Materials Issues for High Power Accelerators

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Materials Issues in High Power Accelerators--Not Limited to Target Radiation Damage

- **Introduction**
- **Radiation effects– the most pervasive issue**
  - Brief review
    - **Metallic alloys**
    - **Ceramics**
    - **Polymers**
  - Damage conditions and materials choices
- **Design-specific issues--examples**
  - Cavitation erosion in pulsed liquid metal targets
  - Irradiation assisted stress corrosion cracking
  - Beam stripper foils
  - Other challenging areas
- **Summary**

High Power Accelerator Facilities

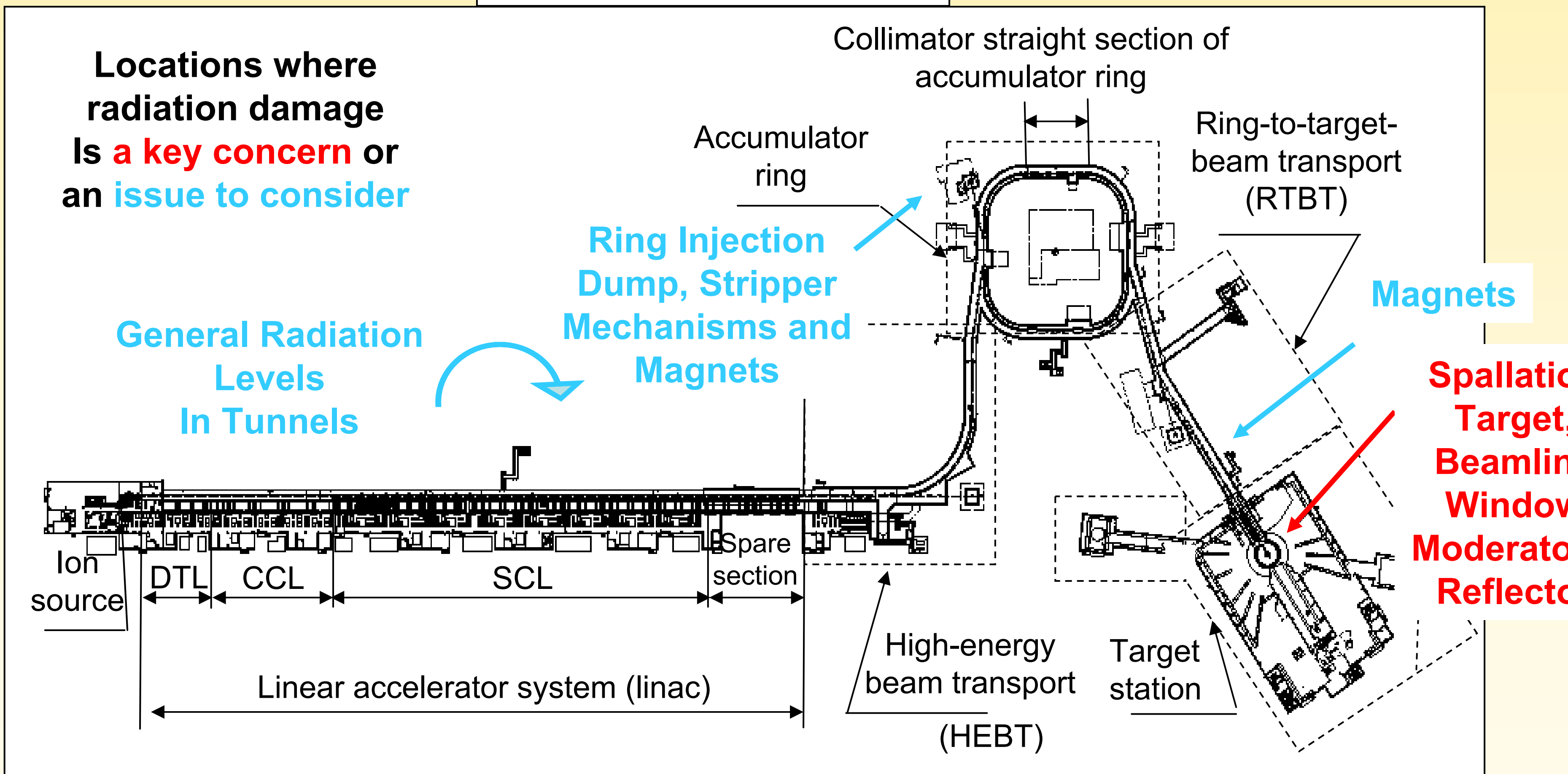
- **Spallation neutrons**
  - Neutron scattering
  - Transmutation of nuclear waste
  - Energy amplification
  - Isotope production
  - In operation: SINQ, LANSCE, ISIS, …
  - Under construction: SNS, J-PARC, …
- **Radioactive ion beams**
  - ISOL (proton)
  - Fragmentation (heavy-ion)
- **Particle physics**
  - Muon and neutrino production
  - …

Most Important of the Materials Questions for Large Accelerator Complexes

- **Where is R&D needed to reduce risk?**
  - Establish new concept viability
  - Select or develop materials that could fulfill intended function
  - Obtain sufficient information to estimate lifetime
  - Qualify materials for applications
    - requires more effort than above activities
    - need prototypical facilities or conditions
    - usual for fission power reactor and research reactor applications
    - not always possible for new accelerator types
- **Where are demands above experienced-based threshold or beyond conventional needs for high performance facilities?**
  - Conditions for which there are few or no data
  - Aggressiveness of service environment

Example--Spallation Neutron Source

Protons: 1 GeV, 2 ma



Radiation Doses for Key SNS Locations

Spallation target module peak displacement dose rate	~ 10^{-6} (10^{-2}) dpa/s (~ 2/3 n, 1/3 p)	Spallation target module peak displacement dose (1 y)	36 dpa
Ring injection dump peak dose rate	~ 2 x 10^{-7} dpa/s (~ 3/5 p, 2/5 n)	Ring injection dump peak dose (1 y)	7 dpa
RTBT second doublet coil peak ionizing dose rate	~ 4 x 10^{-2} Gy/s	RTBT second doublet coil peak ionizing dose (10 y)	10 MGy
Ring injection section magnet coil peak ionizing dose rate	~ 1.5 x 10^{-2} Gy/s (1/5 n, 4/5 p)	Ring injection section magnet peak ionizing dose (10 y)	5 MGy
Coupler peak displ. dose rate Peak Ionizing dose rate	~ 2 x 10^{-16} dpa/s (~ 3/4 n, 1/4 p) ~ 3 x 10^{-4} Gy/s	Coupler peak displ. dose (10 y) peak ionizing dose (10 y)	 6.3 x 10^{-8} dpa 95 kGy

Historical Perspective on Radiation Effects

- **Some radiation effects were observed in minerals in the 19th century, but their origin was not understood**
- **E. P. Wigner, 1946, Journal of Applied Physics 17**

  "The matter has great scientific interest because pile irradiation should permit the artificial formation of displacements in definite numbers and a study of the effect of these on thermal and electrical conductivity, tensile strength, ductility, etc. as demanded by the theory."
- **The full scope of radiation effects in materials was only appreciated after high neutron flux fast spectrum reactors were operated in the 1950's and 1960's**

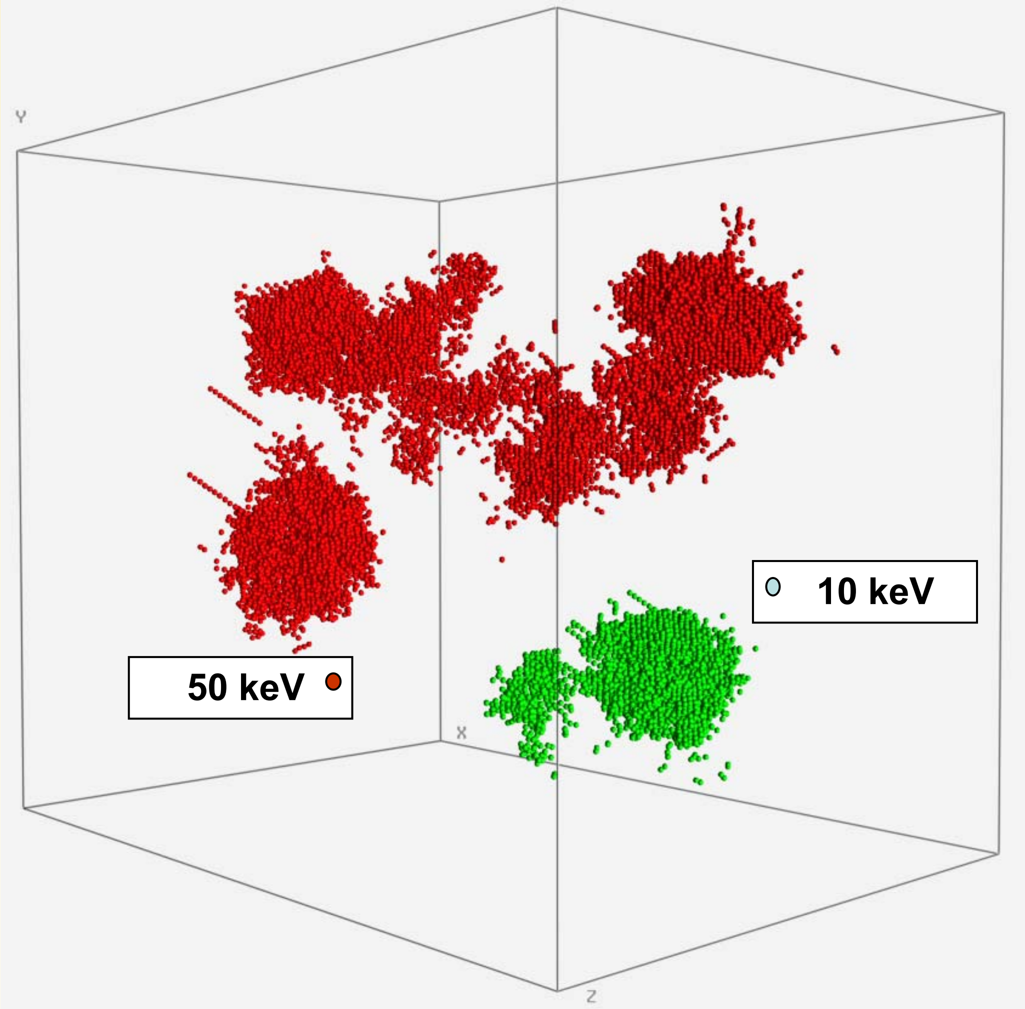
Radiation Effects in Materials

- Virtually every property of materials can be changed by irradiation
  - Dimensions
  - Mechanical properties (strength, ductility,…)
  - Physical properties (electrical, optical, thermal…)
  - • • •
- Underlying these changes are the production of defects and defect clusters, alterations in structure on a nanoscale (e.g., dislocations, voids, precipitates) compositional segregation, electronic ionization and excitation …
- Targets of high power accelerators experience roughly the same levels of damage as the highest flux fission reactor cores and first walls of future fusion reactors

Origins of Radiation Effects in Materials

- **Displacement of atoms (nuclear stopping)**
  - –Dominant damage process for metals
  - –Important for ceramics, semiconductors
  - –Could be significant for polymers (usually neglected)
  - –Dose unit--displacement per atom, *dpa*
  - –One dpa is the dose at which on average every atom in the material has been energetically displaced once
- **Ionization and excitation (electronic stopping)**
  - –Generally can be neglected for metals
  - –Important for polymers
  - –Can be important for ceramics, semiconductors
  - –Dose unit--Gray, *Gy*, the dose for absorption of 1 J/Kg

Displacement Damage Occurs in Cascades



- High energy particles, e.g., GeV protons may produce atomic recoils at much higher energies than fission neutrons
- Large-scale atomic simulations demonstrate that subcascade formation leads to similar defect production

Molecular Dynamics Simulations of peak damage state in iron cascades at 100 K
R. E. Stoller, ORNL

Origins of Radiation Effects in Materials

- **Transmutation reactions**
  - –Transmutation products, especially He and H from proton- and neutron-induced reactions, exacerbate damage
  - –Customary unit of measure is appm transmutant per dpa, e.g., *appm He/dpa*
- **Typical highest damage rates--10^{-6} dpa/s, $>10^{3}$ Gy/s**
  - –High power accelerator target 100 appm He/dpa
  - –High flux reactor core 0.2 appm He/dpa
  - –Fusion reactor first wall 15 appm He/dpa

Hierarchy of Reactions Leading to Property Changes in Metallic Alloys

Displacement of Atoms

Diffusion and Aggregation of Defects

Evolution of Microstructure

Embrittlement, Swelling, Irradiation Creep

What is it? Why is it important?

Time and Energy Scales for Radiation Effects by Displacement Damage

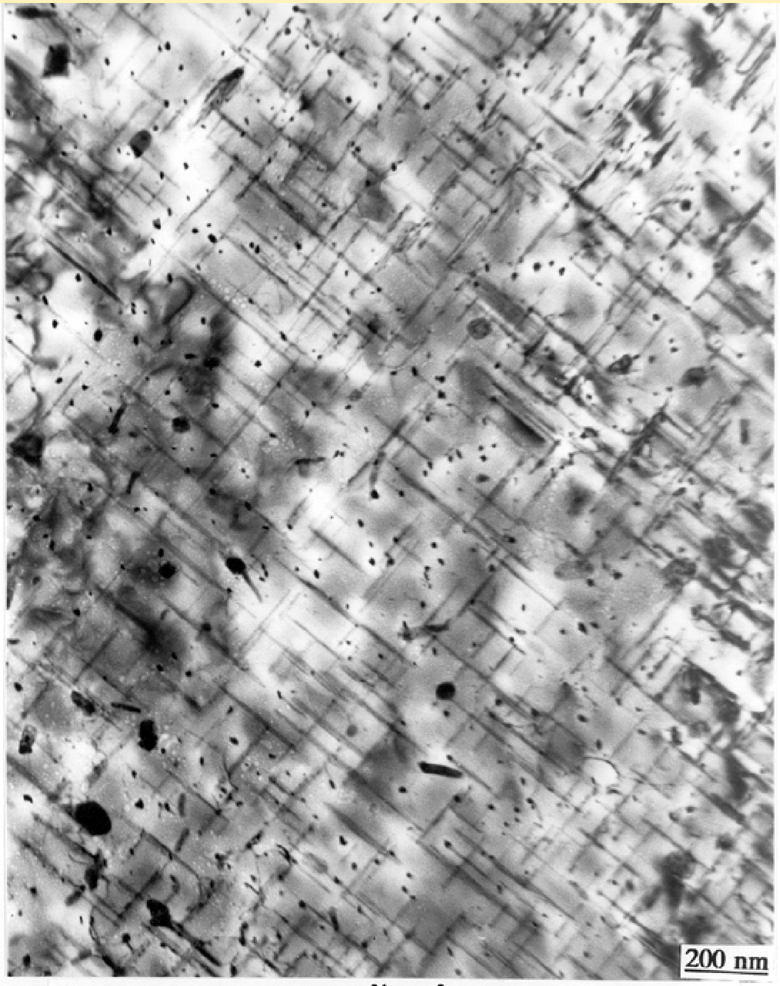
Time	Energy
Cascade Creation 10^{-13} s	Neutron or Proton 10^{5} - 10^{9} eV
Unstable Matrix 10^{-11} s	Primary Knock-on Atom 10^{4} - 10^{5} eV
Interstitial Diffusion 10^{-6} s	Displaced Secondary 10^{2} - 10^{3} eV
Vacancy Diffusion 10^{0} s	Unstable Matrix 10^{0} eV
Microstructural Evolution 10^{6} s	Thermal Diffusion kT

Radiation-induced Swelling

- Volume increase accounted for by a distribution of nanoscale cavities
- Interstitials absorbed at dislocations; vacancies absorbed at cavities
- Tens of percent at tens of dpa in structural alloys not designed to be swelling-resistant
- Theory and critical experiments have led to knowledge of mechanisms

Low Swelling Alloys Have Been Designed by Combining Theory with Critical Experiments

SA 316 irradiated to $8x10^{26}$ n/m^2 at 585 °C in EBR-II



A7 (Si, P) irradiated to $7x10^{26}$ n/m^2 at 555 °C in EBR-II

Importance of Swelling

- Significant concern between 0.3 and 0.6 T_m
- Overall dimensional increase of components
- Sensitivity to gradients in dose, dose rate and temperature can lead to distortions
- Fabricated geometries not preserved
- Cavity distributions possible easy paths for fracture
- May place limits on component lifetimes
- May affect particle transport and thermal hydraulics
- Not expected to be a problem in components operating < 0.3 T_m (e.g., SNS target)
- Could be a problem for higher temperature targets and beam dumps

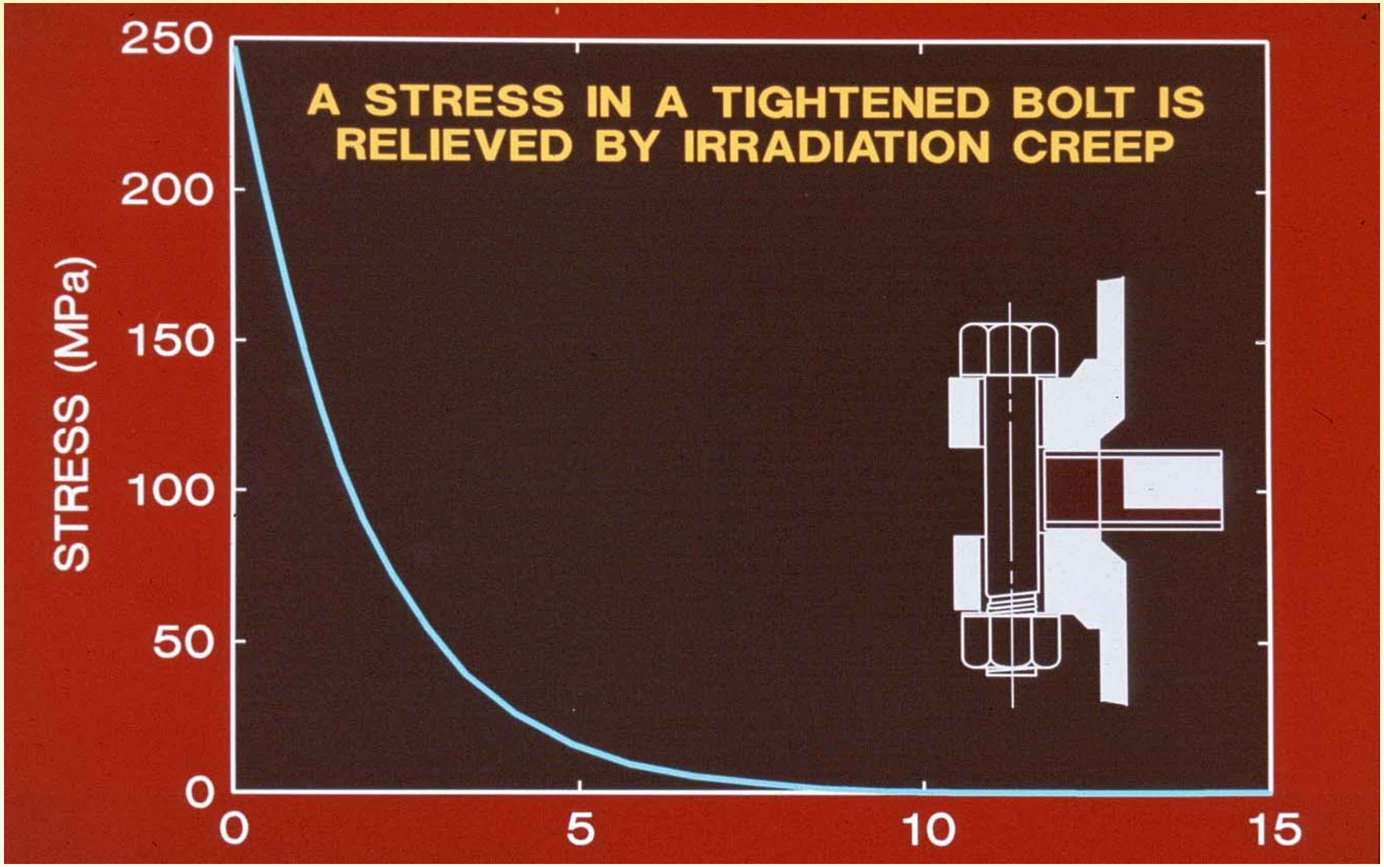
Radiation-induced Creep

- **Shape change in response to applied stress, or relaxation under constraint**
- **Vacancies and interstitials partition asymmetrically**
  - to differently oriented dislocations
  - between dislocations and other sinks for defects (cavities, grain boundaries, …)
  - because of short time unequal stochastic fluctuations in absorption of vacancies and interstitials
- **Occurs at all temperatures of interest**
- **At high temperatures, $T > 0.55\ T_m$, it is overwhelmed by thermal creep**

Radiation-Induced Creep

Two manifestations of the same phenomenon

- **Relaxation of stresses**
- **Continuing dimensional change**



Importance of Radiation-induced Creep

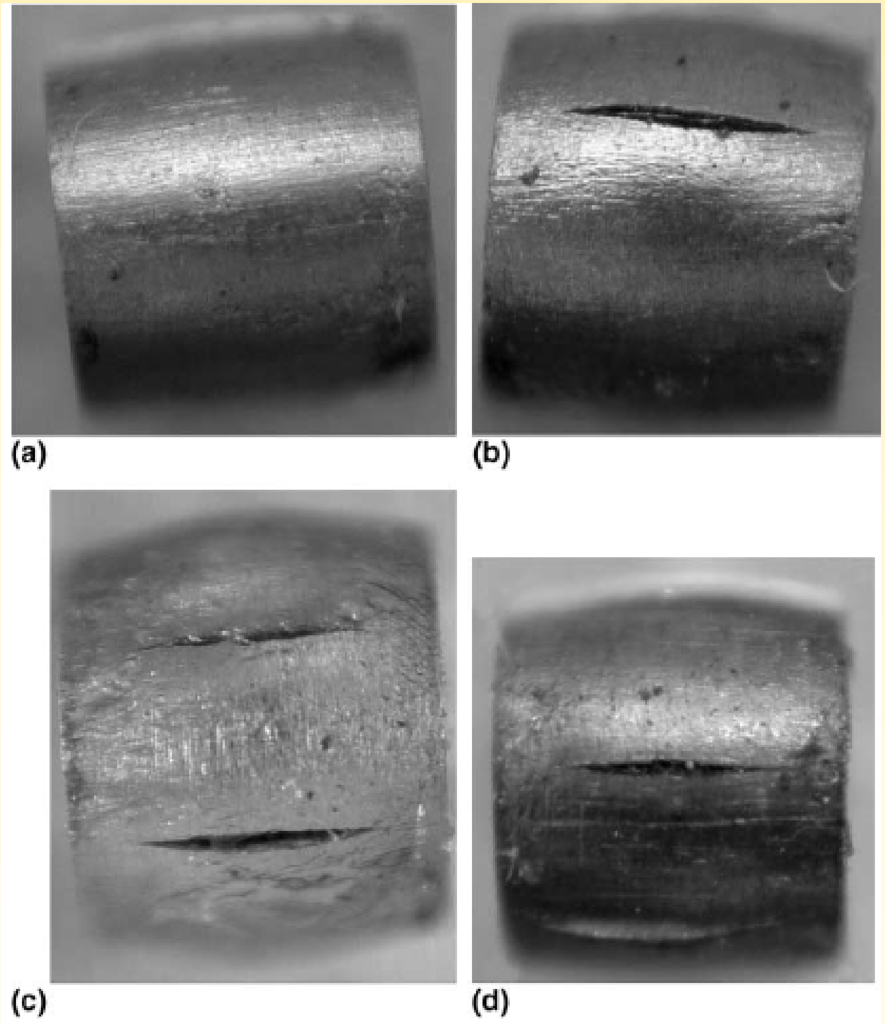
- Relaxation of engineered stress distributions
- Dimensional instability in shapes and sizes--linear dimension changes of several percent at high doses
- May be beneficial in relaxing stresses produced by radiation-induced swelling
- Significant problem for tight tolerance geometries, e.g., in fast neutron spectrum reactor cores
- Could affect particle transport and thermal hydraulics
- Not expected to be a problem in liquid metal accelerator targets with open structure (e.g., SNS target)

Radiation-induced Embrittlement

- **Hardening and loss of ductility**
- **Caused by vacancy and interstitial clusters, dislocation loops, precipitates and cavities that restrict deformation by dislocation glide**
- **Simultaneous weakening of grain boundaries**
  - by radiation-induced solute segregation and precipitation at grain boundaries
  - by accumulation of transmutation products on grain boundaries, especially He from (n, α) reactions
- **Various forms of embrittlement can occur at all temperatures**
  - at low temperatures, embrittlement mainly by matrix hardening
  - at higher temperatures, embrittlement mainly by helium and solute segregation at grain boundaries

Failures Can be Caused by Embrittlement

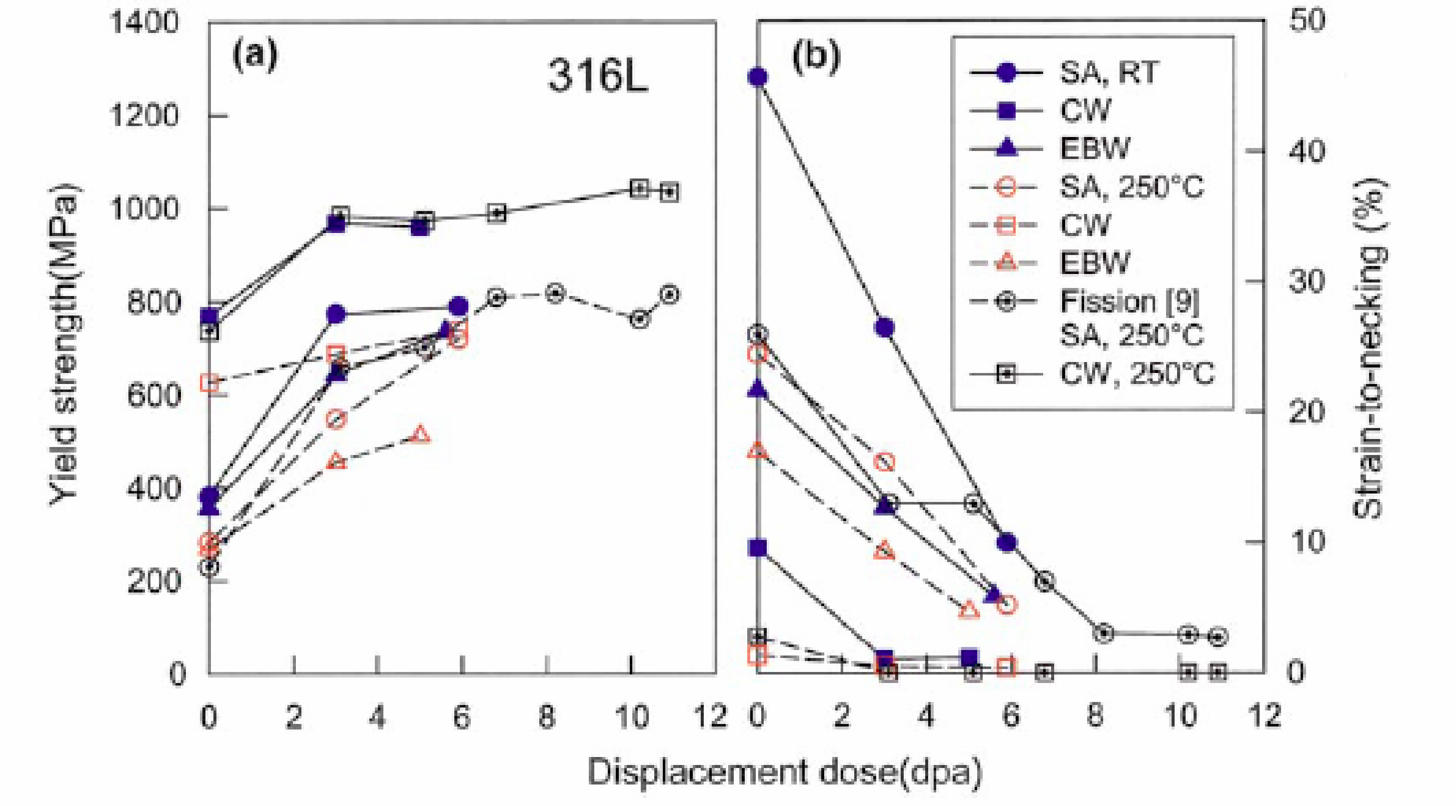
- Micrographs of tungsten compression specimens
- Irradiated with 800 MeV protons and compression tested to 20% strain at room temperature
- (a) before irradiation, (b) after 3.2 dpa, (c) after 14.9 dpa, and (d) after irradiation to 23.3 dpa.



S. A. Maloy, et al., J. Nucl. Mater., 2005
(LANSCE irradiations)

Irradiation-induced Hardening/Loss of Ductility

- Yield stress and strain-to-necking vs displacement dose for AISI 316L in solution annealed, 20% cold-worked and electron-beam welded conditions
- Filled and empty symbols--test temperatures of 25 and 250 º C, respectively
- Data from fission reactor irradiations ($T_{test} = T_{irrad}$ = 250 º C) are included

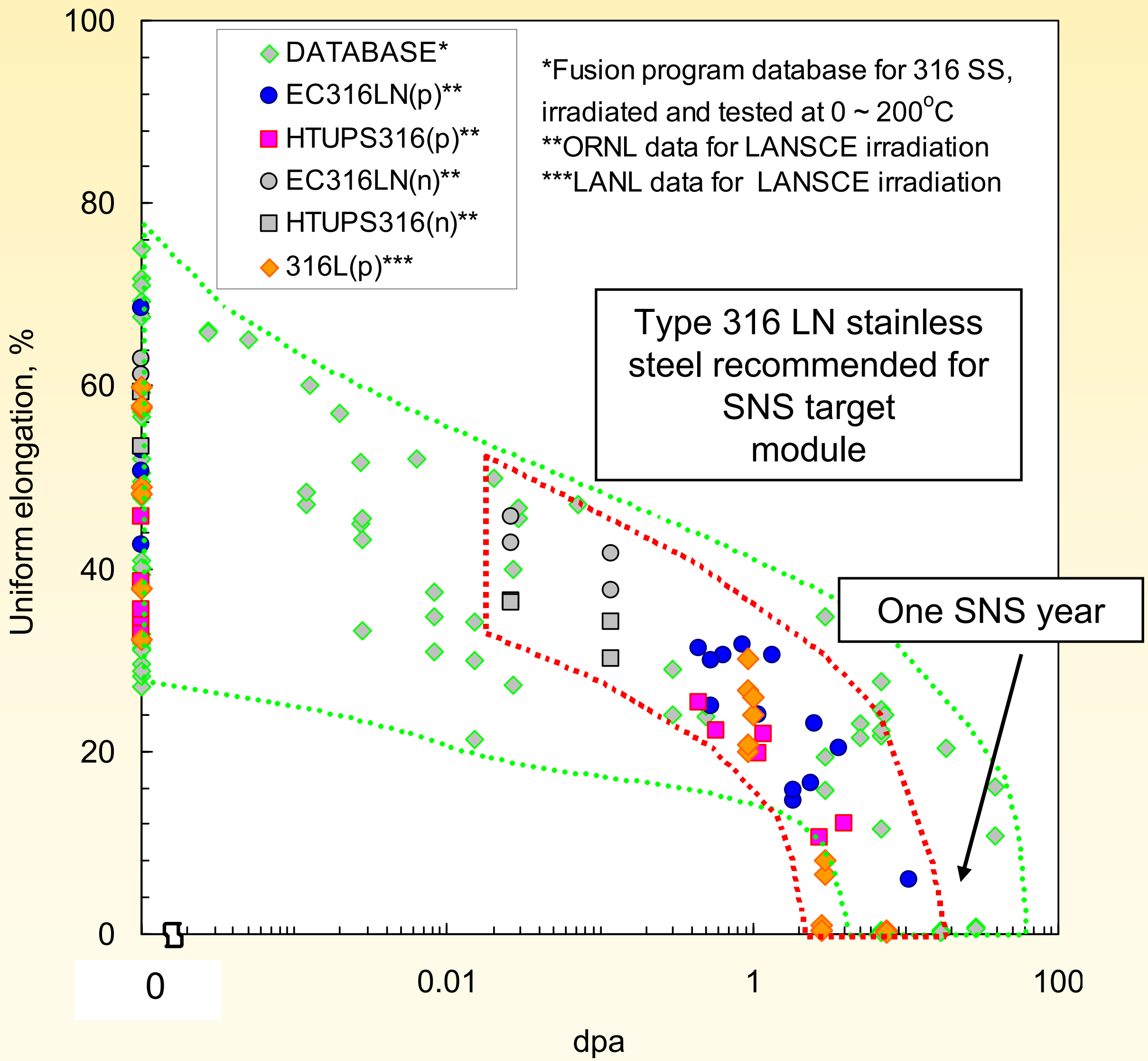


J. Chen, et al.,
J. Nucl. Mater. 2005
(SINQ irradiations)

Importance of Embrittlement

- Can lead to structural failure of components
- Possible crack formation and loss of vacuum or coolant integrity
- May necessitate early replacement of components
- Under fission reactor-like conditions component lifetimes can be tens of dpa
- Primary radiation effects issue for low temperature liquid metal target containers (e.g., SNS target)
- For higher temperature targets embrittlement must be considered together with other radiation effects

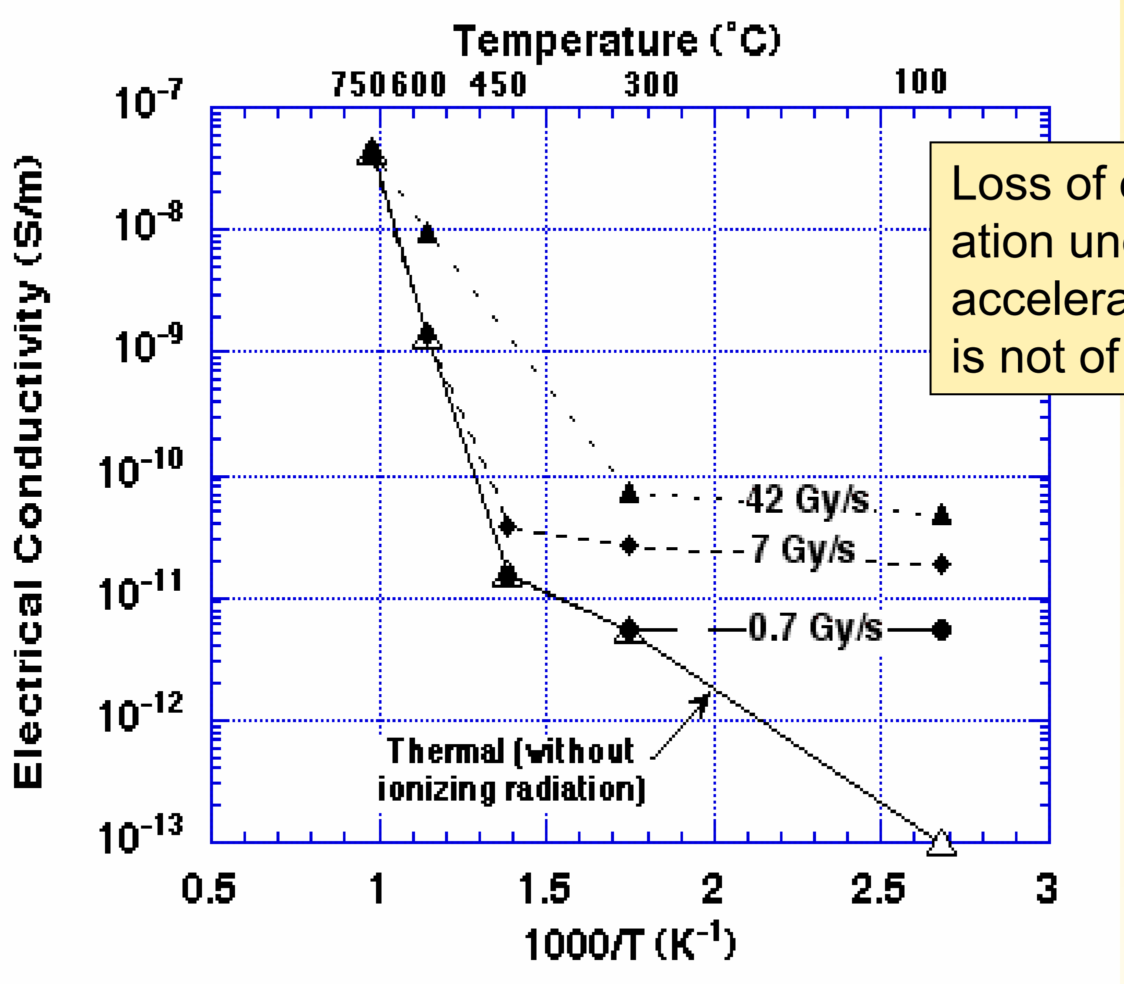
SNS Materials R&D on Ductility of Stainless Steels



Radiation Can Affect Ceramics through Three Types of Processes

- **Permanent defect production by knock-on collisions and nuclear reactions**
  - Displacement damage
  - Transmutations
- **Displacement production via ionization (radiolysis) processes**
  - Occurs in SiO_2, alkali halides, etc.
  - Does not occur in Al_2O_3, BeO, AlN
- **Radiation-induced conductivity (RIC)**
  - Transient excitation of valence electrons into conduction band

Electrical Conductivity in Fine Grained 99.99% Pure Alumina Cable (CR 125)

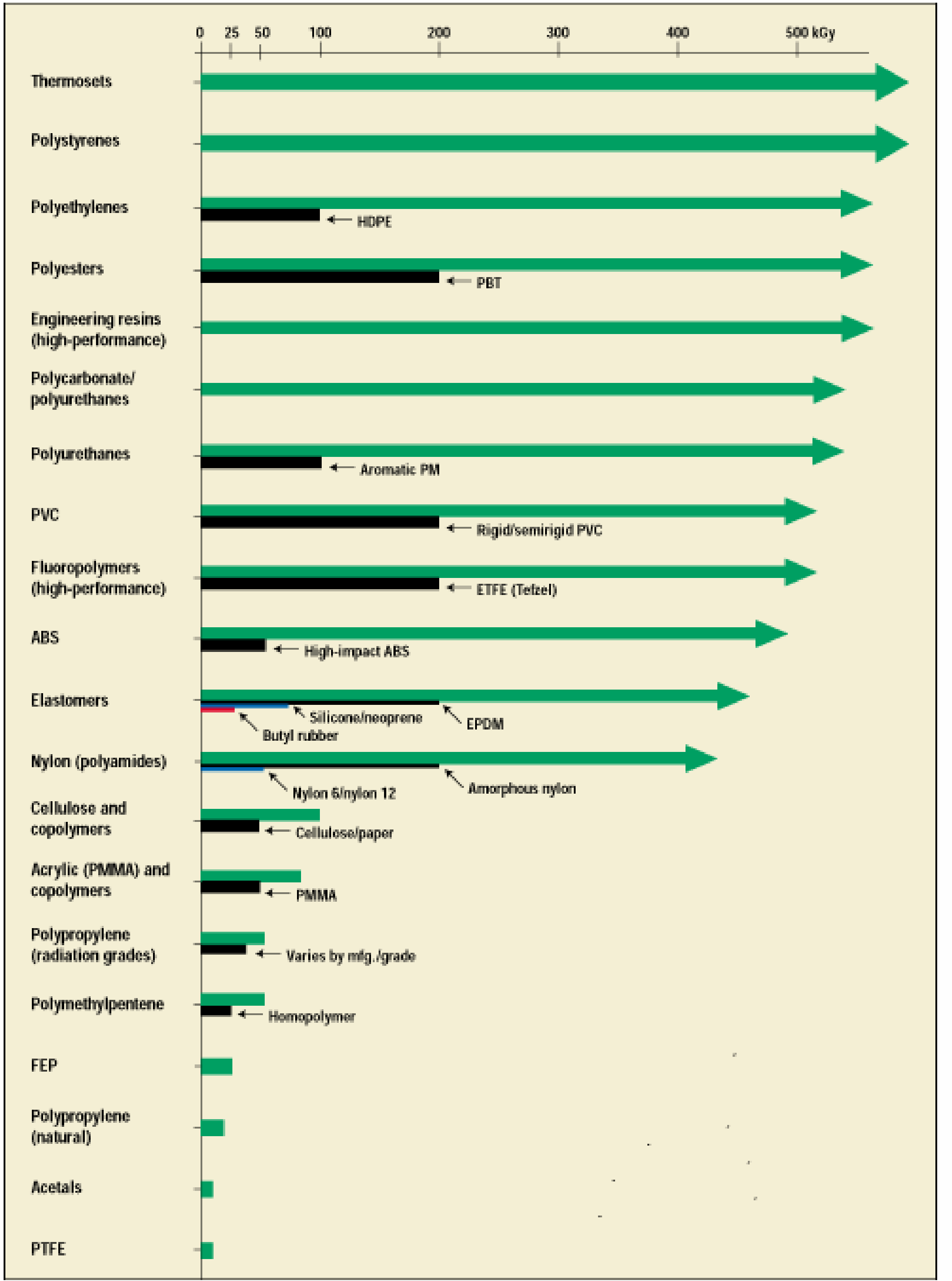


Loss of electrical insulation under typical accelerator conditions is not of concern

Basics of Radiation Effects on Polymers

- **Comparatively low doses can change properties**
  - Why? Typically very high molecular weight—therefore, a large fraction (tens of percent) of molecules can suffer at least one event in doses of order 10 kGy
- **Predominant changes are chain scission and cross-linking (other changes: release of small molecules, altering chemical compostion, i.e., gas formation; modification in types of bonding, …)**
- **For a given polymer, radiation type and temperature, either cross-linking or scission usually dominates**
- **Cross-linking increases molecular mass, lowers solubility and can improve mechanical properties**
- **Scission generally degrades properties**
- **Sensitivity depends on irradiation conditions and environment. Vacuum can improve dose endurance over air by an order of magnitude. Irradiation at higher T can give improvement.**

Mechanical Properties of Polymers (dose to reduce elongation by 25%)



K. J. Hemmerich, Med. Dev. & Diag. Ind. Magazine, Feb. 2000

Recommendations for Use of Polymers in Accelerator Components

- **Radiation effects in polymers become significant over the range from ~ 1 kGy to $\gtrsim 10^3$ kGy, depending on the material**
- **Acetal, polypropylene, and PTFE (teflon) should be avoided except for very low dose applications**
- **Top performers include PI (polyimide) and PS (polystyrene)**
- **High performance fluoropolymers like Viton are in an intermediate range.**
  - However, "Viton" is a general name for entirely different formulations. Specific data must be consulted

Examples of Design-Specific Materials Issues 1) Cavitation Erosion in Hg; 2) IASCC; 3) Beam Stripper Foils; 4)Corrosion in Pb-Bi

- **Cavitation erosion (pitting) in short pulse/high power/liquid Hg target**
  - Origin of effect
  - Potential lifetime-limiting problem for target
  - Research to characterize and mitigate damage
    - LANSCE accelerator (WNR facility) tests
    - Vibratory horn
    - High repetition pulse experiments
    - Surface carburization treatment
    - US, European, Japanese collaboration
- **This topic covered in other presentations**

Irradiation-Assisted Stress Corrosion Cracking

- For water-cooled stainless steel or nickel-based alloys in radiation fields, need to consider IASCC
- Damage based on irradiation embrittlement (above) may not be worst case for water-cooled structures
- Discuss dpa limits, fabrication and chemistry

  - First reported in Boiling Water Reactors (BWRs) in 1962
  - Observed in 300 series stainless steels and high nickel alloys
  - Earlier, components affected were either small (bolts, springs), or designed for replacement (control blades, instrumentation tubes)
  - Recently, more structurally significant components of reactor cores such as core shrouds have also been degraded

Some Facts About IASCC

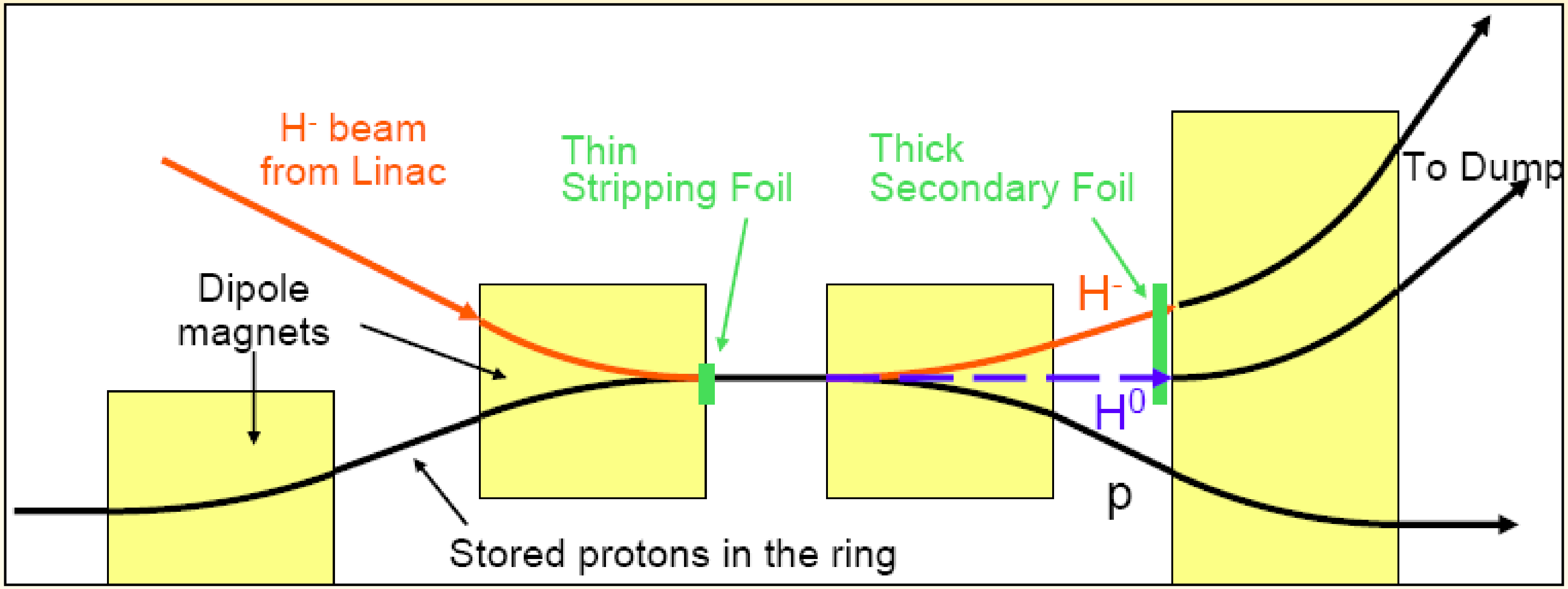
- An intergranular cracking phenomenon
- Requires displacement damage, water, stress
- Threshold in BWRs ~ 0.5 dpa
- Threshold in PWRs ~ several dpa
- Most available data in range 270-370 C
- Decreasing T may decrease prevalence; there is also evidence to the contrary
- Can be eliminated by controlling O to < 10 ppb, and/or H > 200 ppb
  - Too much H can also cause cracking
  - Crack tips can become acidic without added H

Recommendations on IASCC

- Avoid designs and fabrication that increase stress--e.g., unrelieved residual stresses, sharp corners, other stress raisers
- Avoid high strength alloys--e.g., CW materials
- Use strictly controlled weld procedures
- Consider control of water chemistry--addition of hydrogen/removal of radiolytic oxygen and removal of impurities (e.g., chlorides, sulfates)
- Restrict very long term water-cooled structures under significant stress to < few dpa; if no water chemistry control < 0.5 dpa
- Design for non-routine replacement of "permanent" structures as far as possible
  - 40 years is too long for irrevocable decisions in a complex irradiation environment

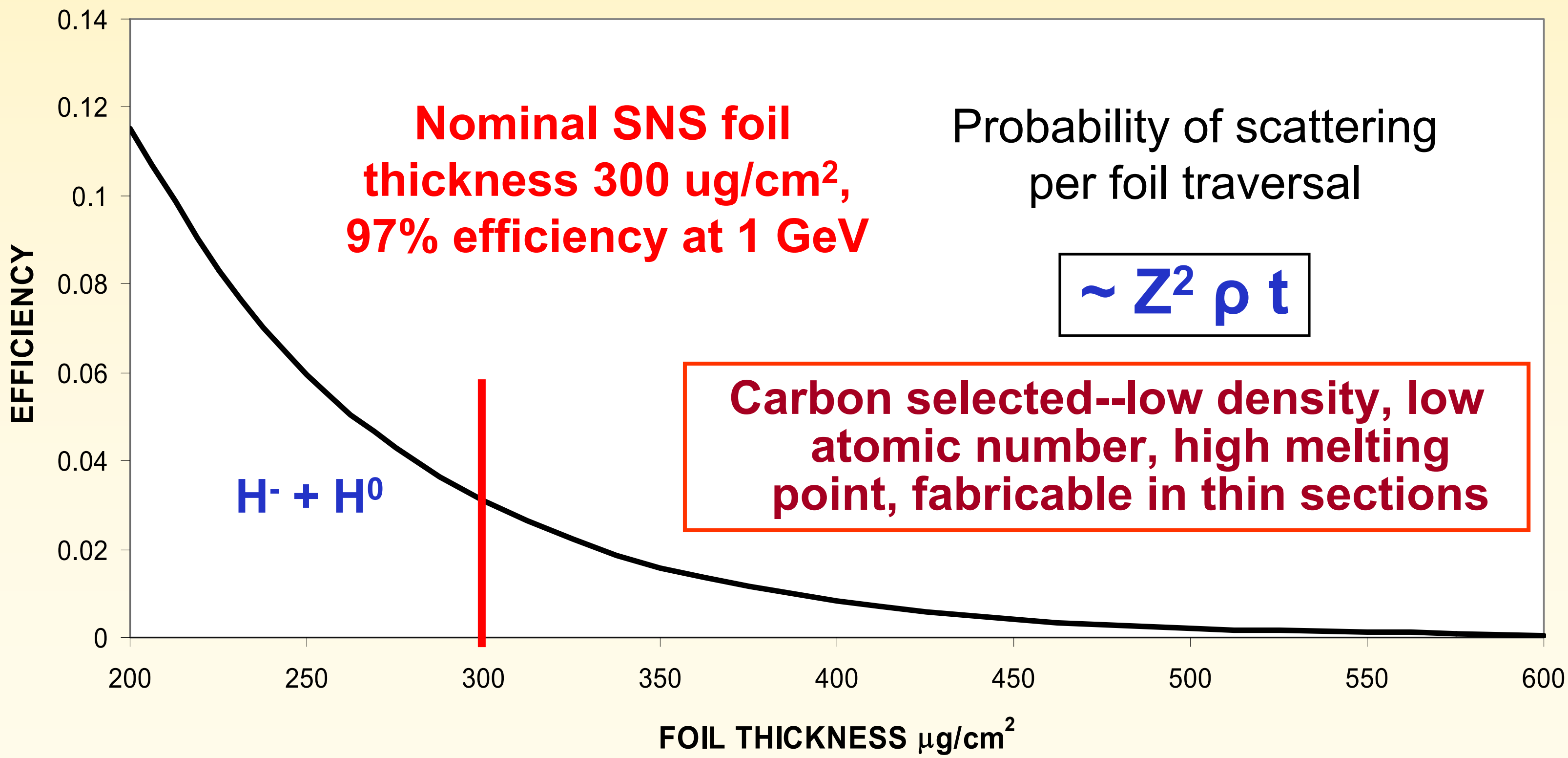
H^- Beam Stripper Foils

- Multi-Turn Charge-Exchange Injection creates short pulse of protons in Ring from long pulse Linac
- Two electrons are removed by the stripping foil, injected protons are merged with previously accumulated beam
- The secondary foil strips the H^- and H^0 which survive the first foil



Need Foil Thick Enough to Strip Electrons

- Average proton in SNS ring will pass through stripper foil 6 to 7x
- Thicker foil runs at higher T, scatters circulating beam, and increases activation levels
- Select thin foils with low atomic number and low density



Tradeoff between Stripper Foil and Ring Injection Dump Capabilities

Small **Foil Size/Thickness** **Large**

- **Less efficient H^+ production**
  - More power to dump
  - More radiation damage in dump
- **Lower loss and activation**
  - Lower foil temperature
  - Fewer foil hits by stored beam
  - Fewer large amplitude particles injected into ring

- **More efficient H^+ production**
  - Less power to dump
  - **Less radiation damage in dump**
- **More losses and activation**
  - Higher foil temperature
  - More foil hits by stored beam
  - More large amplitude particles injected into ring

Foil size and thickness optimization problem involves both materials and accelerator physics/engineering

High Power Accelerators Create Needs, Challenges and Opportunities

- **Unusual conditions in high power accelerators, especially (but not limited to) targets, compared with most of the current knowledge in radiation effects**
- **Challenge--ensure that high power accelerator targets and components will fulfill intended service**
  - Near term project oriented R&D
- **Opportunity--behavior of materials under previously unexplored irradiation conditions**
  - Fundamental radiation materials science
- **Continuing need--Materials irradiation facilities to serve high power accelerator community**

Acknowledgements

- Target materials--T. S. Byun, Yong Dai, Jim DiStefano, Ken Farrell, Martin Grossbeck, John Haines, John Hunn, Stuart Maloy, Steve Pawel, Bernie Riemer, Joe Strizak, Hans Ullmaier, Steve Zinkle
- Radiation damage calculations--Phil Ferguson, Franz Gallmeier, Monroe Wechsler
- Carbon stripper foils--Mike Plum

Further Reading

- W. Sommer, et al. "Materials selection and qualification processes at a high-power spallation neutron source," Mater. Char., 43 (1999) 97-123
- G. S. Bauer and H. Ullmaier, "Materials related work for the ESS target stations," J. Nucl. Mater. 318 (2003) 26-37
- Y. Dai, et al., "An overview of the behavior of structural materials after irradiation in SINQ targets at temperatures below 400 ºC," this conference
- Y. Oyama and K. Kikuchi, "Status of the J-PARC project on materials issues," J. Nucl. Mater. (in press)
- L. K. Mansur, "Materials research and development for the spallation neutron source mercury target," J. Nucl. Mater. 318 (2003) 14-25

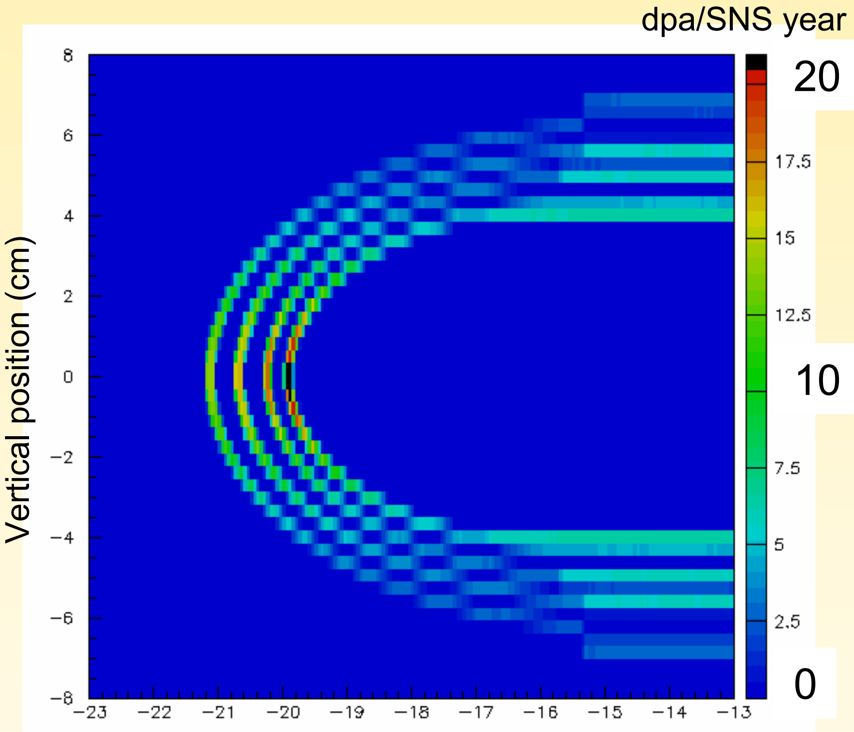
Materials R&D Experiments for SNS

Expt.	E	dpa	He/dpa	H/dpa	Hg Flow	T (° C)	σ	Cycles	Cavitation
SNS	≤ GeV	≥ 10	≥ 50	≥ 500	high	≤ 200	high	$\leq 10^9$	yes
3-beams	~ MeV	≥ 10	≥ 50	≥ 500					
p into liq.	~ MeV	≤ 5			high	≤ 200	high		
LANSCE	< GeV	≥ 10	≥ 50	≥ 500	(H_2O)	≤ 200			
SINQ	< GeV	≥ 10	≥ 50	≥ 500	(H_2O)				
HFIR	~ MeV	≥ 10	≤ 50	≤ 10	(H_2O)				
TC loop					low	≤ 300			
P loop					high	≤ 300			
Tensile					static	~ 25	high		
Fatigue					static	25	high	$\leq 10^9$	
WNR					static	25	high	≤ 200	yes
Vib. horn					static	25	high	$\leq 10^9$	yes
Impact					static	25	high	$\sim 10^6$	yes

High Energy Accelerator Radiation Damage Differences with Fission or Fusion Neutrons

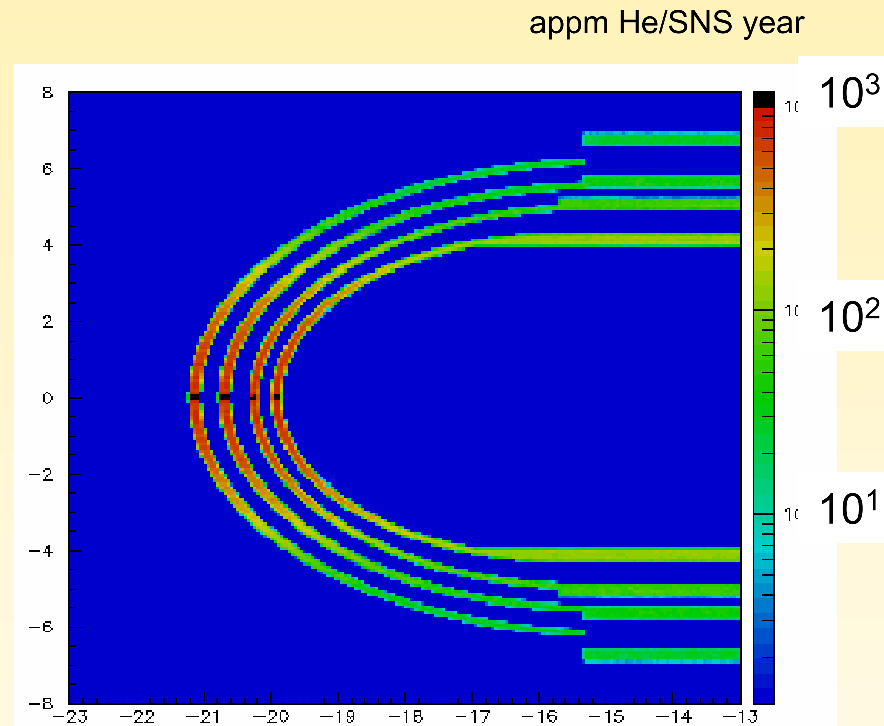
- **Highest particle energies**
  - GeV vs. ≤ 14 MeV
- **Instantaneous damage rates**
  - 10^{-2} vs. 10^{-6} dpa/s for pulsed beams (time average ~ 10^{-6} dpa/s)
- **He and H transmutation rates**
  - GeV protons ~ 500 appm H/dpa
    100 appm He/dpa
  - Fusion 10 "
  - Fission 0.2 "
- **Wide range of other transmutations**

SNS Target Radiation Damage



Axial position (cm)

Displacements



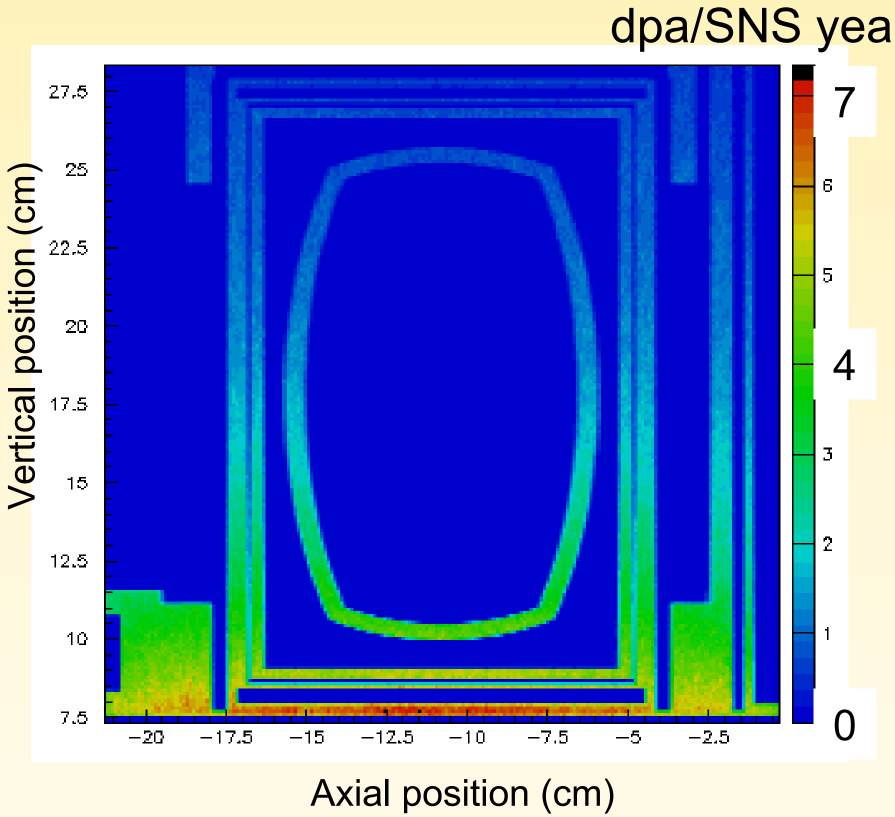
Axial position (cm)

Helium production

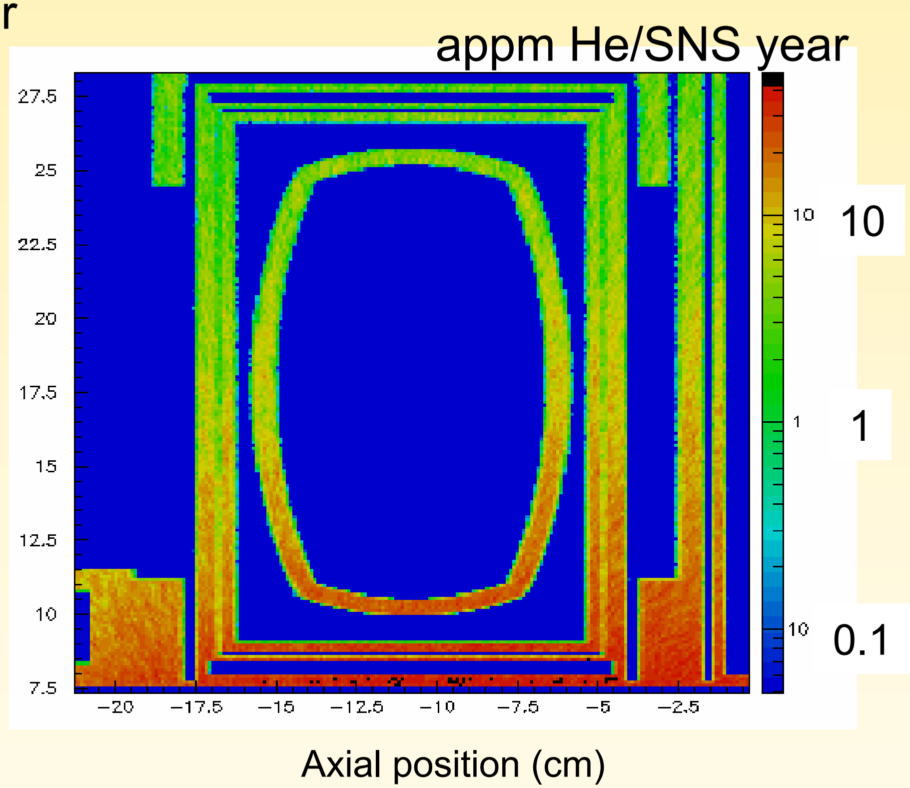
Maximum dpa rate is ~ 21 dpa/SNS year (~36 dpa/year)

(SNS year = 5,000 h)

Moderator Vessel Radiation Damage



Displacements

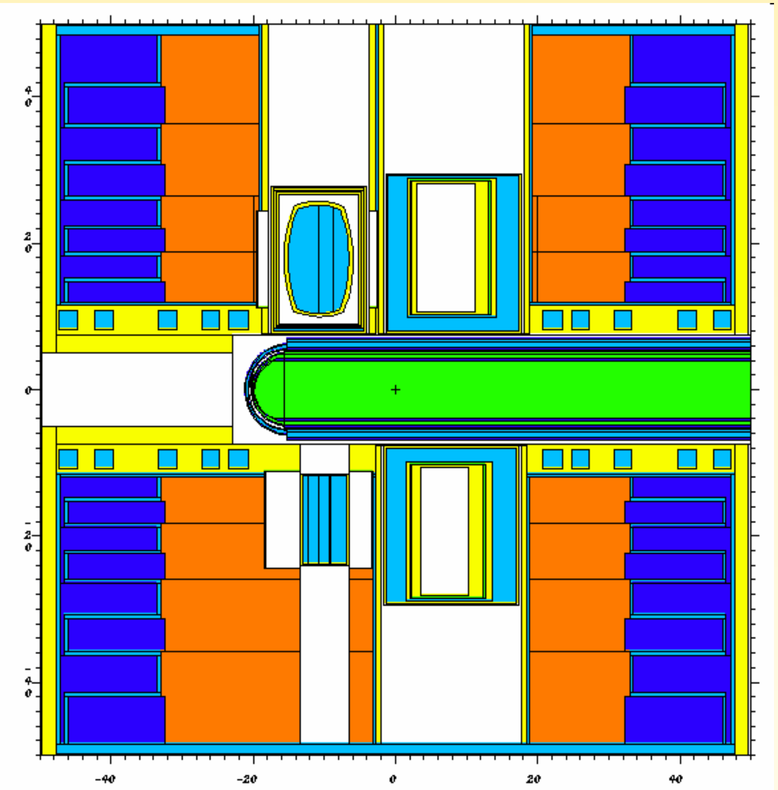


Helium

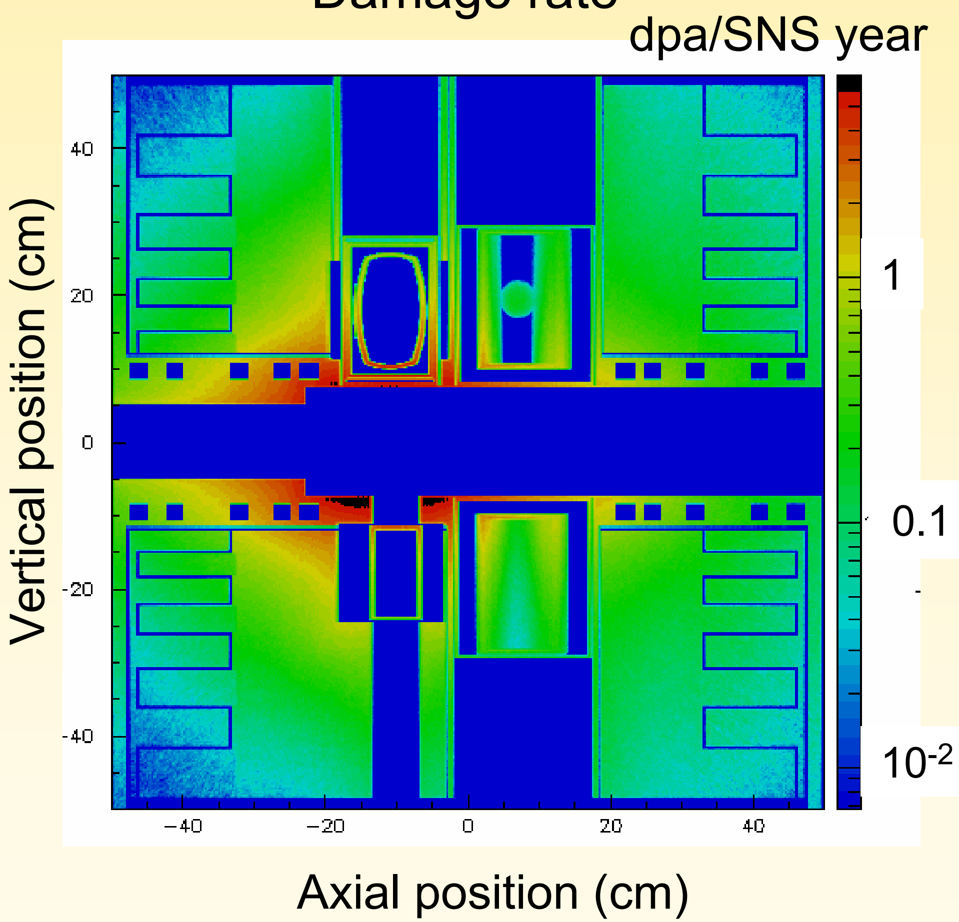
- **Maximum dpa rate is less than 8 dpa/SNS year**
- **Maximum He production less than 50 appm He/SNS yr (~6 appm He/dpa)**

Reflector Radiation Damage

Geometry



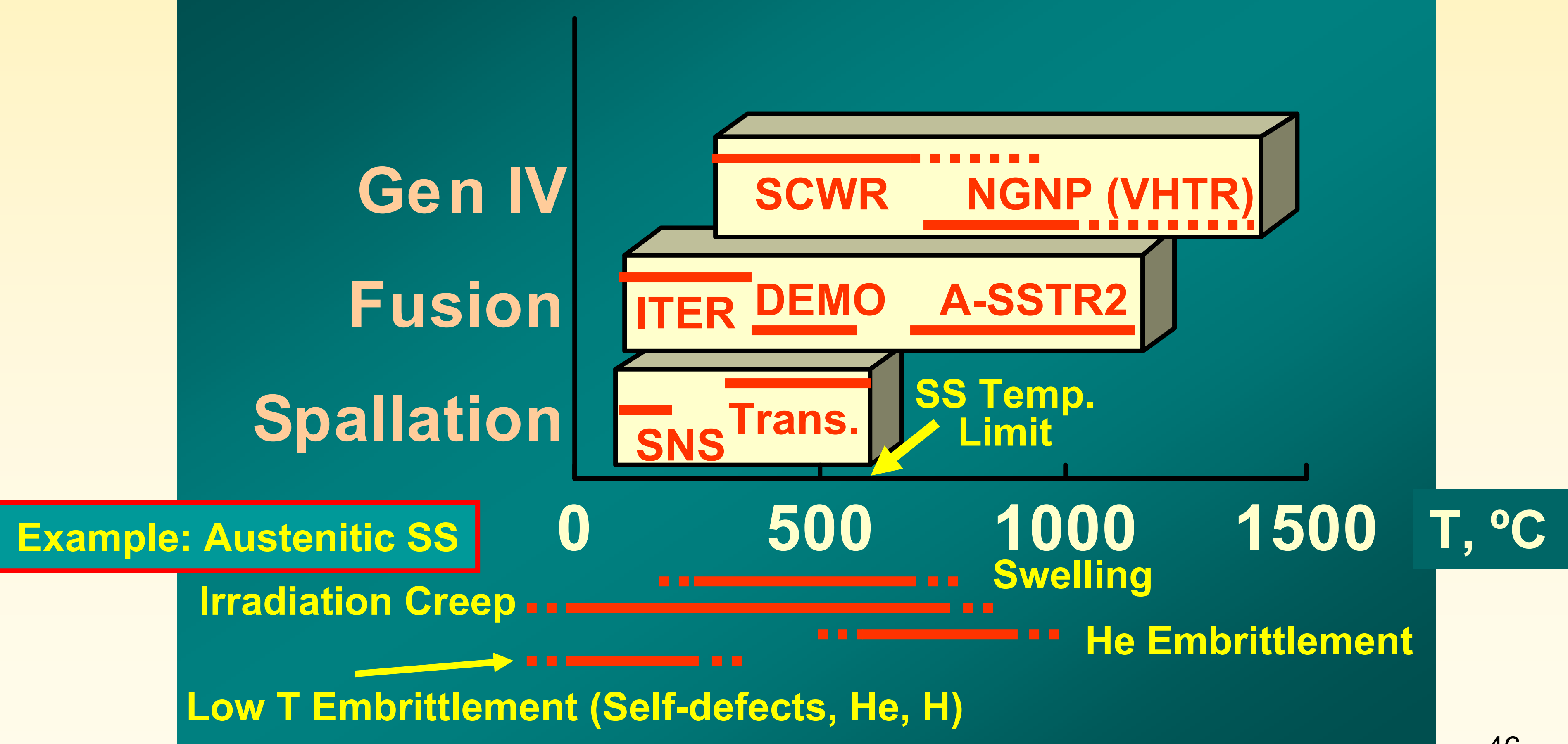
Damage rate

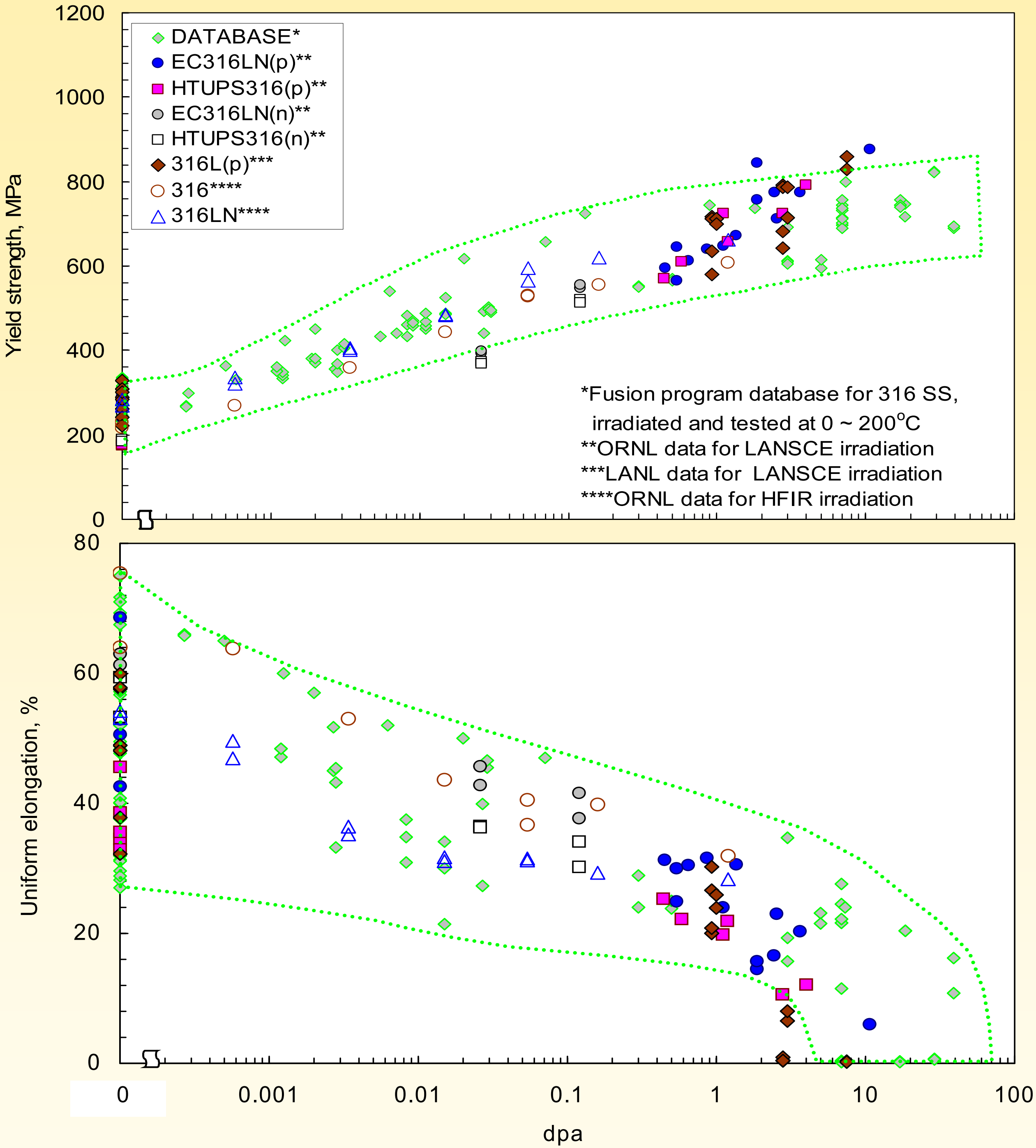


Maximum displacement rate of ~7 dpa/SNS y in Al 6061, less in steel and Be
Maximum He ~40 appm He/SNS y in Al 6061 and ~30 appm He/SNS yr in Be

Overlap in Temperature for Fusion, Generation IV Fission Reactors and Spallation Facilities

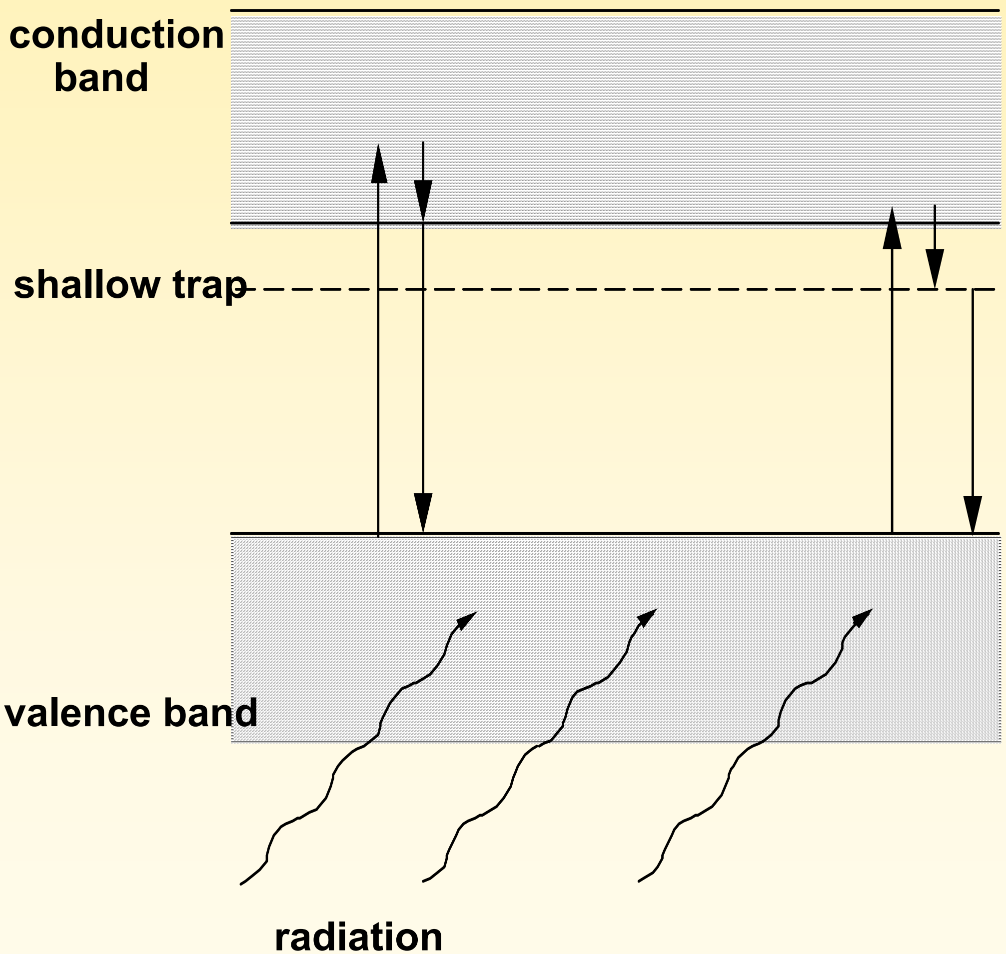
Operating Temperatures and Radiation Effects



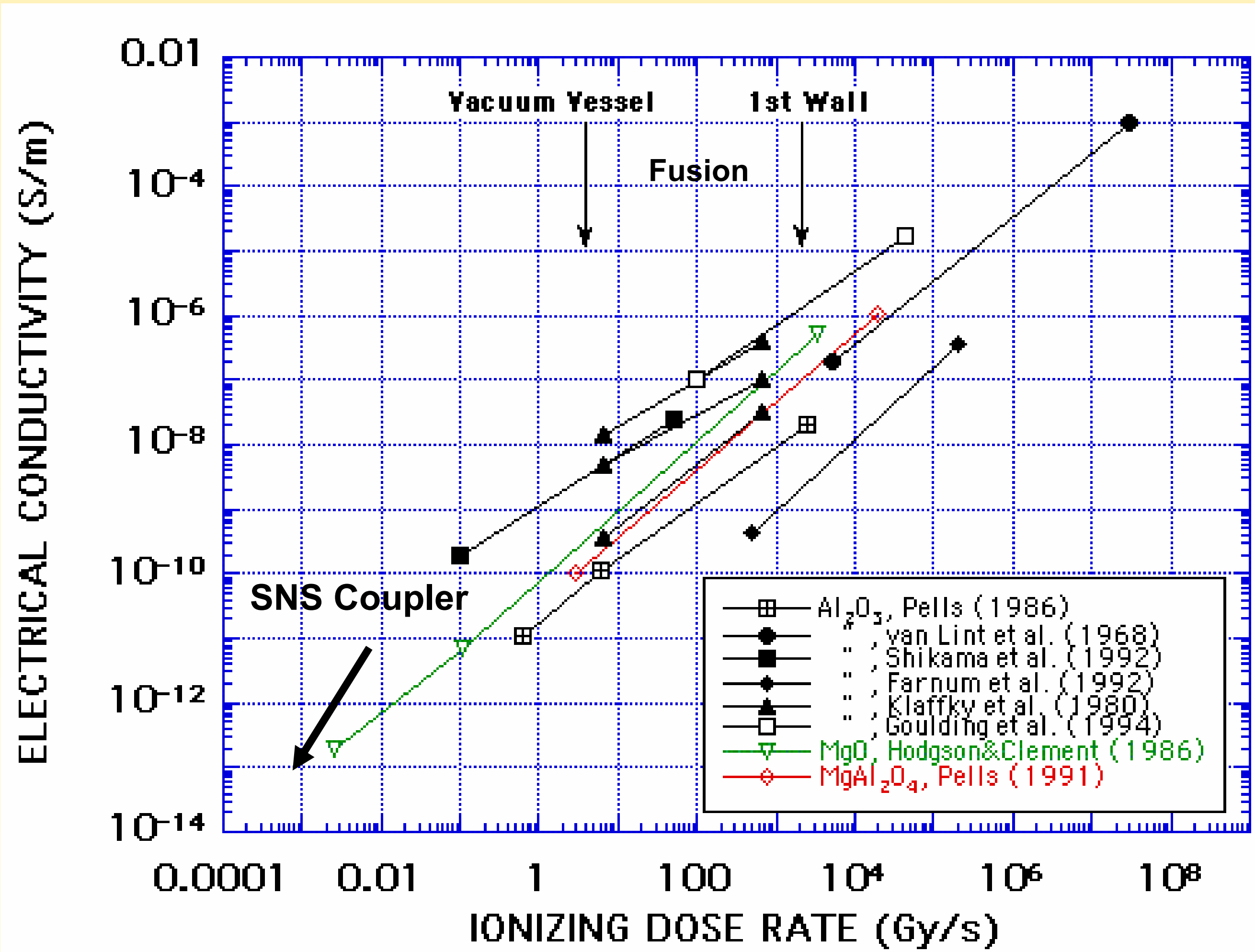


SNS R&D on Irradiated Yield Strength and Ductility of Stainless Steels

Radiation-Induced Conductivity in Insulators



Summary of RIC Data for Oxide Ceramics

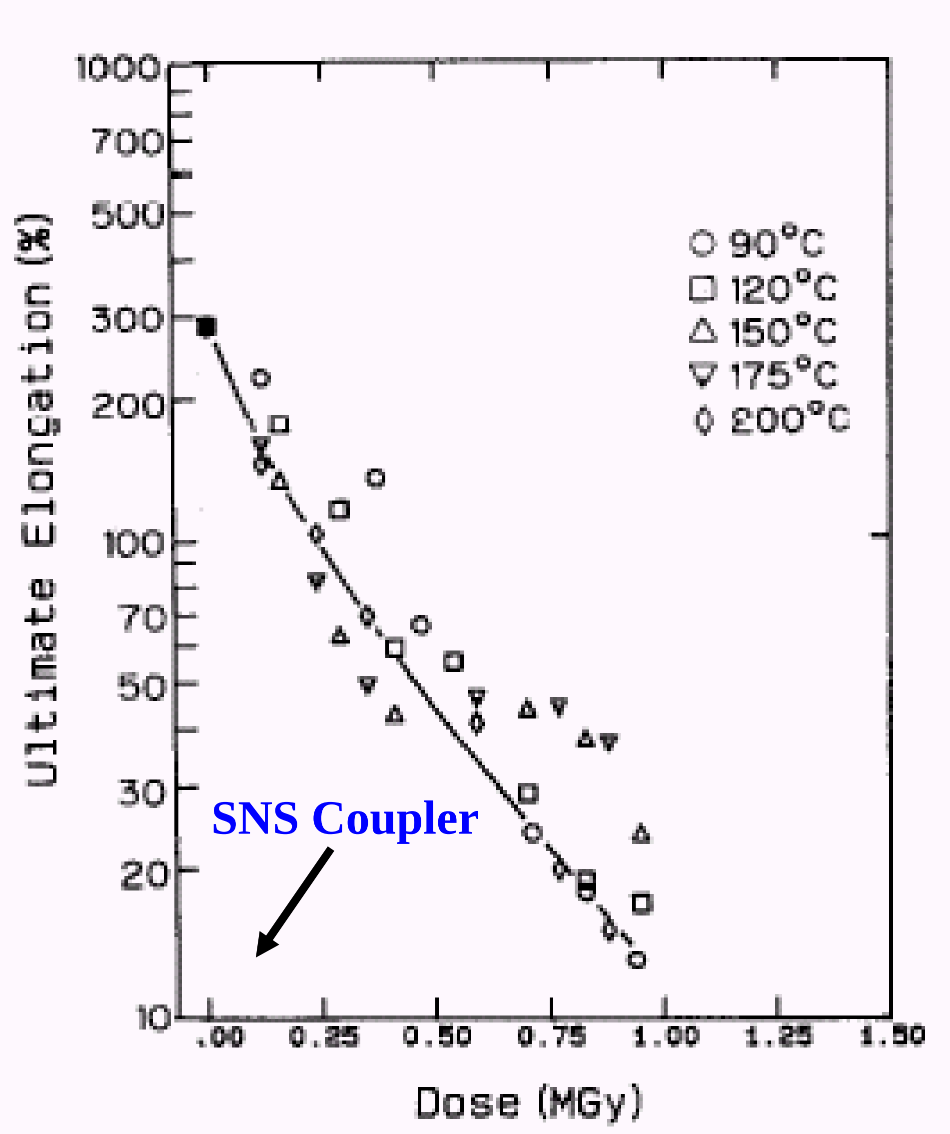


$$\sigma_{RIC} = \sigma_0 + K\phi^d$$

d~1.0

K depends on electron trap concentration

Decrease in Elongation of Viton Elastomer Irradiated at Various Temperatures



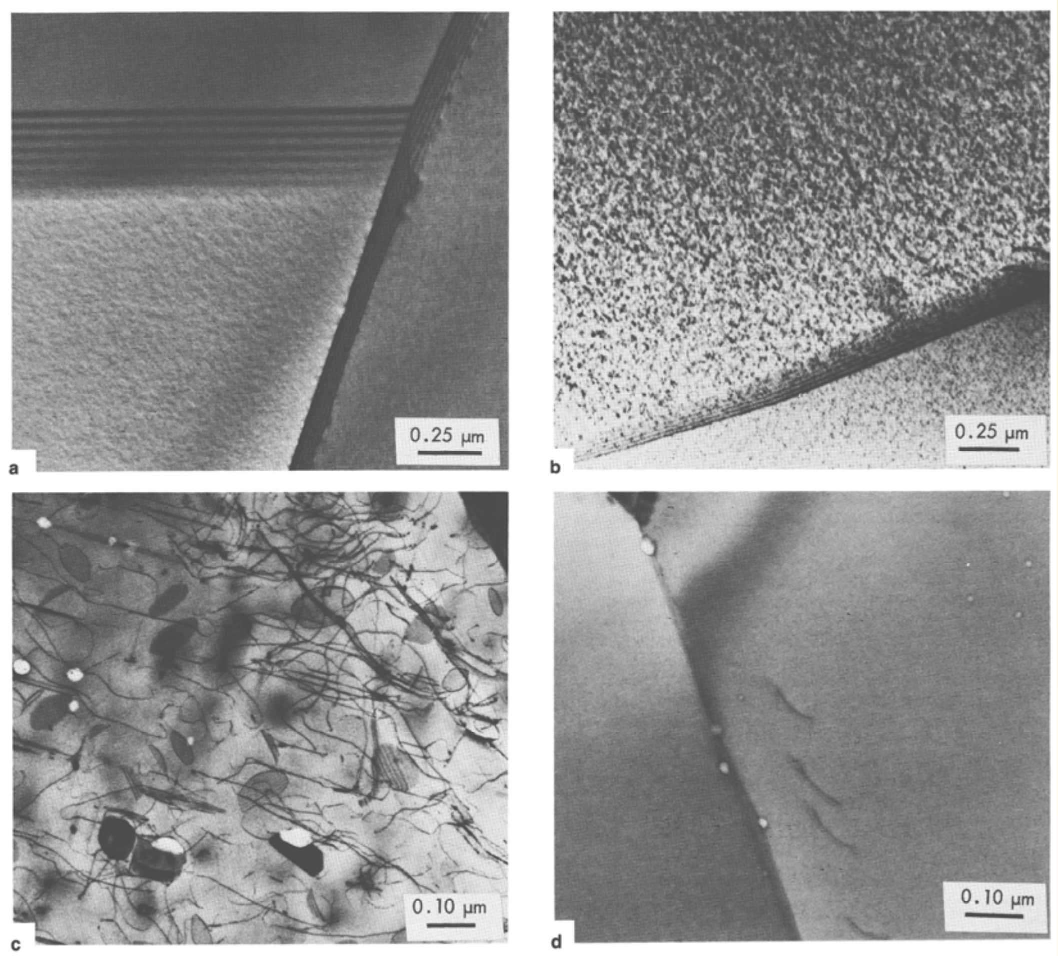
M. Ito, Radiat. Phys. Chem. 47(1996)607-610

Basics of Radiation Effects on Polymers

- **Although polymers are often classed as cross-linking or scission (degrading) types under irradiation, our research has shown that the ratio of cross-links to scissions depends strongly on LET. Energetic heavy ions cause much more cross-linking than γ or e^-, because of much higher LET and can lead to reclassification from scission type to cross-linking type**
- **Range of sensitivity for producing significant degradation spans more than three orders of magnitude in dose, for example, for reduction in elongation by 25%:**
  - $\gtrsim$1kGy PTFE (Teflon)
  - $\gtrsim 10^3$ kGy PI, PS (Polyimide, Polystyrene)
- **Sensitivity also depends on irradiation conditions and environment. Irradiation in vacuum can improve dose endurance over that in air by an order of magnitude. Irradiation at higher temperatures can give improvement.**

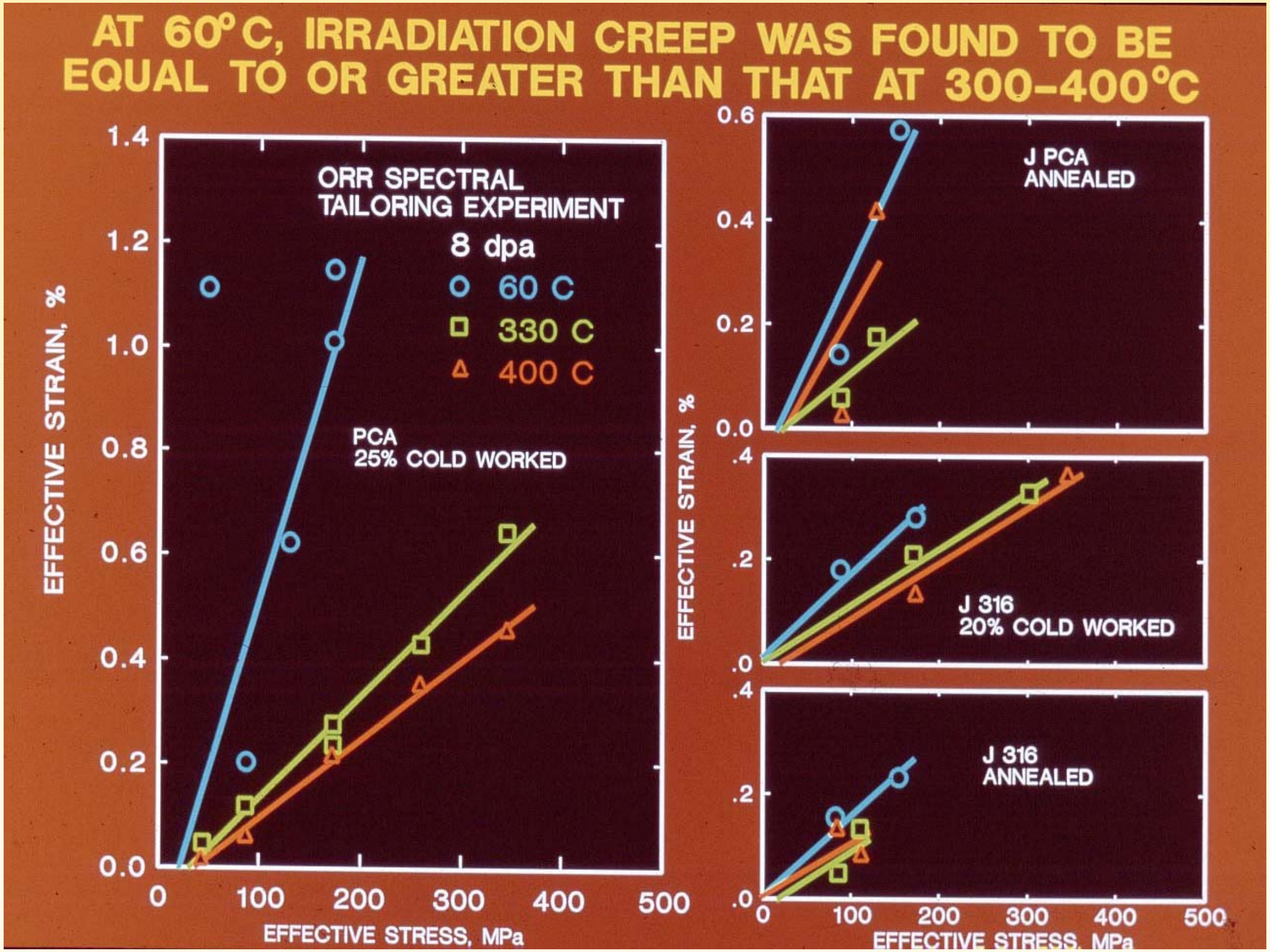
Type and Distributions of Microstructural Features are Strong Functions of Temperature

Not irradiated | Irradiated 300 ºC



Irradiated 500 ºC | Irradiated 700 ºC

Radiation-induced Creep



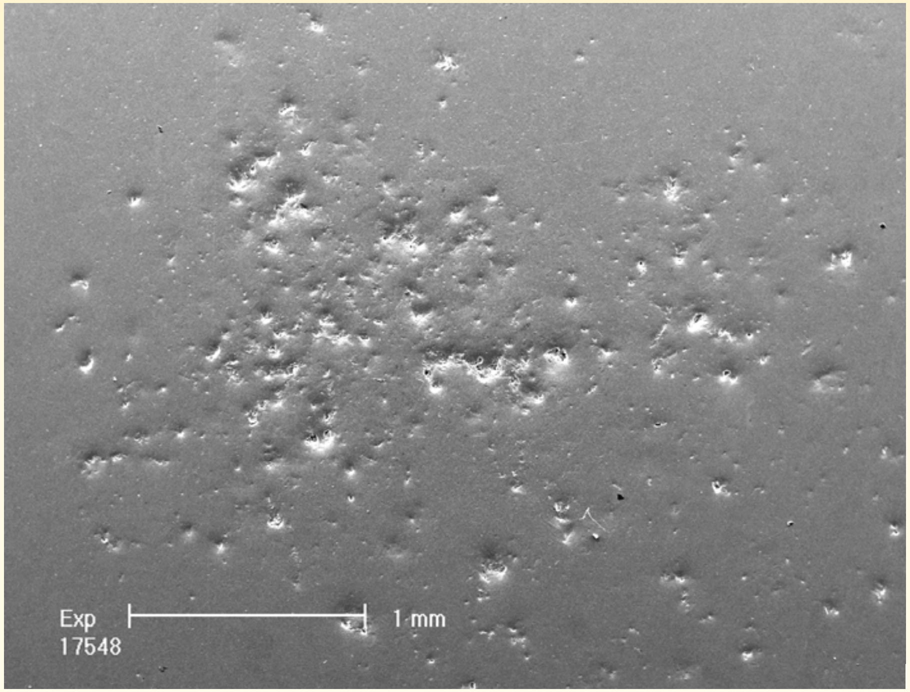
Approximate Radiation Dose Limits

- **People << 1 Gy (Sv) (ALARA)**
- **Polymers: 10^2 to 10^7 Gy**
- **Semiconductors: ~10^{13} n/cm^2, ~10^2 Gy (10^{16} to 10^{17} for SIC JFETs at 300˚C)**
- **Piezoelectric crystals: 10^{14} to 10^{19} (?) n/cm^2**
- **Ta capacitors: ~ 10^{15} n/cm^2, ~10^5 Gy**
- **Organic lubricants: 10^{16} n/cm^2, ~10^6 Gy**
- **Graphite, $MoSi_2$ lubricants: ~ no degradation up to 10^{19} n/cm^2**
- **Magnets: 10^{18} n/cm^2: up to 30% increase in coercive force and magnetic remanence**
- **Glass: 10^{20} n/cm^2 (>10% dimension change); 10^8 Gy (optical darkening saturates)**
- **Ceramics:**
  - ~10^9 Gy, ~ 10^{20} n/cm^2 (radiolysis-sensitive ceramics)
  - >10^{21} n/cm^2 (> 1 dpa) for most oxides, carbides and nitrides
- **Metals: > to >> 10^{21} n/cm^2 (> 1 dpa); ignore ionizing radiation**

Compiled by S. J. Zinkle, ORNL

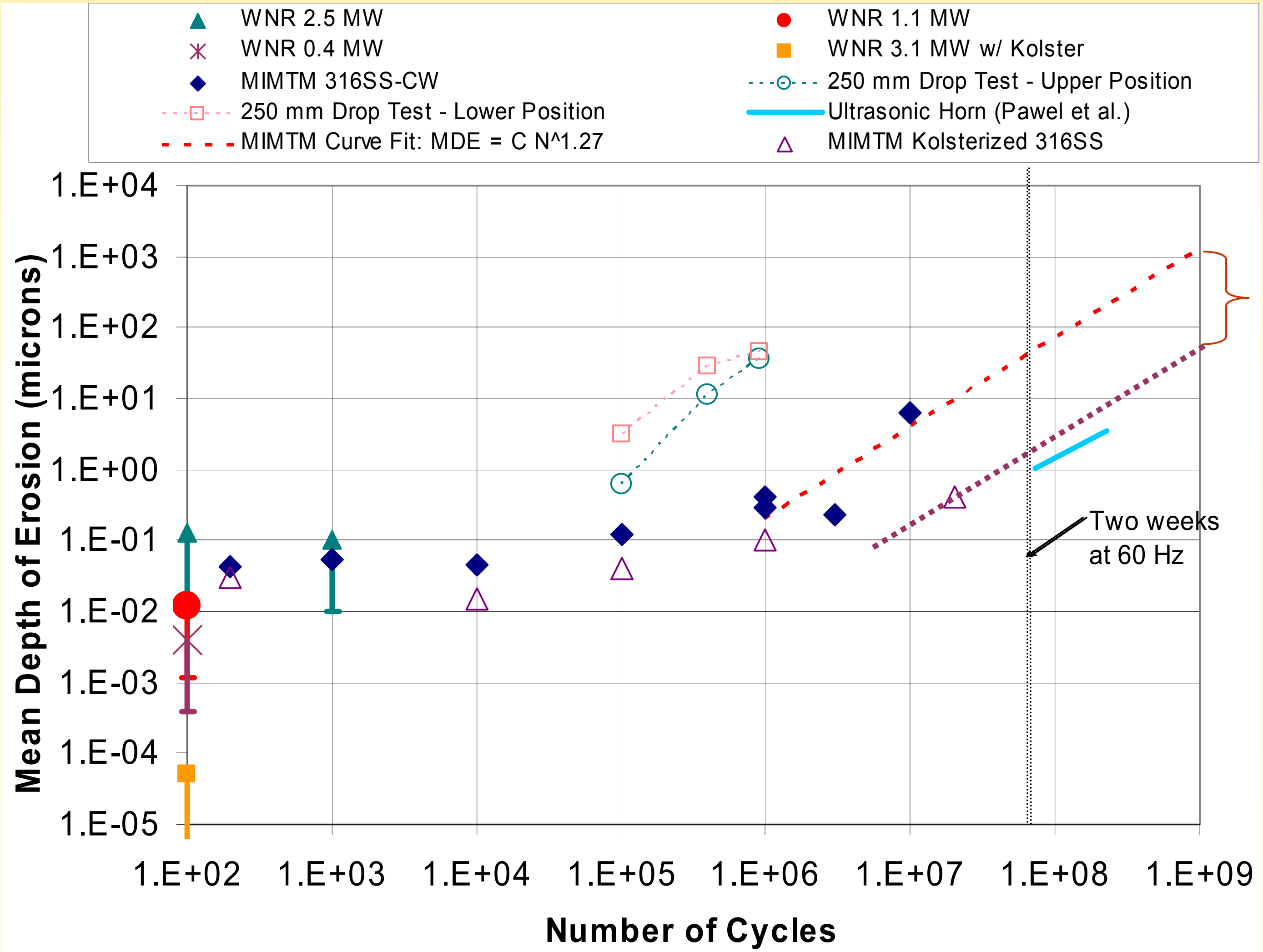
Cavitation Bubble Collapse Leads to Pitting Damage

- **Large tensile pressures occur due to reflections of compression waves from steel/air interface**
  - These tensile pressures cavitate the mercury
  - Damage is caused by violent collapse of cavitation bubbles under subsequent interaction with large compression waves



Damage in region with large pits for bare 316SS-LN diaphragm after July 2001 LANSCE-WNR tests

Summary of Pitting Erosion Tests



MIMTM device data used for extrapolation because 100 pulse damage is slightly worse than 1 MW equivalent in-beam damage

Extrapolating--estimated mean depth of erosion in SNS at 1 MW for 2 weeks < 50 μm