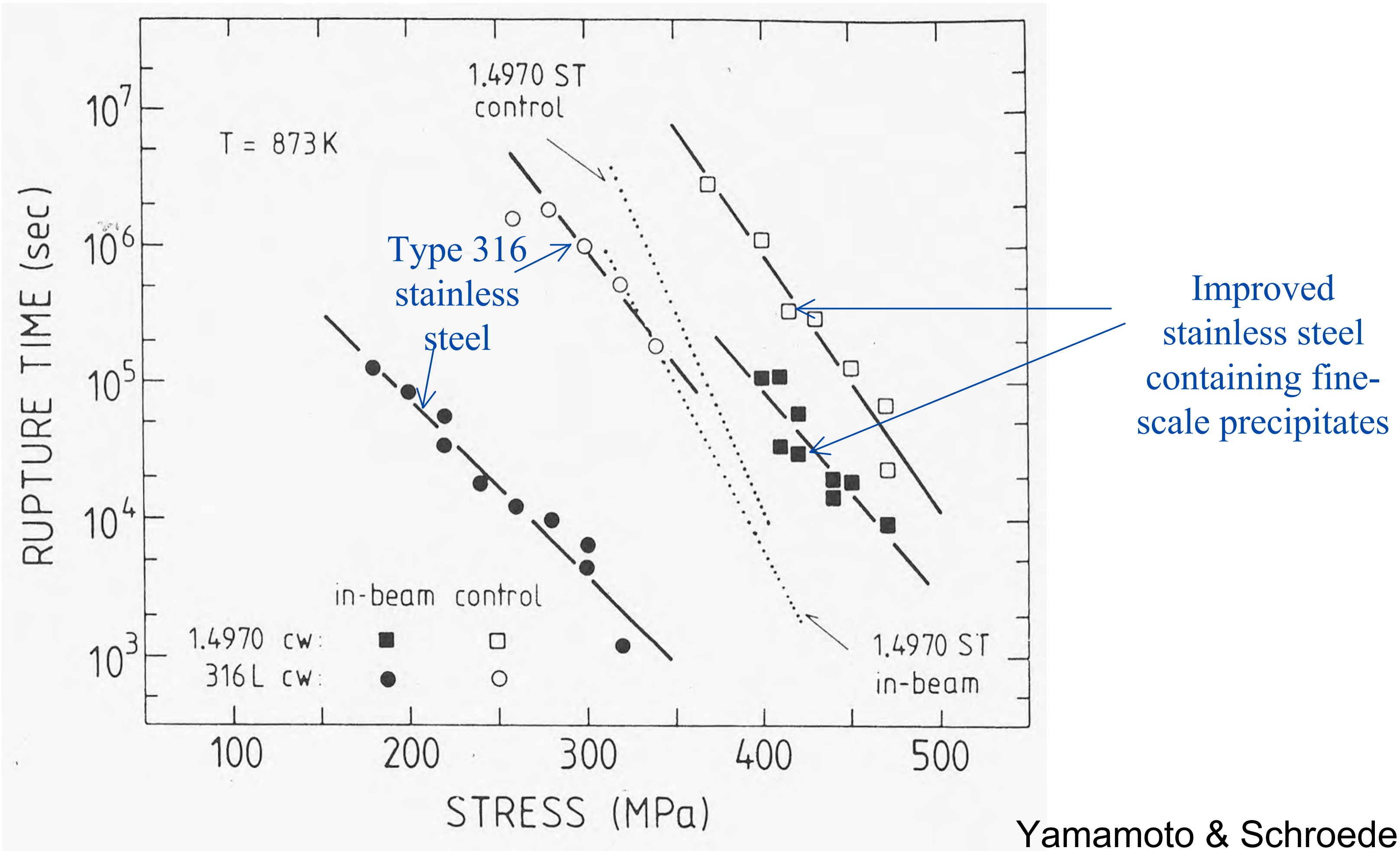


Effect of applied stress on the rupture lifetime of austenitic stainless steel annealed at 750˚C

- 100 appm He reduces rupture lifetime of stainless steel at 750˚C by 10-100X due to formation of grain boundary He cavities



Fine-scale matrix precipitates can trap He and provide greatly improved He embrittlement resistance



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

Austentic steels are typically more sensitive to helium embrittlement effects than ferritic/martensitic steels

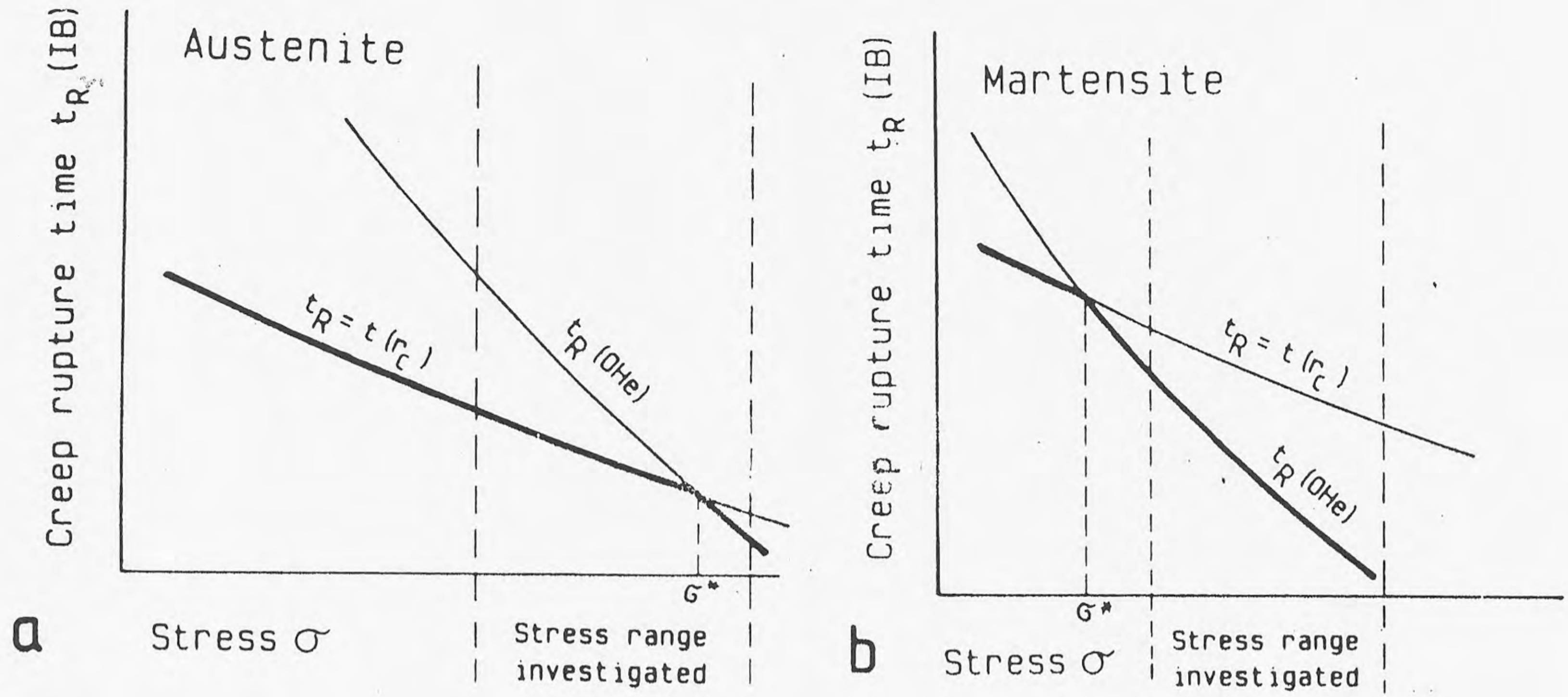


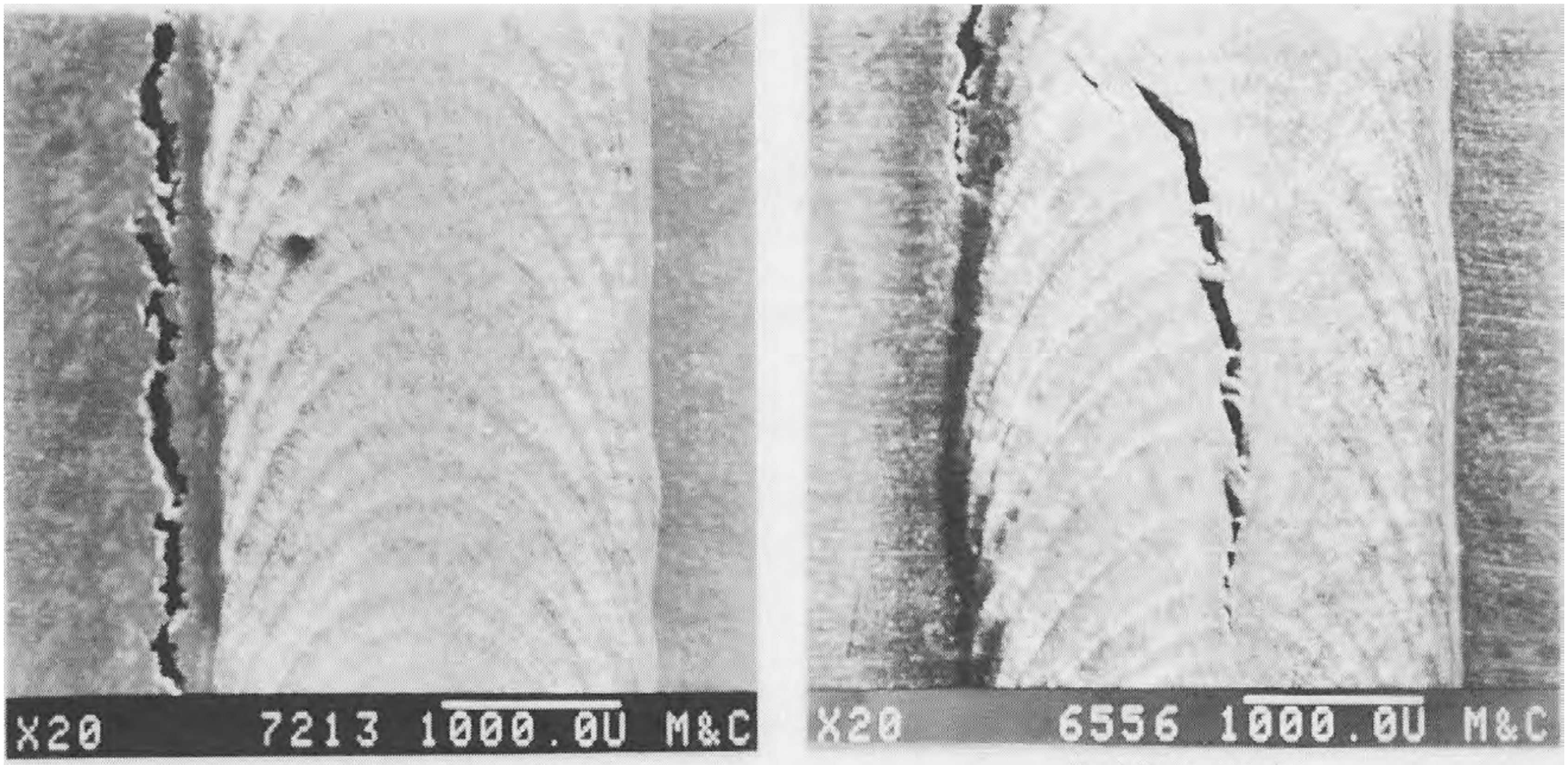
FIG. 7—*Comparison of fracture time* t_R *as a function of applied stress* σ *between unimplanted controls* (t_R(OHe)) *and stable gas-driven growth as a lifetime controlling mechanism* ($t_R = t(r_c)$) (*schematic*). *Indicated is the stress range investigated and the transition stress* σ*, *at which 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

Cracking is observed in welded metals containing He (from nuclear transmutations)

Occurs in heat affected zone for low He concentrations

At high He concentrations, cracking also occurs in the fusion zone (weld bead)

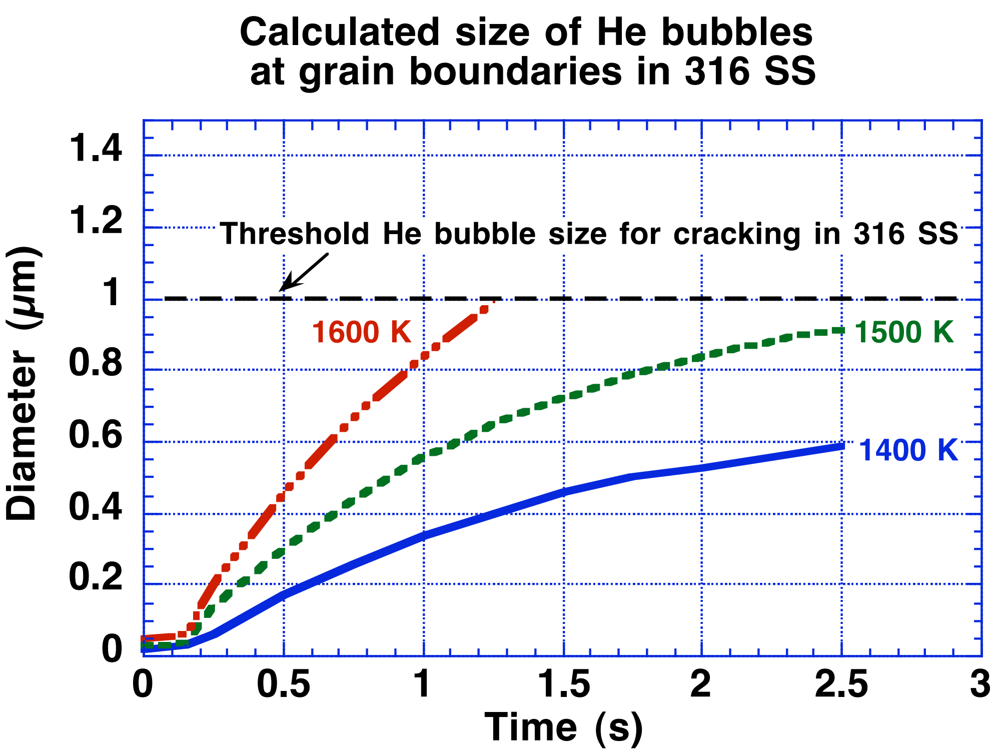


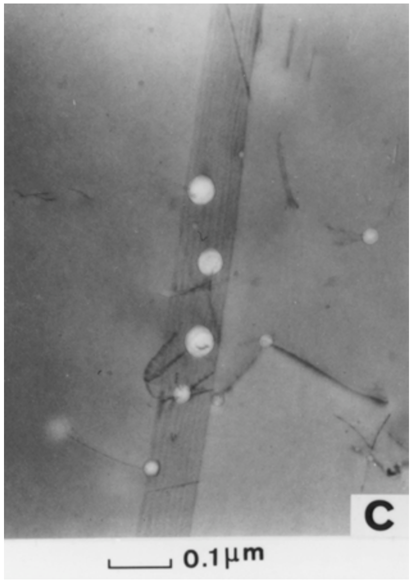
(2.5 appm) (105 appm)

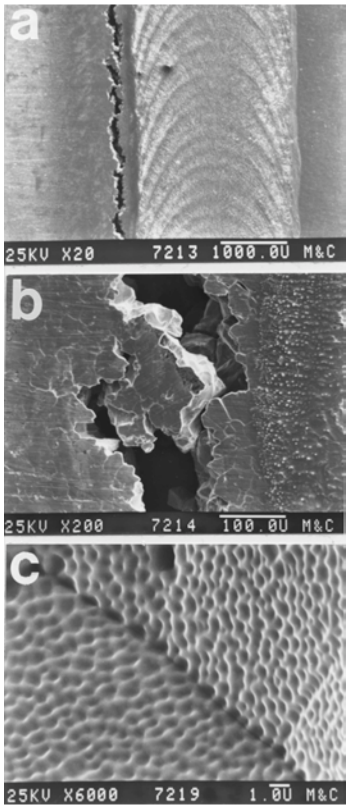
Heat affected zone (HAZ) Weld bead HAZ

Effect of transmutant He on welding behavior of metals

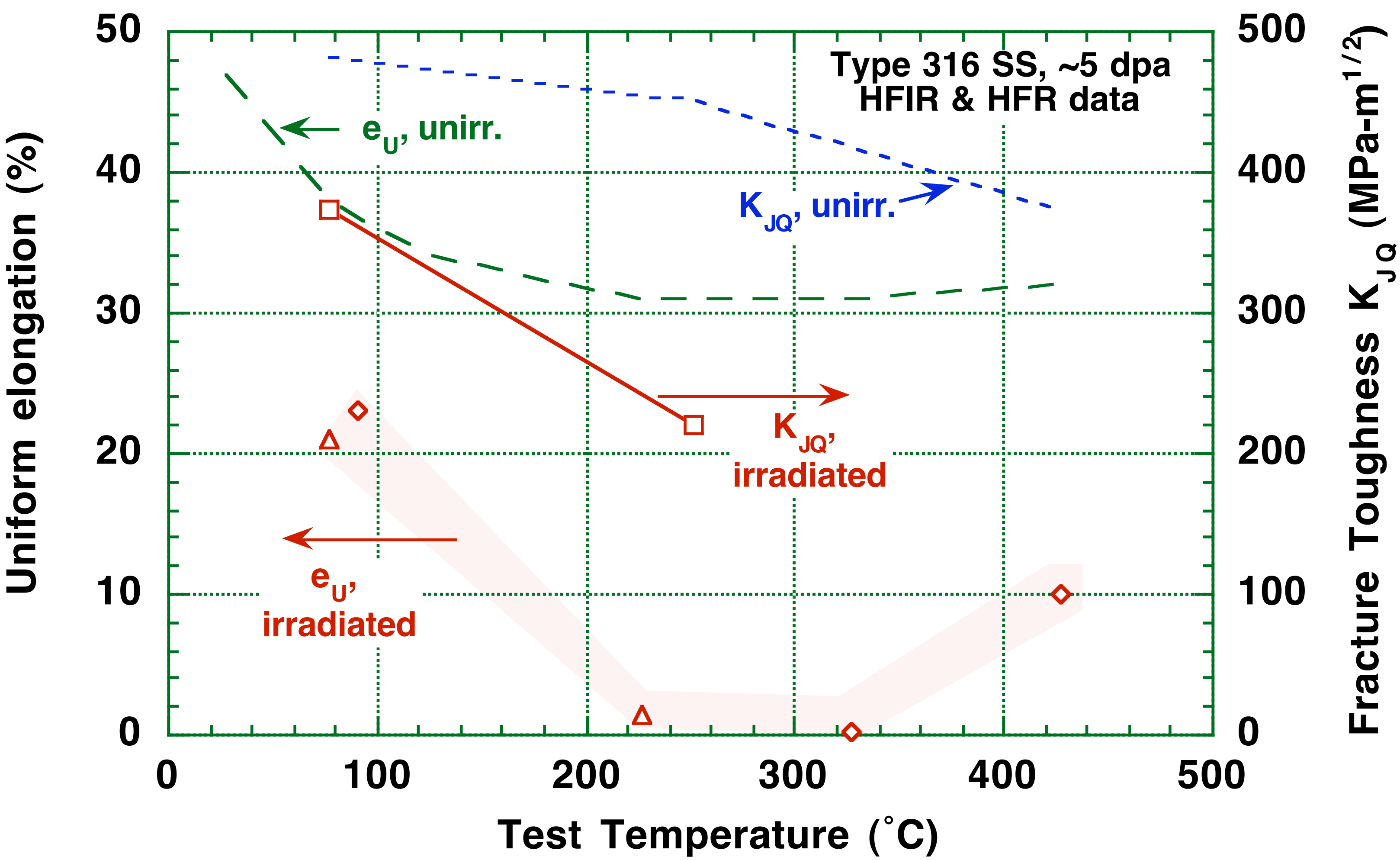
- Irradiated materials with He contents above ~1 appm cannot be fusion-welded due to cracking associated with He bubble growth; the lower temperatures associated with FSW may allow repair joining of irradiated materials





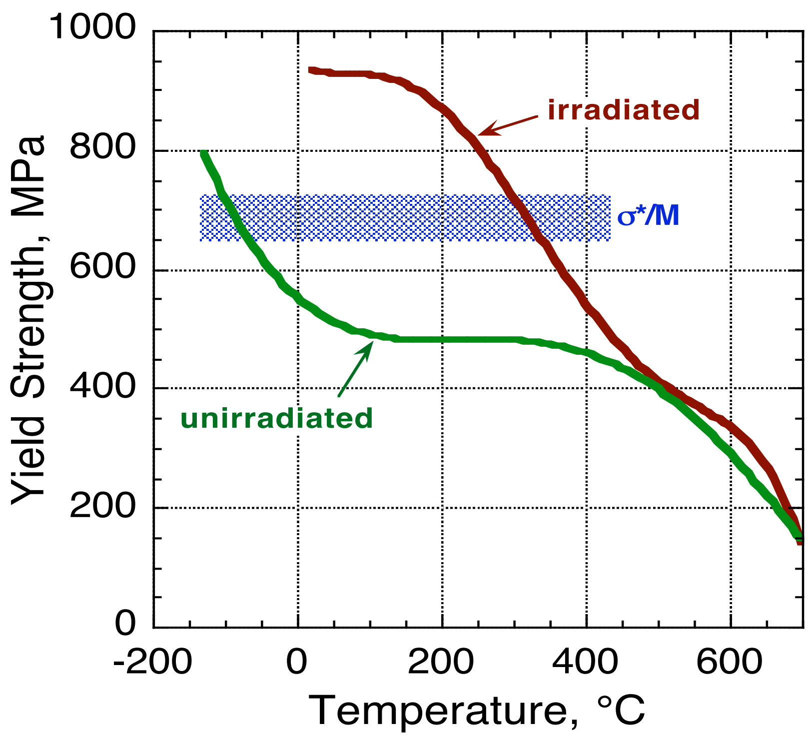


Irradiation of Austenitic Stainless Steel in Mixed Spectrum Reactors causes Pronounced Loss in Elongation and Significant Reduction in Fracture Toughness

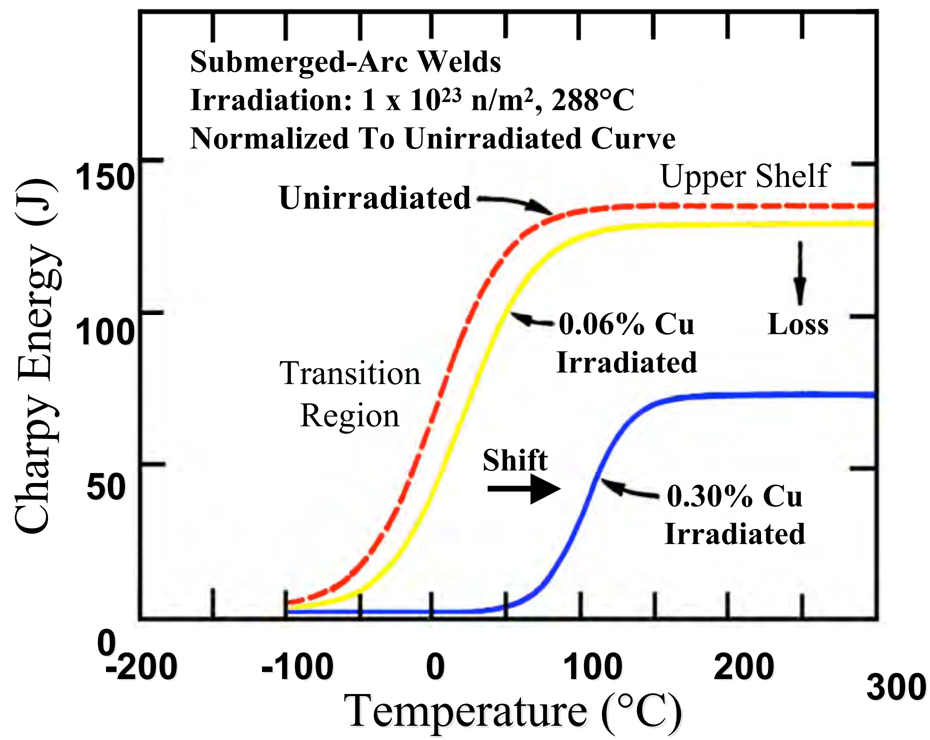


Fracture Toughness of Irradiated BCC Structural Alloys

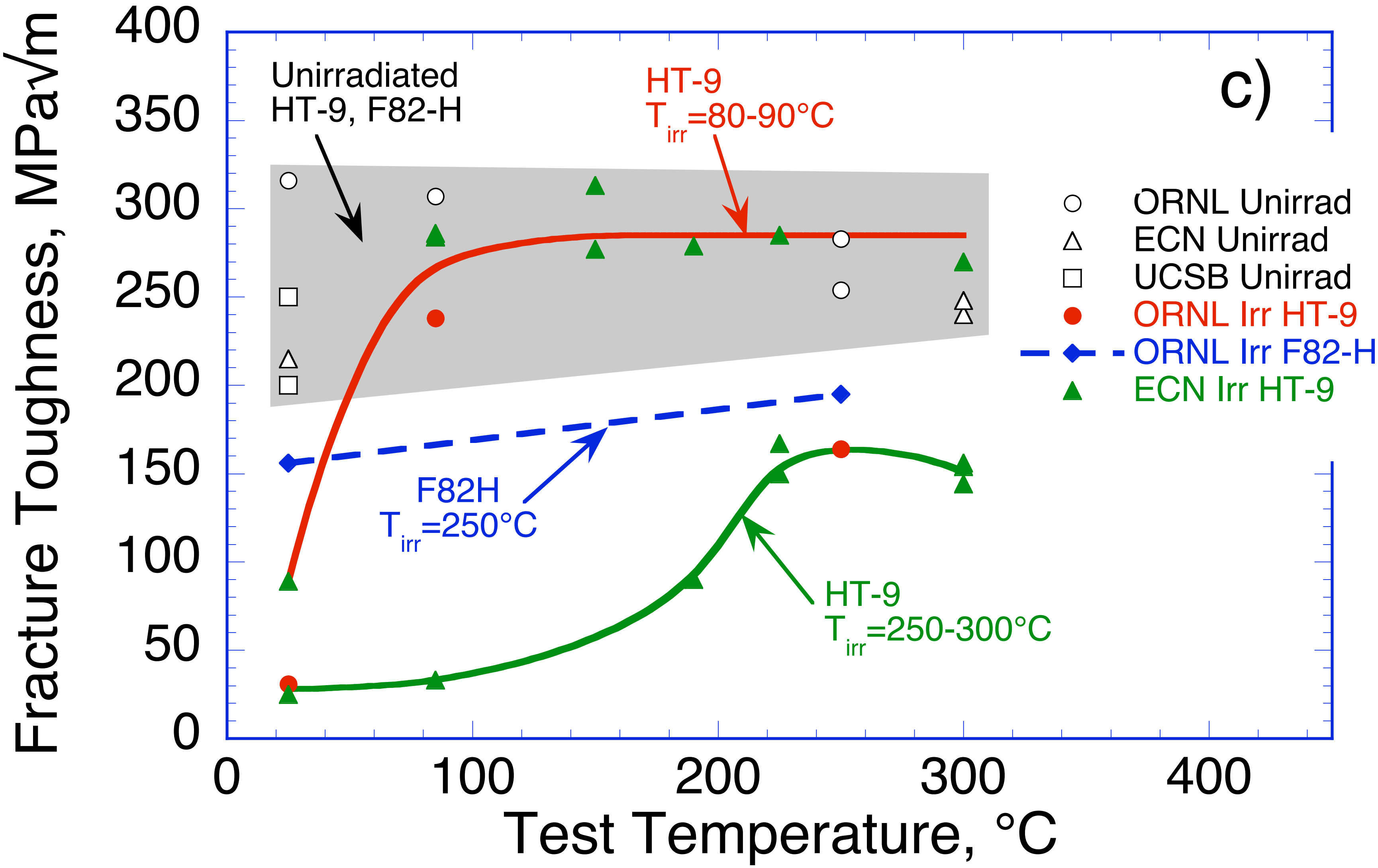
- There are two general approaches to mitigate DBTT increases
  - Reduce radiation hardening by alloying modifications (e.g., low-Cu RPV steels)
  - Increase σ*



- Significant improvements in resistance to low temperature radiation embrittlement can be achieved by selective alloying (e.g., reduced Cu in reactor pressure vessel steels)



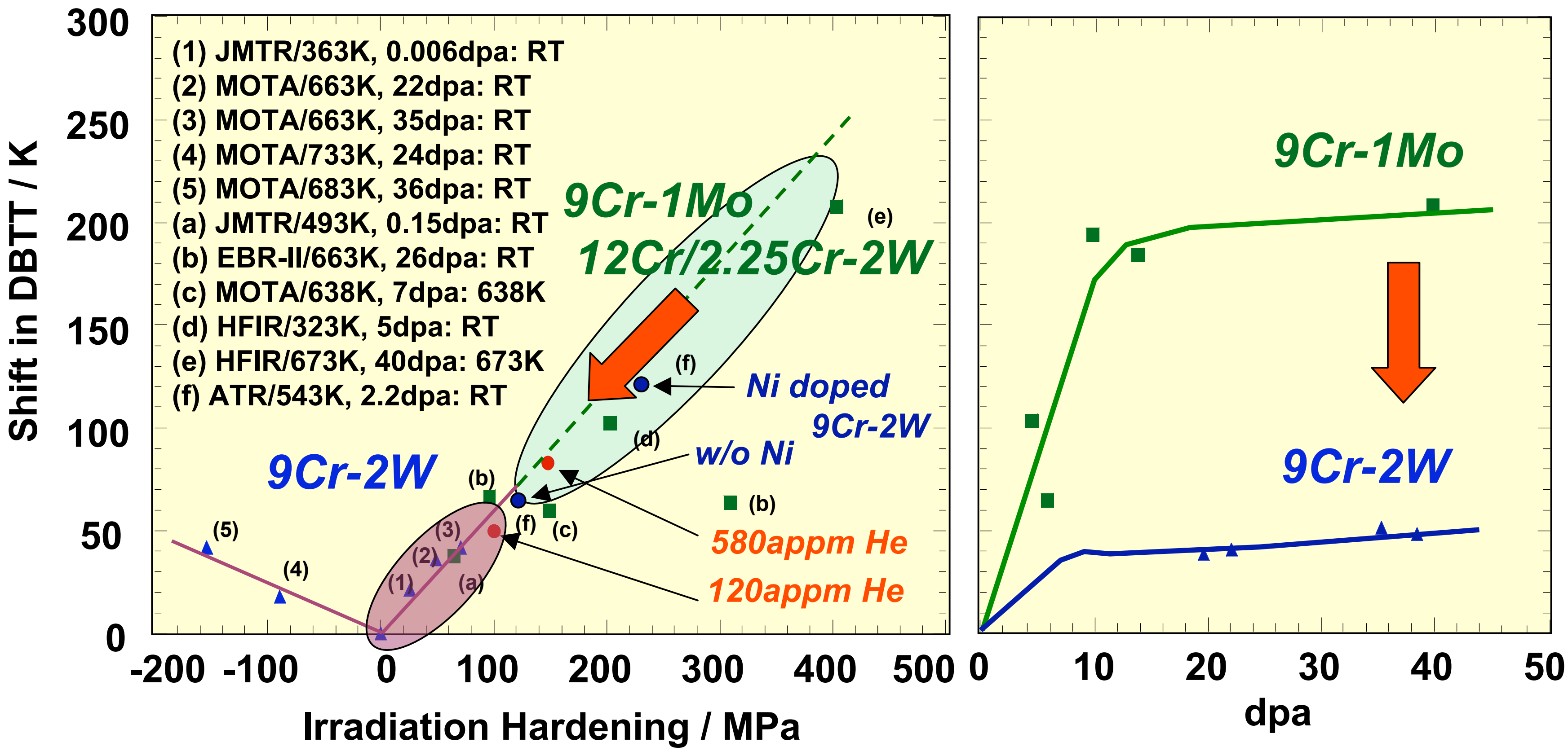
Comparison of the fracture toughness of 9Cr and 12Cr steels



A.F. Rowcliffe et al., J. Nucl. Mater. 258-263

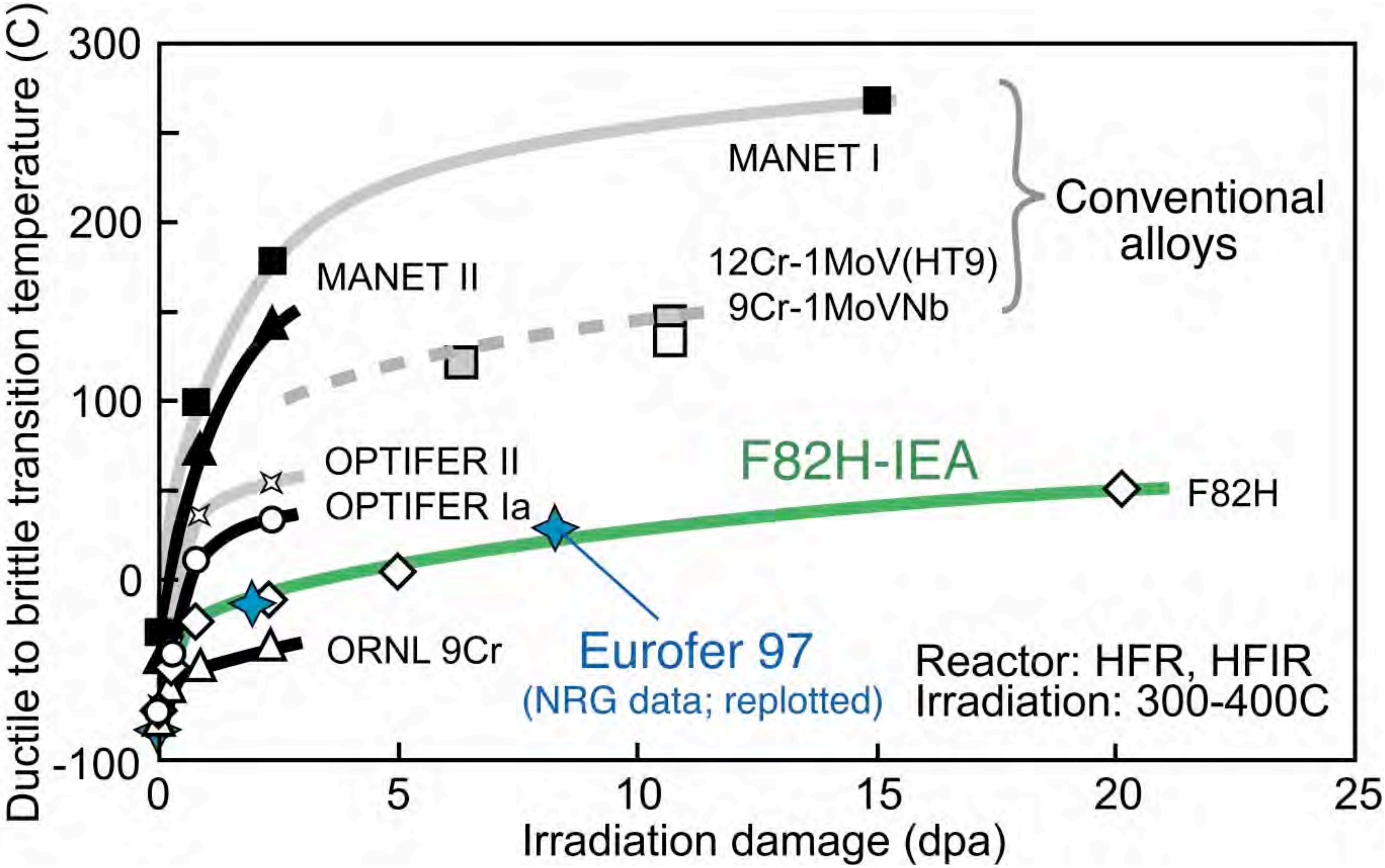
Fusion Reduced Activation Ferritic/martensitic Steels Exhibit Superior Irradiation Performance Compared to Conventional 9Cr-1Mo and 12Cr Steels

● Suppression of irradiation embrittlement

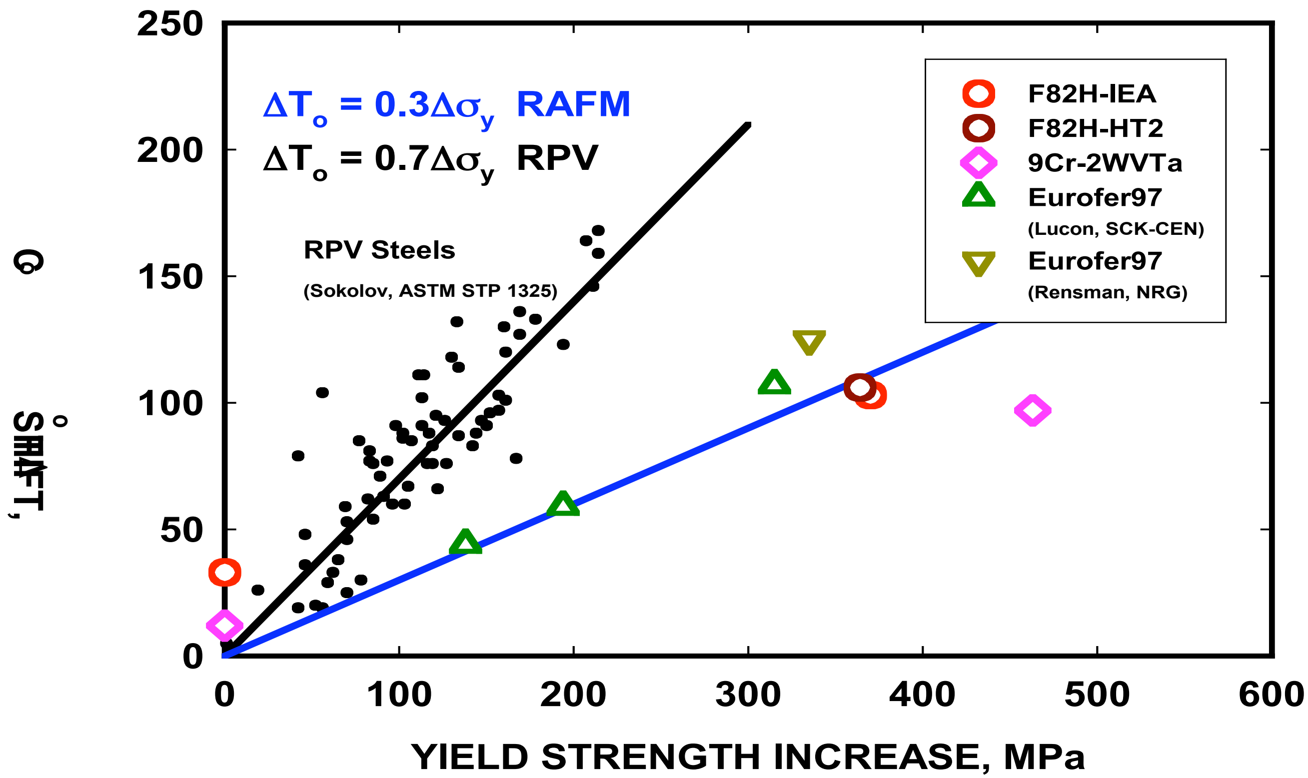


A. Kohyama, ANS TOFE (Madison, WI, 2004)

Effect of Neutron Irradiation on DBTT of Ferritic/martensitic Steels

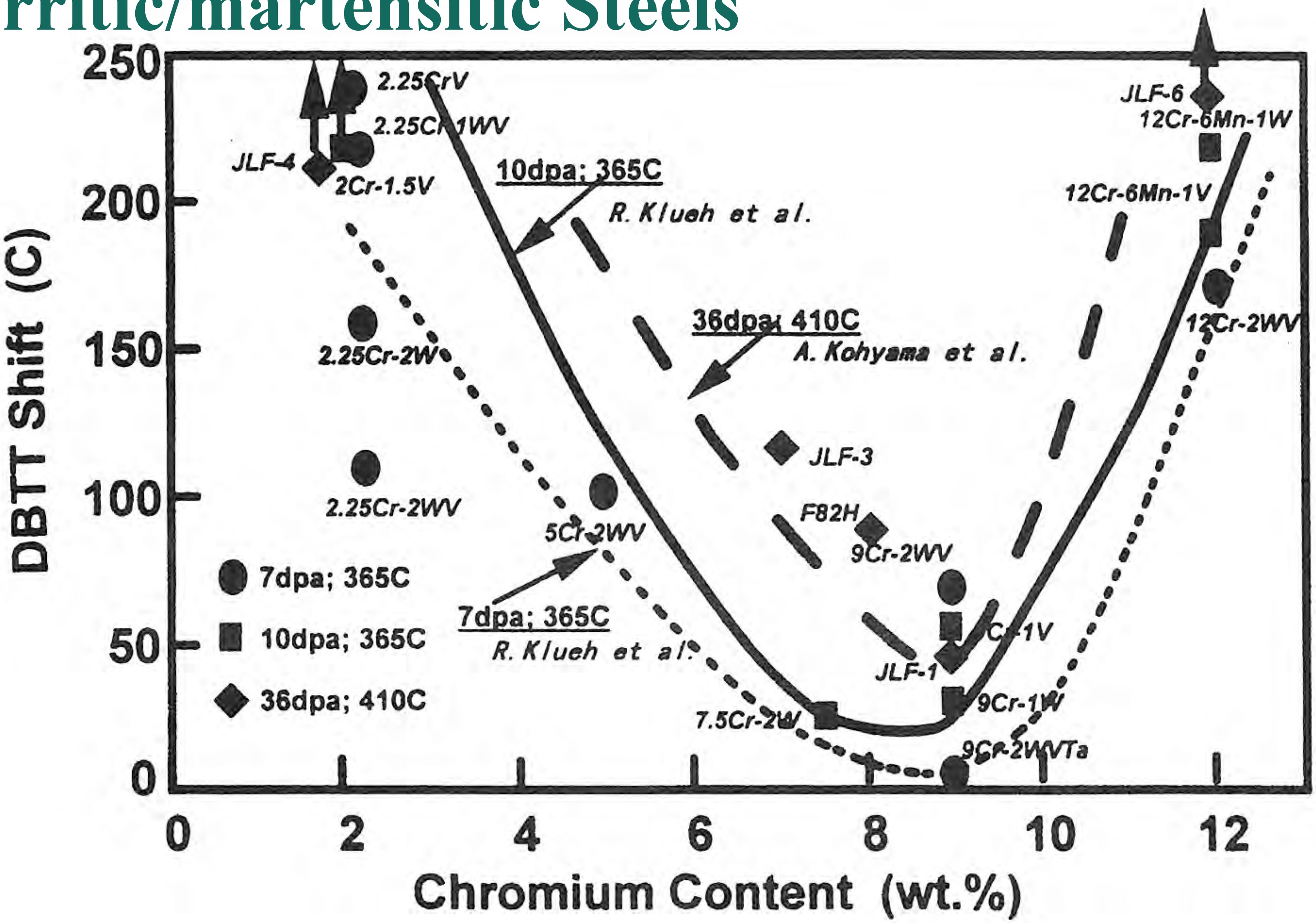


8-9%Cr RAFM Steels Exhibit Less Embrittlement Per Unit of Hardening Than Low-Alloyed RPV Steels



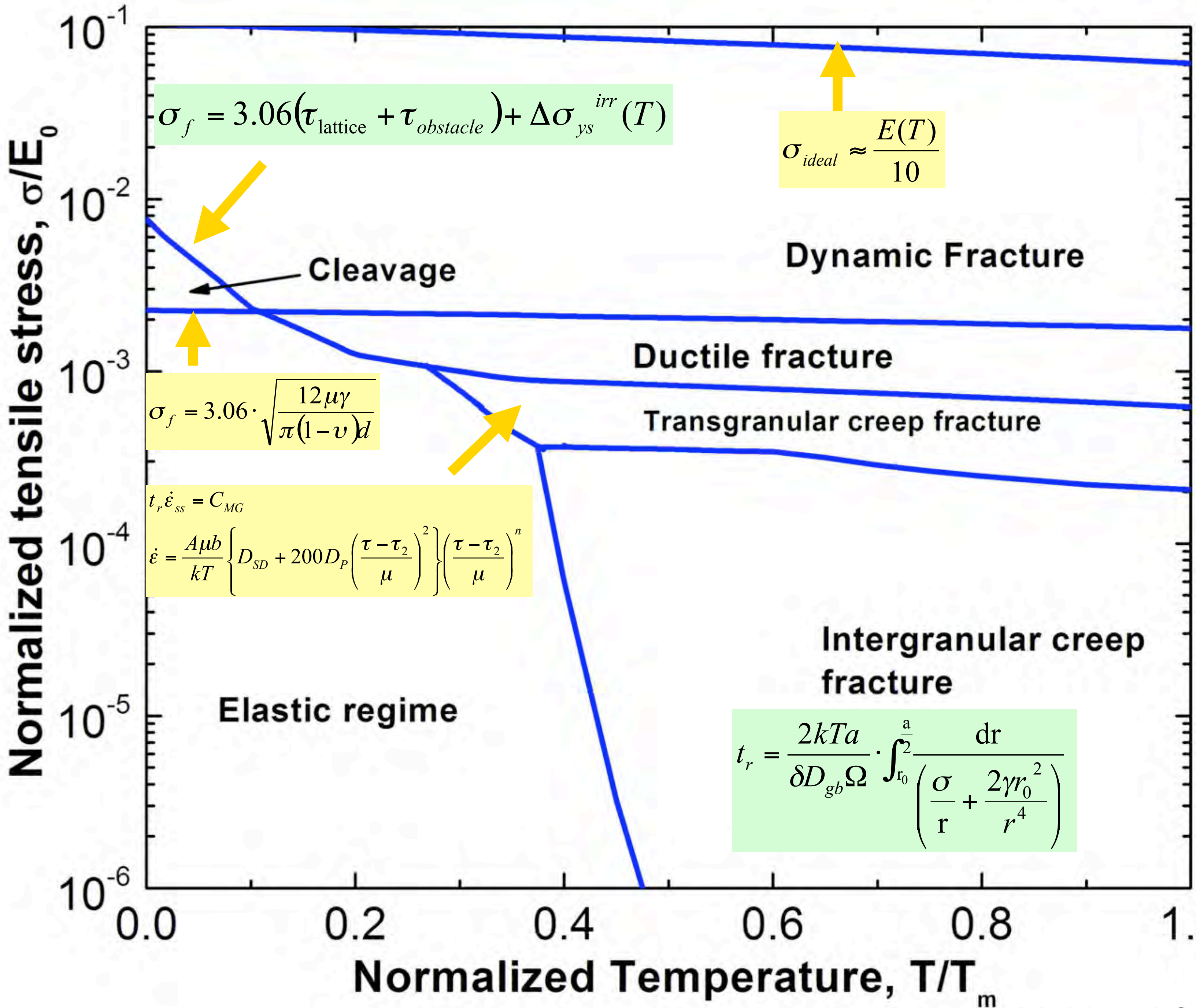
M.A. Sokolov, ICFRM12, J. Nucl. Mater. 367-370 (2007)

Effect of Cr Content on the Ductile-Brittle Transition Temperature of Irradiated Ferritic/martensitic Steels

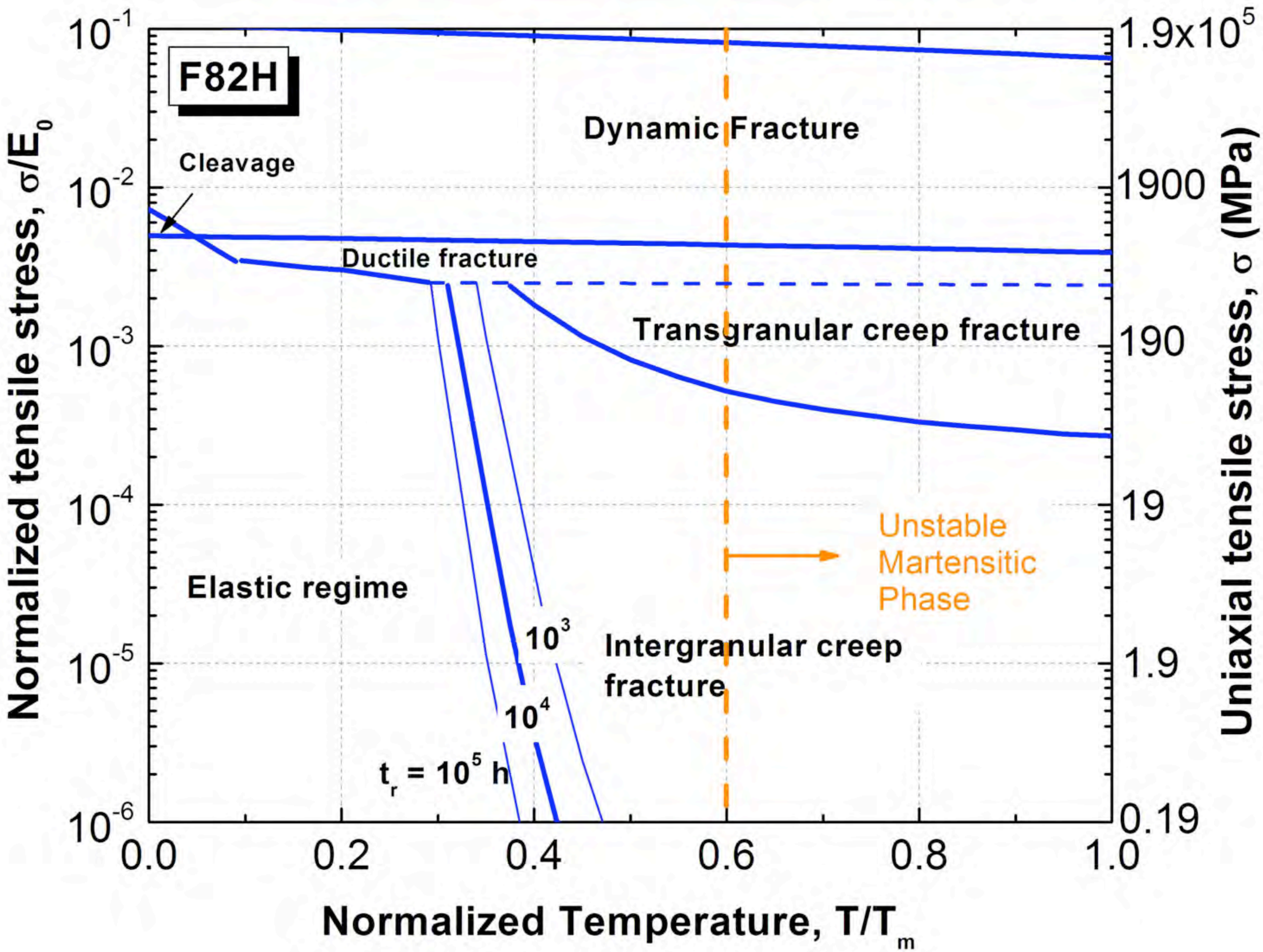


Caution: thermomechanical treatments not optimized!

Klueh and Harries 2001

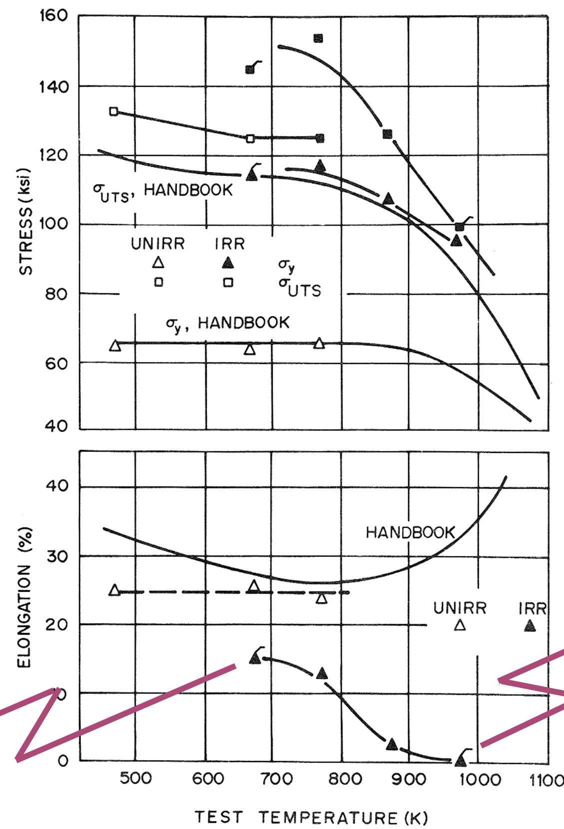


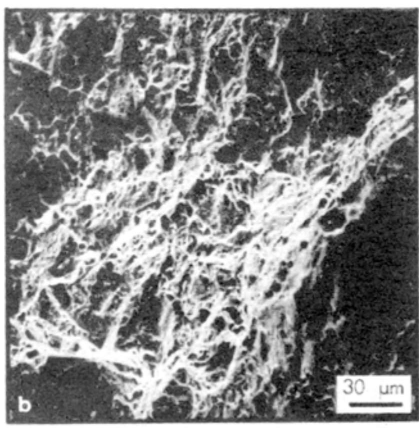
M. Li and S. Zinkle



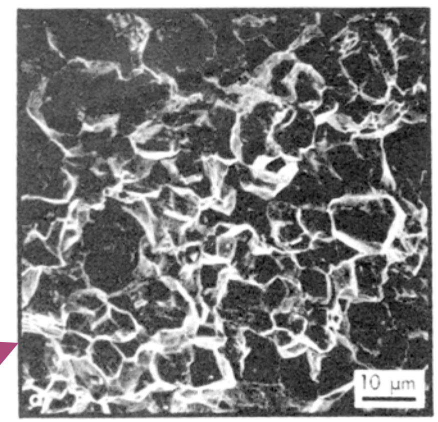
F82H
Dynamic Fracture
Cleavage
Ductile fracture
Transgranular creep fracture
Unstable
Martensitic
Phase
Elastic regime
Intergranular creep
fracture
10^3
10^4
$t_r = 10^5$ h
Normalized tensile stress, σ/E_0
Uniaxial tensile stress, σ (MPa)
Normalized Temperature, T/T_m
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
$1.9x10^5$
1900
190
19
1.9
0.19
0.0
0.2
0.4
0.6
0.8
1.0

Ni Base Alloys Have Excellent High Temperature Strength Properties But Are Highly Susceptible to Ground Boundary Embrittlemen





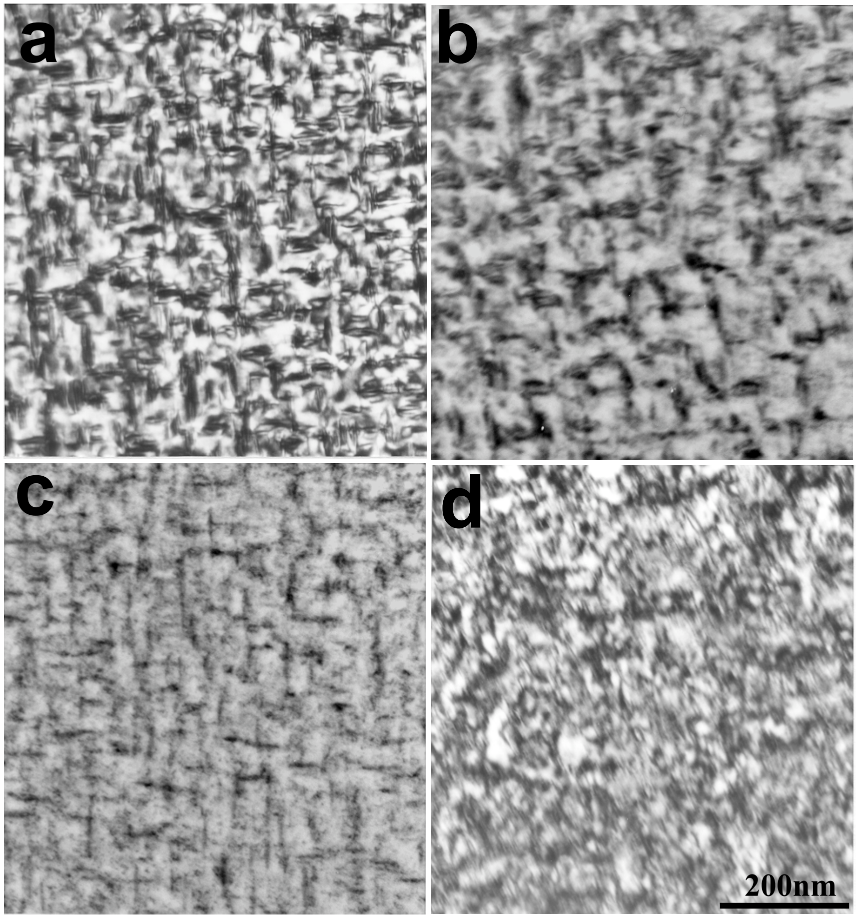
Fracture is transgranular

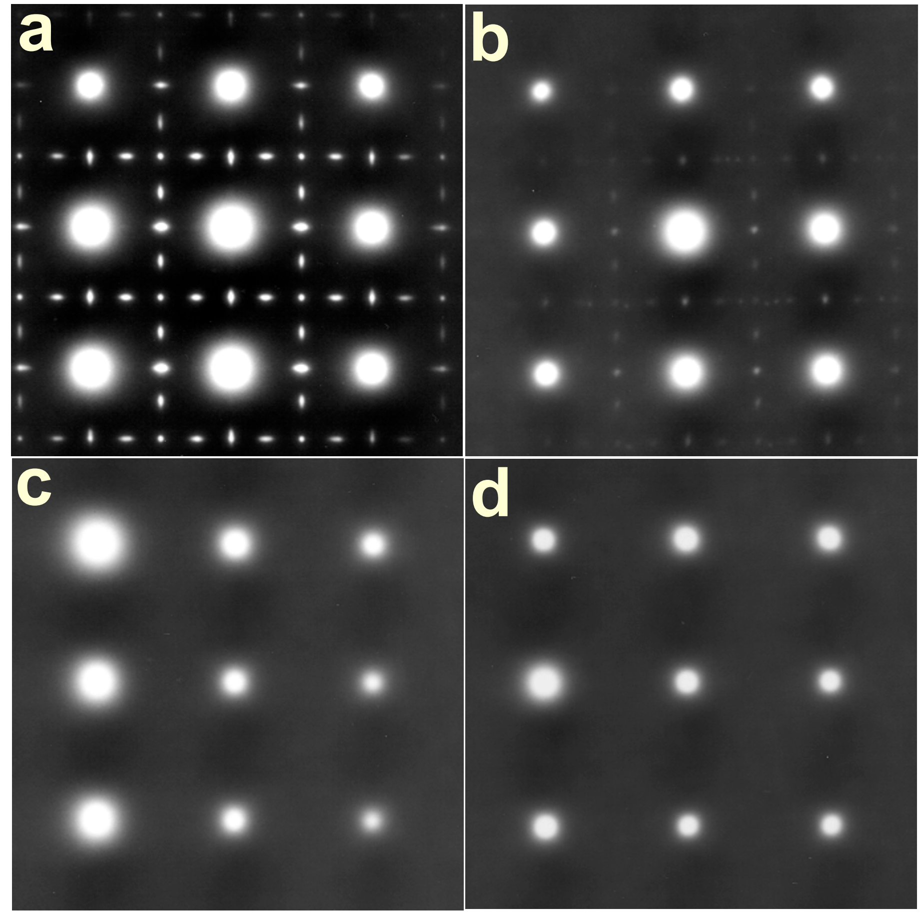


Fracture is Highly Intergranular

Tensile properties of Nimonic PE-16 irradiated at 500 C to ~6 dpa). Heat treatment: 4 h at 1040C + 1 hr at 900 C + 8 hr at 750 C.
(Flags signify 2 h @ 1040 C + 16 hr at 700 C.)

Precipitates in Inconel 718 dissolve during irradiation





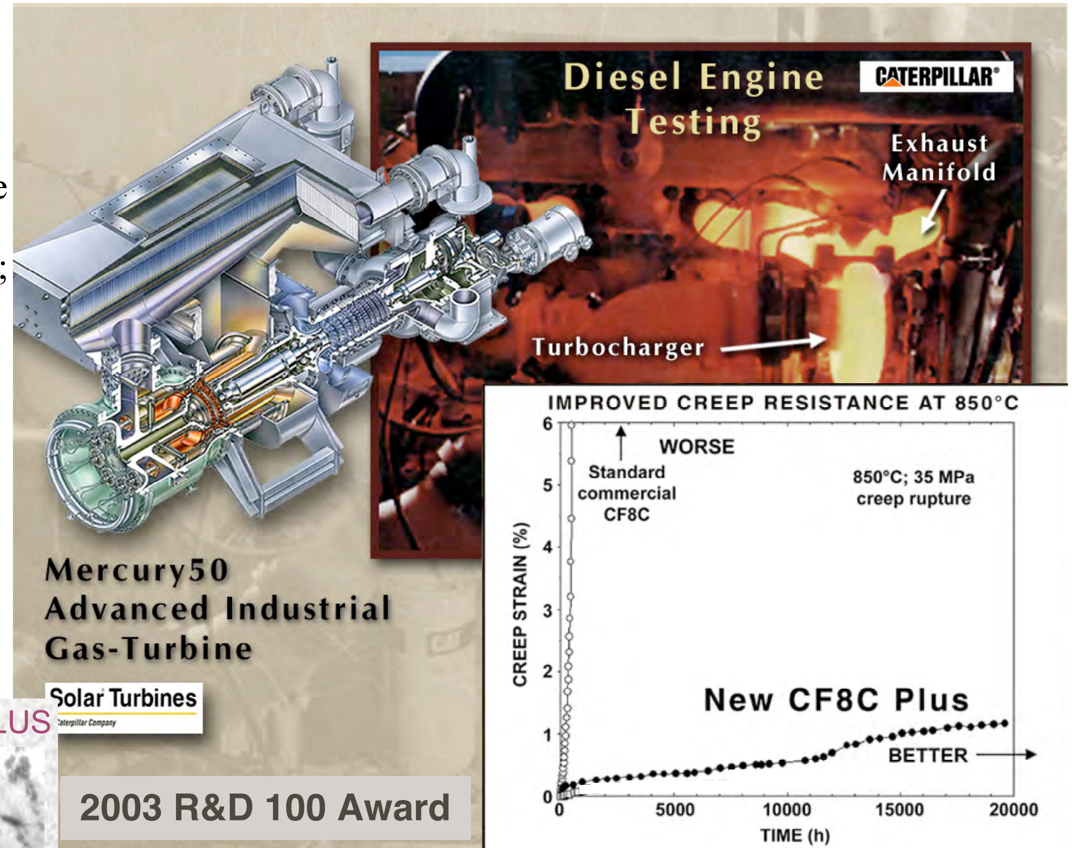
Inconel 718 solution annealed at 1065C/30min and aged 750C/10h+ 650C/20h: (a) pristine, and 3500 keV Fe-ion irradiated at 200 C to (b) 0.1 dpa, (c) 1 dpa, and (d) 10 dpa.

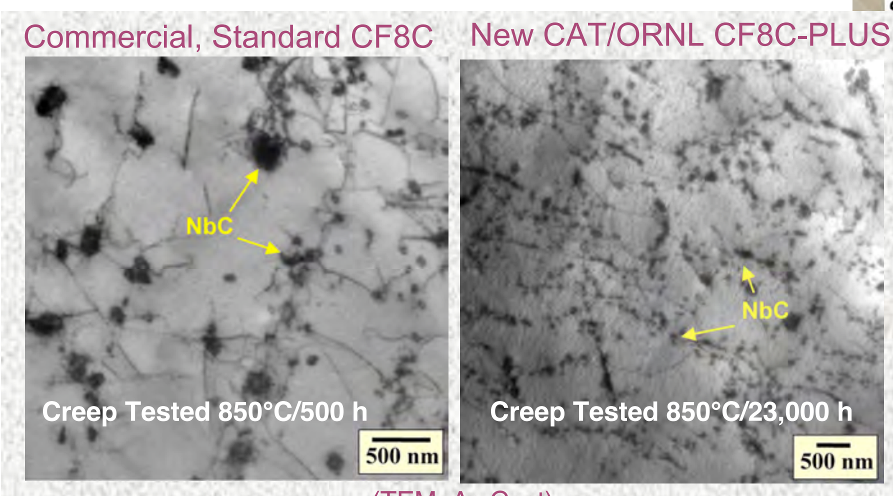
Overview of Improved Steels

- Steels can exhibit a wide range of properties depending on detailed composition and thermomechanical treatment
- Four generations of ferritic steels based on materials science principles have been commercialized (R. Viswanathan, Adv. Mater. & Processes 162, No. 8 (2004) 73)
  - 1st generation 1960-1970; 2nd generation 1970-1985; 3rd generation 1985-1995; 4th generation currently emerging
- Fusion 9Cr ferritic/martensitic steels are based on "2nd Generation" steels developed around 1985; fusion steels are comparable to 3rd generation commercial steels
  - Fusion substitution of W for Mo (reduced activation) was also pursued for "3rd generation" commercial steels
- Future steel development options will likely be based on evolutionary (ingot metallurgy/ classical precipitation) and revolutionary (nanoscale oxide dispersion strengthening) approaches

Microstructural Evolution In Irradiated Stainless Steels Provided the Key for Developing Improved High Temperature Alloys

- ***Reactant Effects*** (ie. Ti, V+Nb enhance MC formation)
- ***Catalytic Effects*** (ie. Si enhances Fe_2Mo or M_6C)
- ***Inhibitor Effects*** (ie. C, P or B retard the formation of Fe_2Mo or FeCr sigma phase during aging, G-phase during irradiation)
- ***Interference Effects*** (ie. N forms TiN instead of TiC; N does not form NbN instead of NbC; therefore C and N can be added with Nb, but not Ti)





(TEM, As Cast)

Result of microstructural modification:

- Formation of stable nanoscale MC carbide dispersions to pin dislocations
- Resistance to creep cavitation and embrittling grain boundary phases (ie. sigma, Laves)
- Resistance to dislocation recovery/ recrystallization

P.J. Maziasz

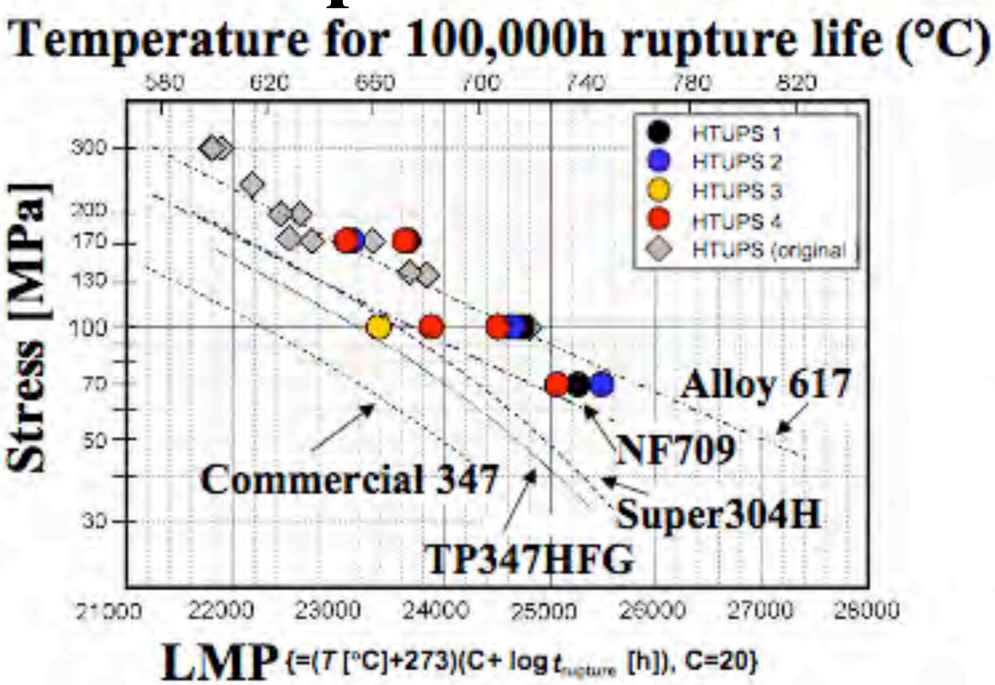
Technology Transfer of CF8C-Plus Cast Stainless Steel

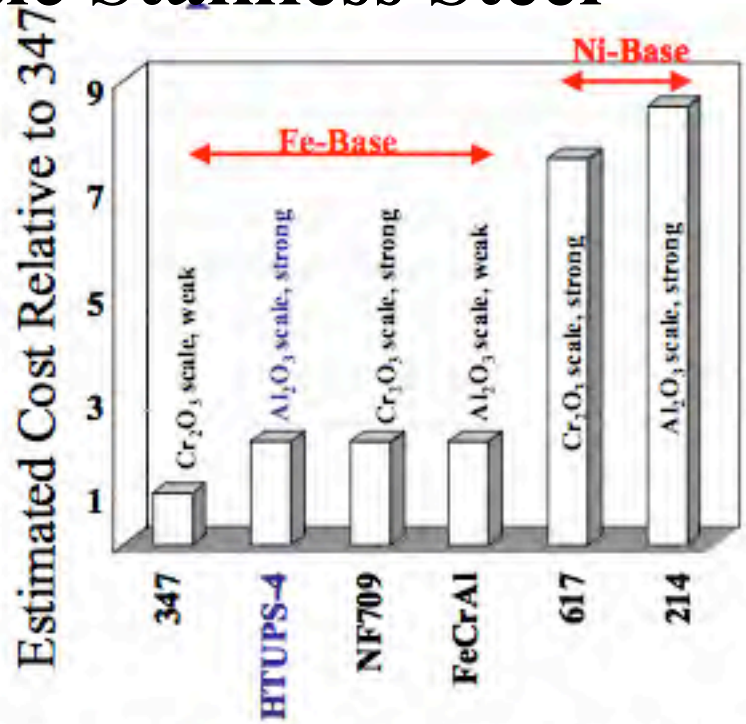
- MetalTek International, Stainless Foundry & Engineering, and Wollaston Alloys have trial licenses in 2005
- MetalTek has cast over 31,000 lb of CF8C-Plus steel through 2005 for Solar Turbines (end-cover, casings), Siemens-Westinghouse (large section tests for turbine casings), ORNL, and a global petrochemical company (tubes/piping).
- Stainless Foundry has cast CF8C-Plus exhaust components for Waukesha Engine Dresser NG engines

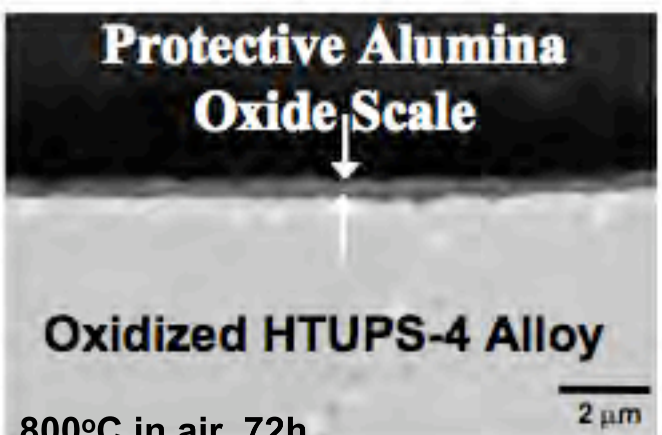
6,700 lb **CF8C-Plus** end-cover cast by MetalTek for Solar Turbines Mercury 50 gas turbine

80 lb **CF8C-Plus** exhaust component cast by Stainless Foundry for Waukesha NG reciprocating engine

Development of New Alumina-Forming, Creep Resistant Austenitic Stainless Steel





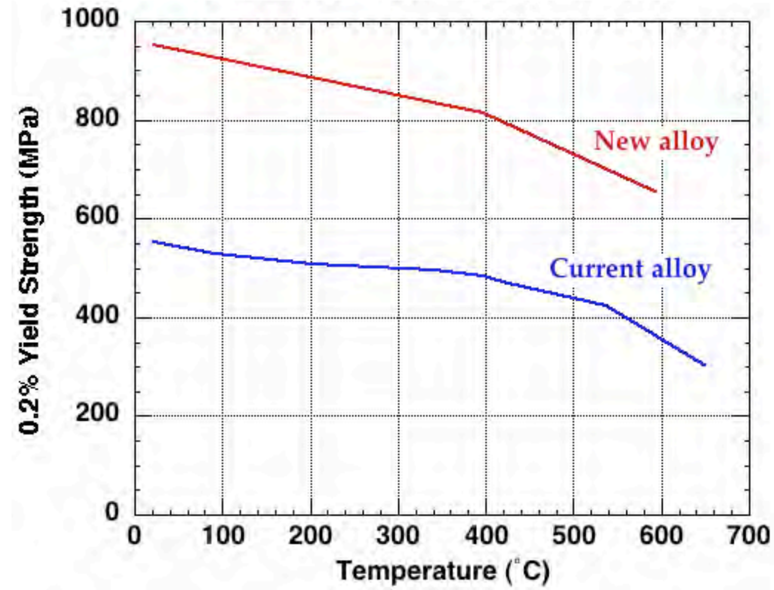


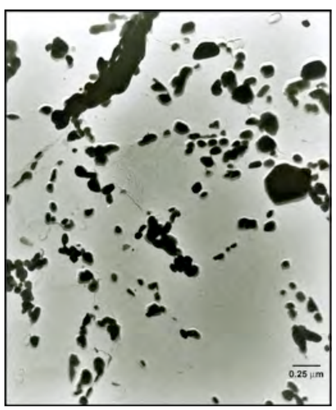
- Designed for 600-800°C structural use under aggressive oxidizing conditions
  - -superior oxidation resistance to conventional chromia-forming alloys
- Comparable cost to current heat-resistant austenitic stainless steels

Y. Yamamoto, M.P. Brady et al., Science 316 (2007) 433

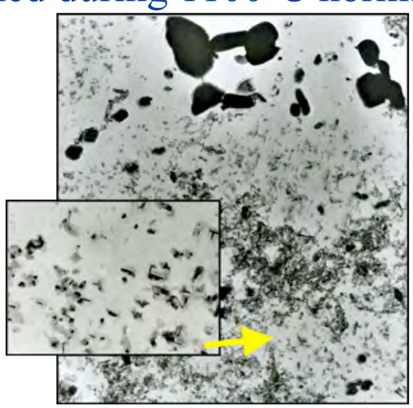
Creep Properties of New 3 Cr Steels have Advantages Over Existing 2 1/4Cr and 9-12Cr Steels

- Creep resistance improved over Japanese 2.25Cr (T23,T24) steels
- May not need to be tempered for high thermal creep strength; no postweld heat treatment?
- Properties better than HT9 and as good or better than modified 9Cr-1Mo steel
  - Long-term creep behavior (>5000 h) still needs to be determined
  - Two 50 ton heats procured by industry are being tested to obtain ASME code approval
  - Improved creep resistance is due to fine V-rich MC needles formed during 1100˚C normalizing

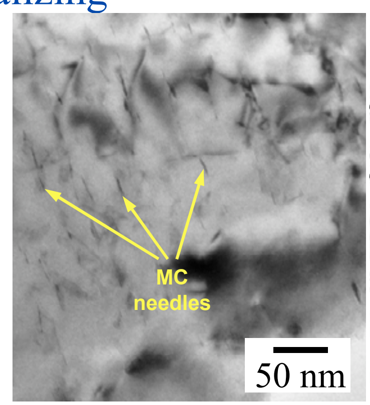




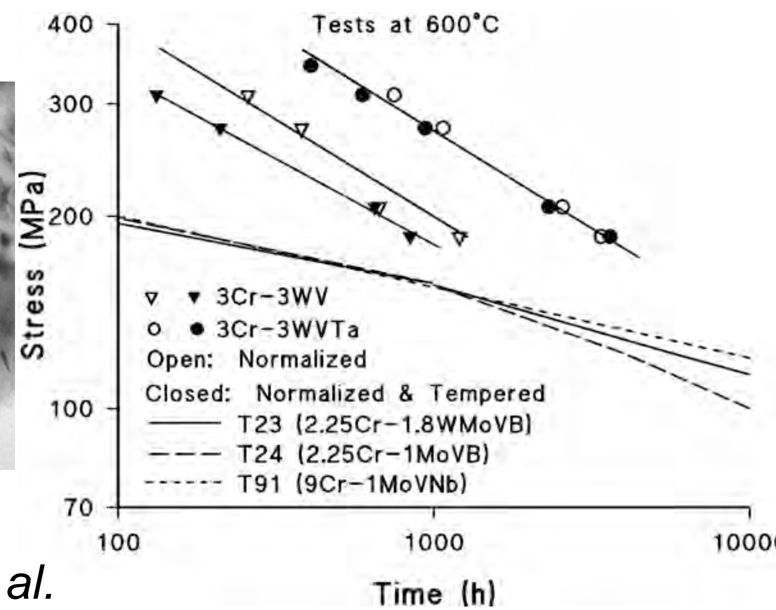
2 1/4 Cr-1Mo



New 2 1/4 Cr-2WV



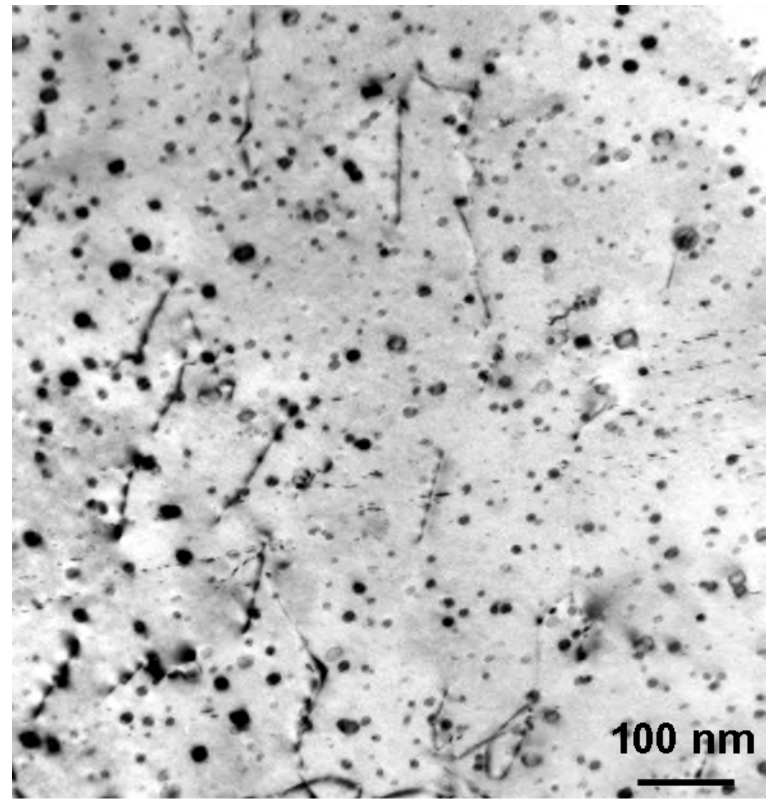
New 3 Cr-3WV



R.L. Klueh et al.

New Thermomechanical treatment (TMT) Process Applied to Commercial 9-12%Cr Steels Yields Improved Microstructure

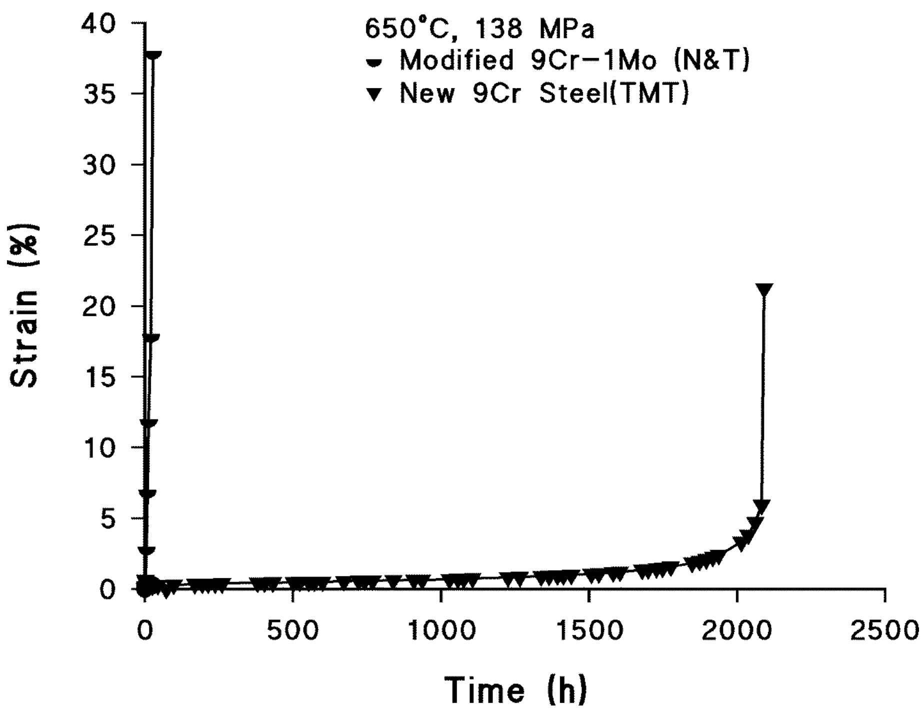
- Commercial 9Cr-1Mo and 12 Cr steels were processed
- TMT (hot rolling) on 25.4–mm plates
  - Several TMT conditions were investigated
- Precipitates formed on dislocations introduced by hot rolling
- Precipitate dispersion is much finer than observed in conventionally processed 9-12Cr steel

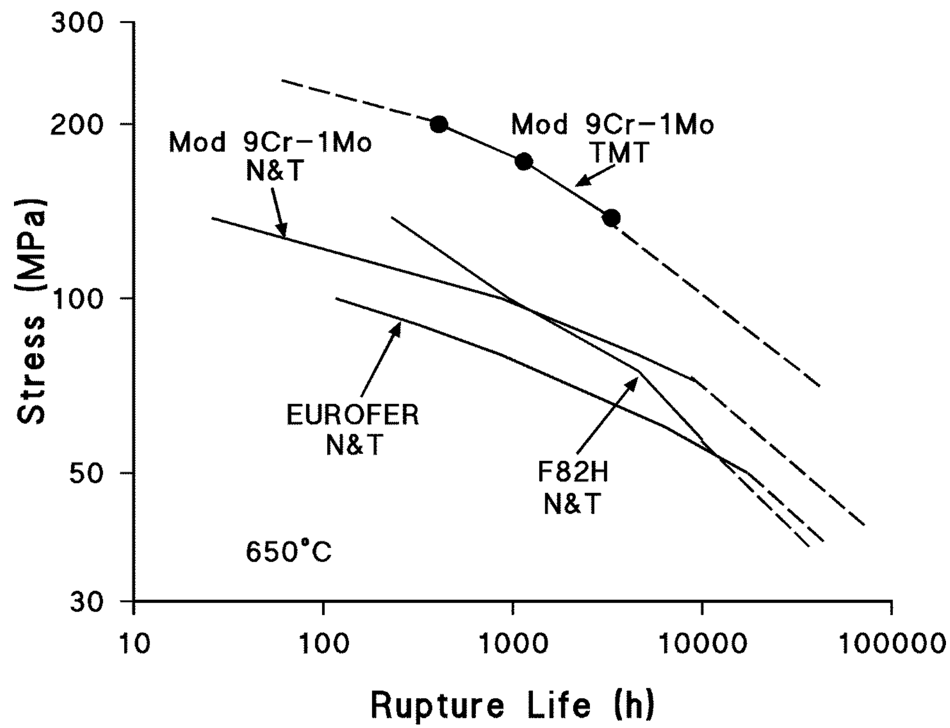


Modified 9Cr-1Mo—New TMT

R.L. Klueh et al., J. Nucl. Mater. *367-370 (2007) 48*

Large Increase in Rupture Life of Modified 9Cr-1Mo at 650°C





- **Thermo-mechanical treatment (TMT) of modified 9Cr-1Mo produced steel with over an order-of-magnitude increase in rupture life**

R.L. Klueh et al., J. Nucl. Mater. 367-370 (2007) 48

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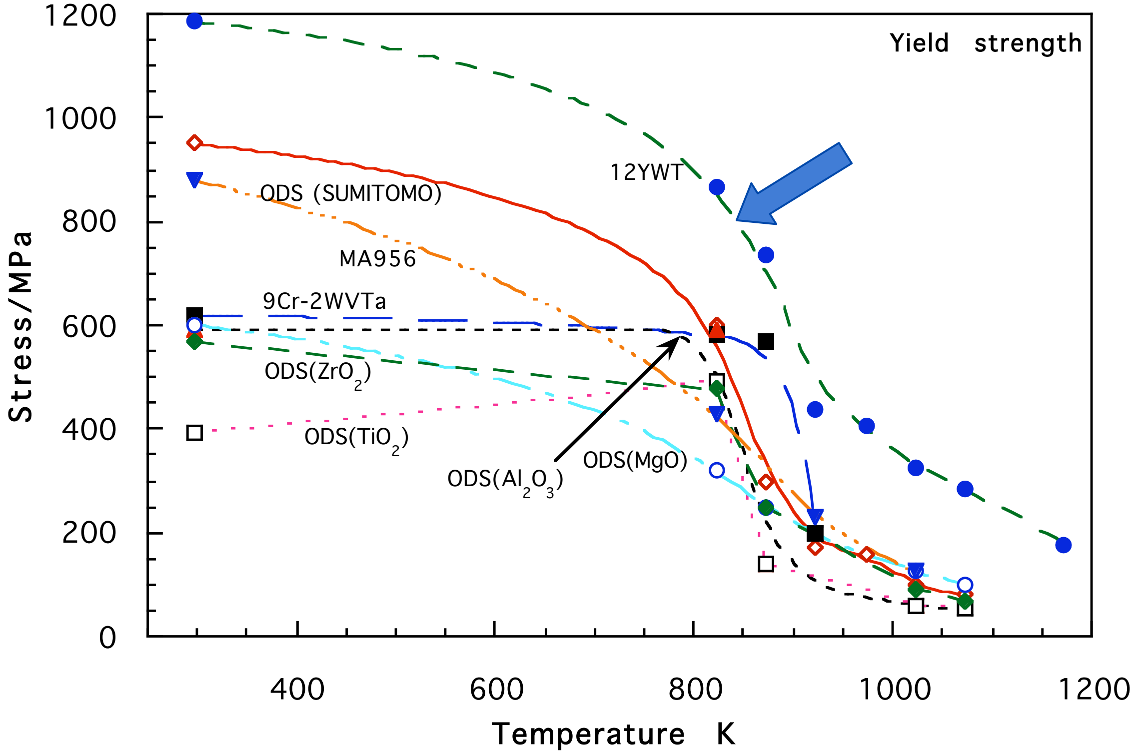
Oxide dispersion strengthened Steels

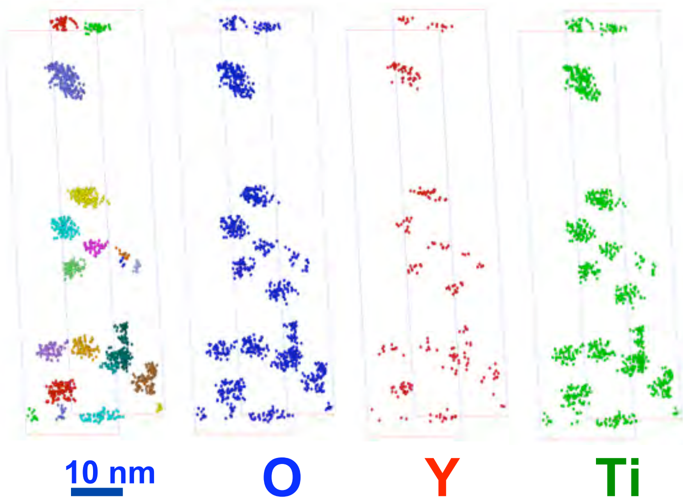
- There are two main options for ODS steels, based on pioneering work by Ukai and coworkers
  - Ferritic ODS steel (typically 12-16%Cr)
  - Ferritic/martensitic ODS steel (typically ~9%Cr)

Steel	Advantages	Disadvantages
12-16% ODS ferritic steel	Higher temperature capability Better oxidation resistance	Anisotropic mechanical properties Lower fracture toughness
9% ODS ferritic/martensitic steel	Nearly isotropic properties after heat treatment Better fracture toughness	Limited to temperature below ~700 C Marginal oxidation resistance at high temperatures

New 12YWT Nanocomposited Ferritic Steel has Superior Strength compared to conventional ODS steels



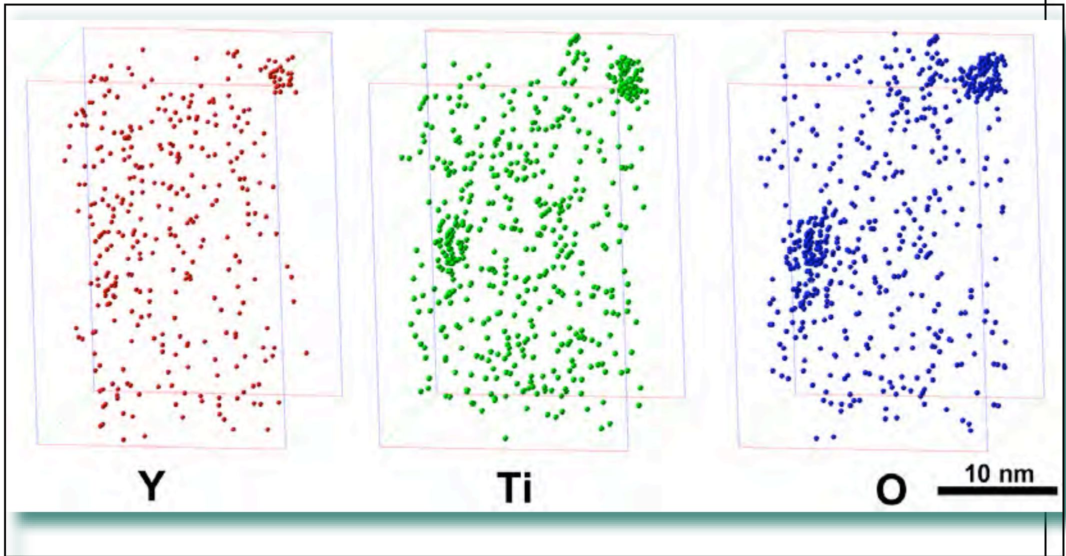
- Thermal creep time to failure is increased by several orders of magnitude at 800˚C compared to ferritic/martensitic steels
  - 2% deformation after ~2 years at 800°C, 140MPa
- Potential for increasing the upper operating temperature of iron based alloys by ~200°C
- Acceptable fracture toughness near room temperature

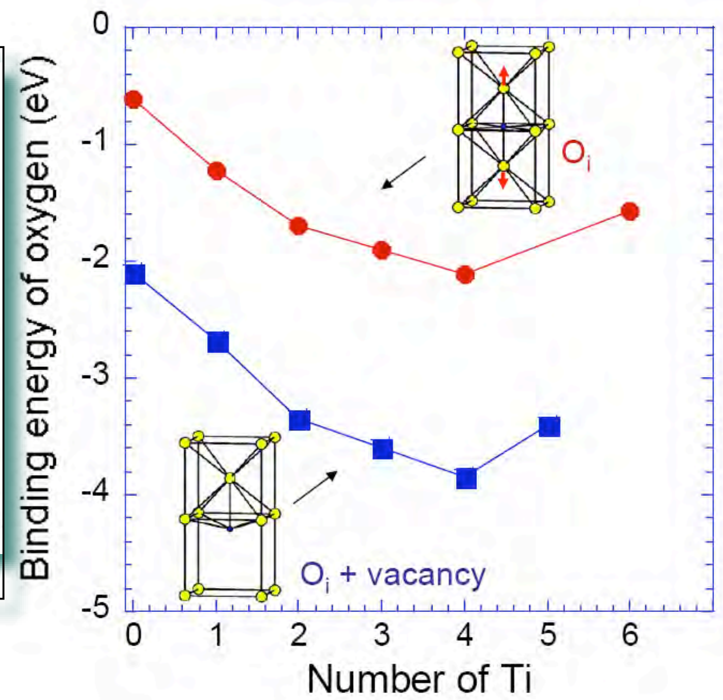


- Atom Probe reveals nanoscale clusters to be source of superior strength
  - Enriched in O(24 at%), Ti(20%), Y (9%)
  - Size : $r_g = 2.0 \pm 0.8$ nm
  - Number Density : $n_v = 1.4 \times 10^{24}/m^3$
- Original Y_2O_3 particles convert to thermally stable nanoscale (Ti,Y,Cr,O) particles during processing
- Nanoclusters not present in ODS Fe-13Cr + $0.25Y_2O_3$ alloy

D.T. Hoelzer et al.

Theory Has Shown That Vacancies Play a Pivotal Role in the Formation and Stability of Nanoclusters





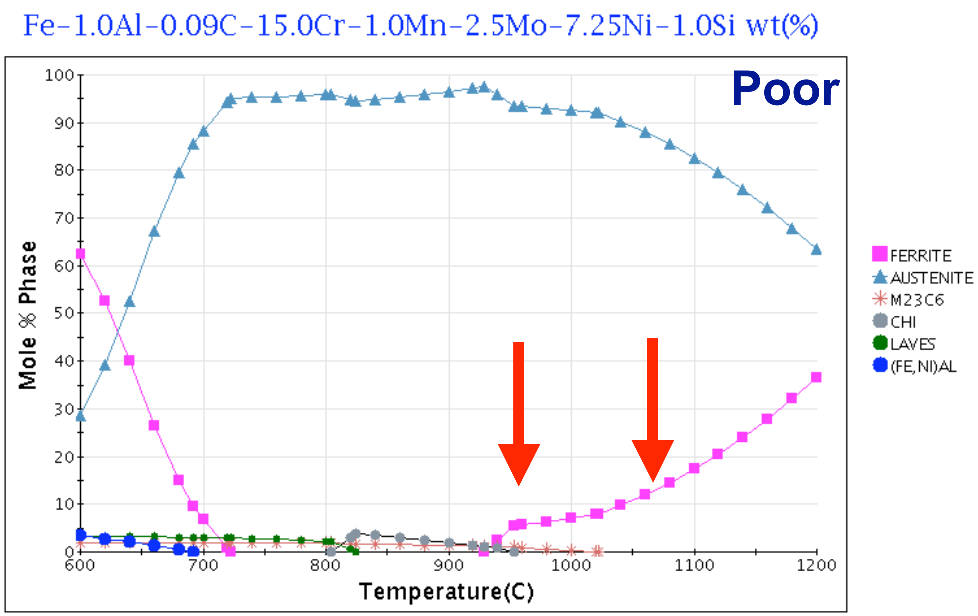
The presence of Ti and vacancy has a drastic effect in lowering the binding energy of oxygen in Fe (by C. L. Fu and M. Krcmar).

Conclusions

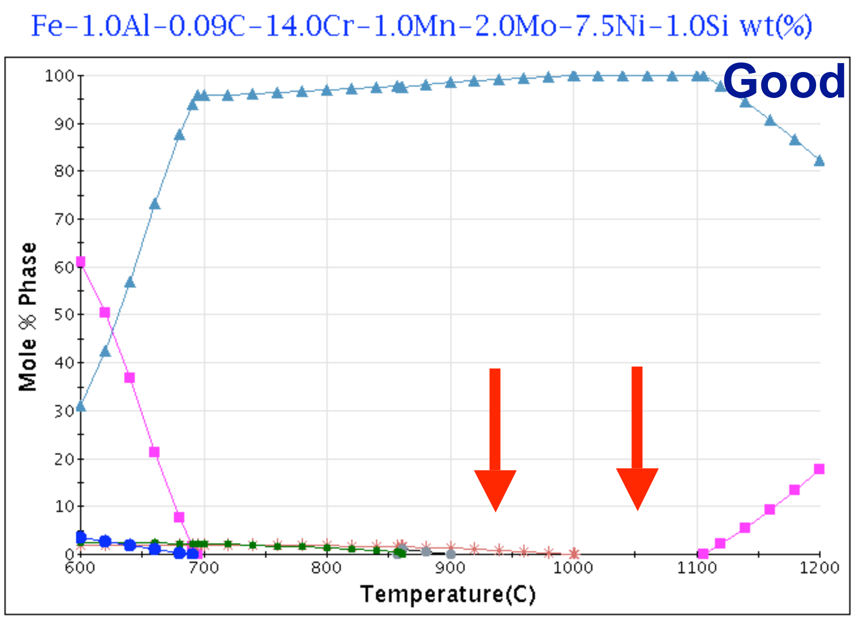
- **Existing structural materials face Gen IV reactor design challenges due to limited operating temperature windows**
  - **May produce technically viable design, but not with desired optimal economic attractiveness**
- **Substantial improvement in the performance of structural materials can be achieved in a timely manner with a science-based approach**
- **Design of nanoscale features in structural materials confers improved mechanical strength and radiation resistance**
  - **Such nanoscale alloy tailoring is vital for development of radiation-resistant structural materials for advanced fission reactors**
- **Continued utilization of modern materials science techniques would be valuable to uncover new phenomena associated with localized corrosion, ultra-high toughness ceramic composites, ultra-high strength alloys, etc.**

Modern computational thermodynamics reveals pathway to improve precipitation hardened stainless steels

Both 15%Cr-7%Ni alloys are within allowable chemical composition



"average" Cr, Mo, Ni



"low" Cr, Mo and "high" Ni

- Within alloy specifications, large differences can be expected with standard heat treatment
- Calculations can lead to composition and heat treatment optimization

J.M. Vitek

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