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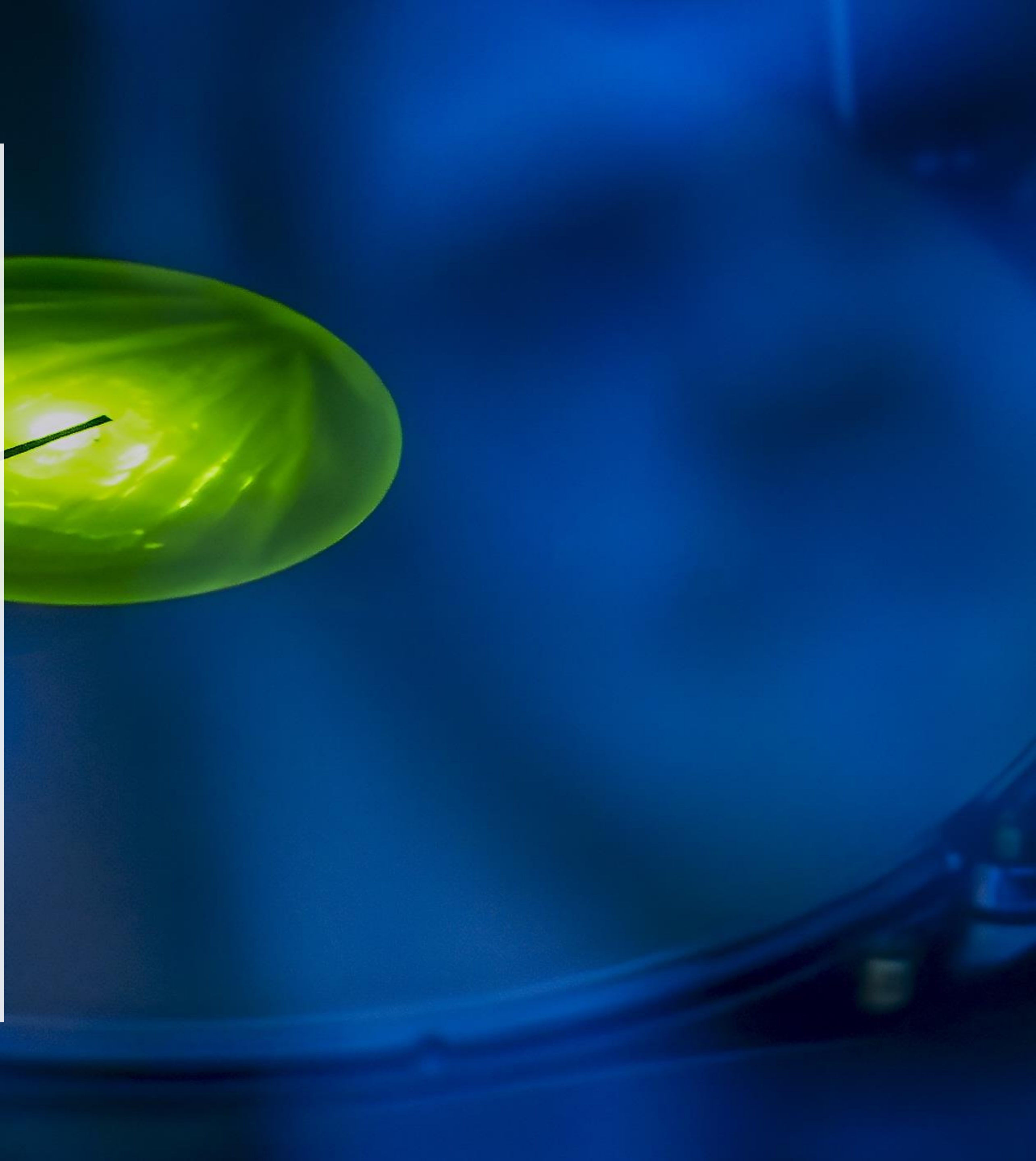
NSUF Foundations of Irradiation Testing Workshop, July 2025

Fundamentals of Radiation Damage in Nuclear Materials

Presented by

Stephen Taller

Oak Ridge National Laboratory



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Who am I?

Stephen Taller

Alvin M. Weinberg Fellow, R&D Staff Scientist

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Research Interests:
-Radiation effects in metals, alloys, and ceramics
-Microstructural characterization of metals and alloys
-Ion irradiation as a surrogate for neutron irradiation
-Advanced alloy development for nuclear applications
-Advanced manufacturing methods for nuclear technology
-Optimizing the nuclear structural materials development cycle

Mean Field Rate Theory

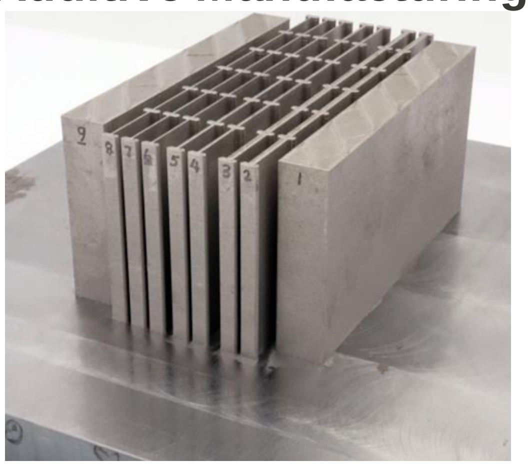
$$\frac{\mathrm{d}C_\mathrm{v}}{\mathrm{d}t} = K_0 - K_\mathrm{iv}C_\mathrm{i}C_\mathrm{v} - K_\mathrm{vs}C_\mathrm{v}C_\mathrm{s} + D_\mathrm{v}\frac{\mathrm{d}^2C_\mathrm{v}}{\mathrm{d}x^2}$$

$$\frac{\mathrm{d}C_\mathrm{i}}{\mathrm{d}t} = K_0 - K_\mathrm{iv}C_\mathrm{i}C_\mathrm{v} - K_\mathrm{is}C_\mathrm{i}C_\mathrm{s} + D_\mathrm{i}\frac{\mathrm{d}^2C_\mathrm{i}}{\mathrm{d}x^2}$$

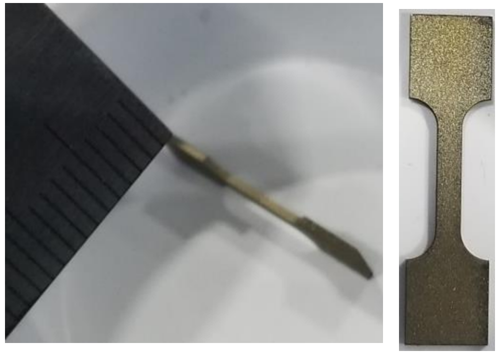
$$\frac{dC_{He}}{dt} = G_{He} - \sum_{i}^{\text{loops, bubbles line}} k_i^2 D_{He} C_{He} + \sum_{i}^{\text{loops, bubbles, line}} N_i \Omega n_g^i \nu_0 exp\left(-E_b^i/_{kT}\right)$$

$$\frac{dn_g^i}{dt} = \frac{k_i^2 D_{He} C_{He}}{N_i \Omega} - n_g^i \nu_0\, exp\left(\frac{-E_b^i}{kT}\right)$$

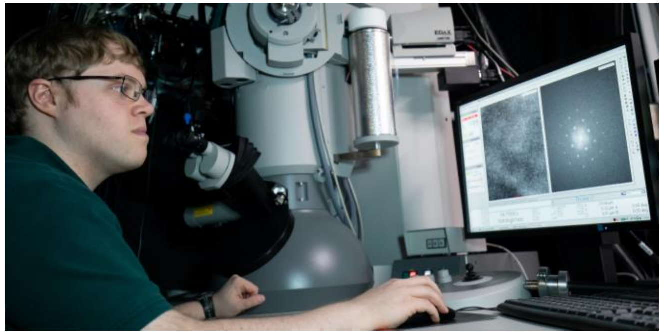
Additive Manufacturing



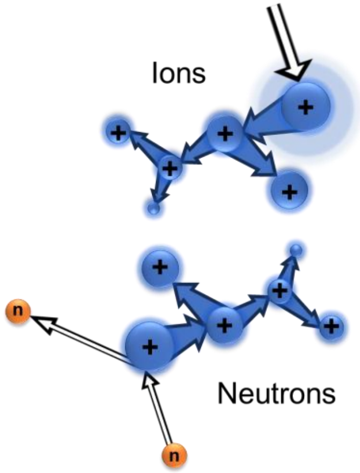
Miniature Mechanical Testing



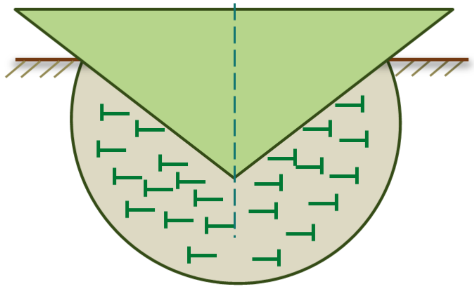
SEM and S/TEM



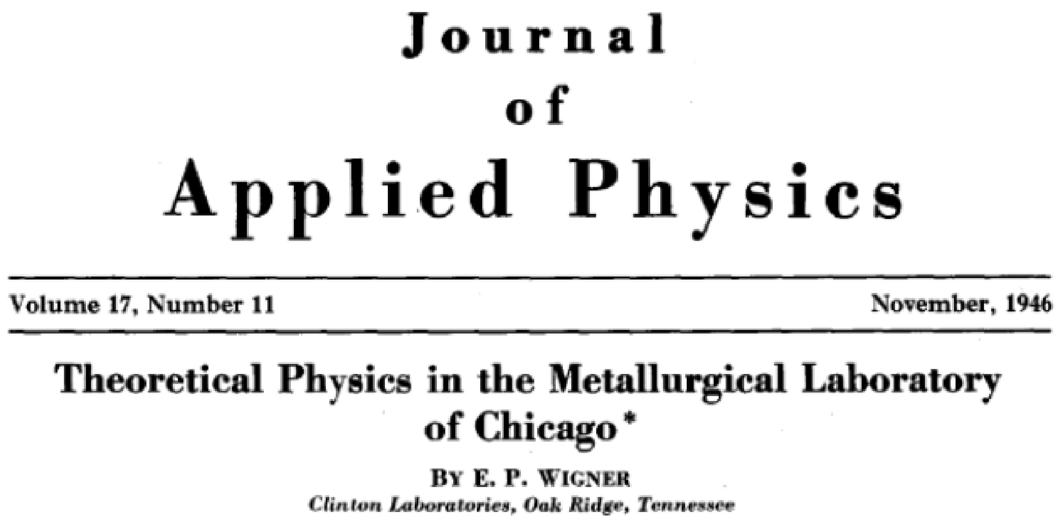
Neutron and Ion Irradiation



Instrumented Indentation Testing



Radiation Damage has been part of the Nuclear Engineering discipline since the beginning



Journal of Applied Physics

Volume 17, Number 11 November, 1946

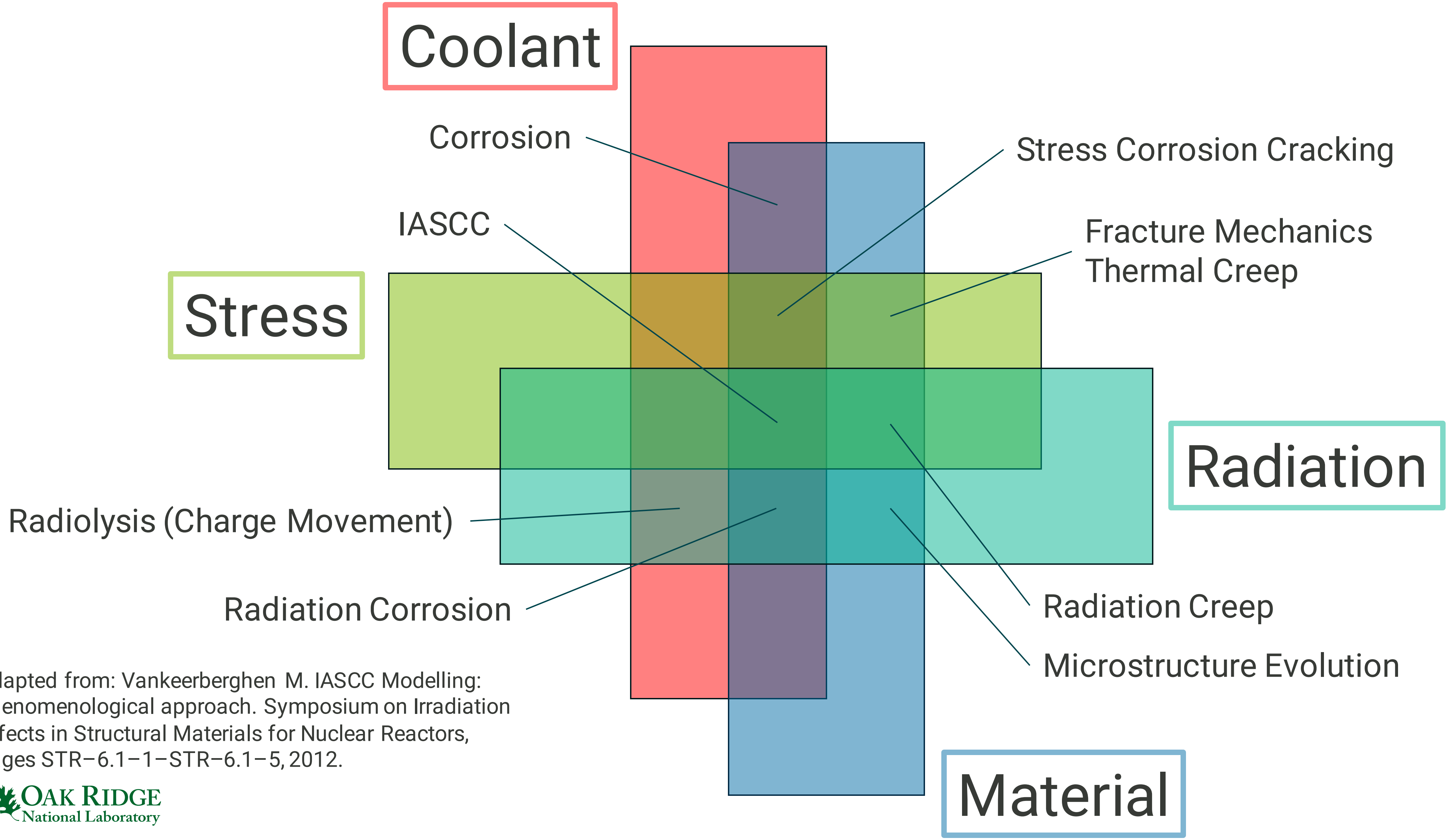
Theoretical Physics in the Metallurgical Laboratory of Chicago*

BY E. P. WIGNER

Clinton Laboratories, Oak Ridge, Tennessee

- Excerpts from Section 3.
- "The **radiation densities, both γ and neutron, are higher** in a plutonium producing pile than can be maintained outside the pile for extended periods of time."
- "The effect of these radiations on the structure of materials was one of our early concerns from the theoretical point of view."
- **"Clearly, the collision of neutrons with the atoms of any substance placed into the pile will cause displacements of these atoms."**
- "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."

Radiation damage is one piece in a complex puzzle



Adapted from: Vankeerberghen M. IASCC Modelling: phenomenological approach. Symposium on Irradiation Effects in Structural Materials for Nuclear Reactors, pages STR–6.1–1–STR–6.1–5, 2012.

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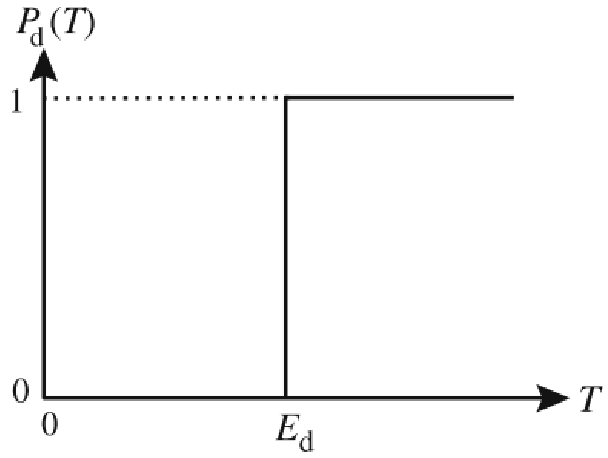
Our Discussion Today Is in 3 Parts

What is radiation damage?

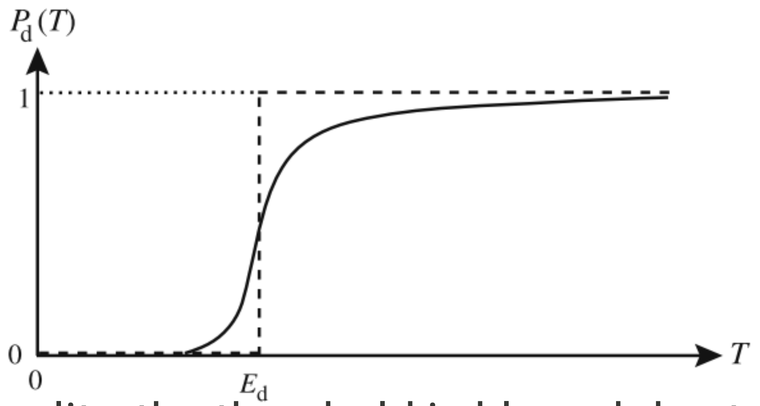
What does it do to materials, broadly?

What are some open questions?

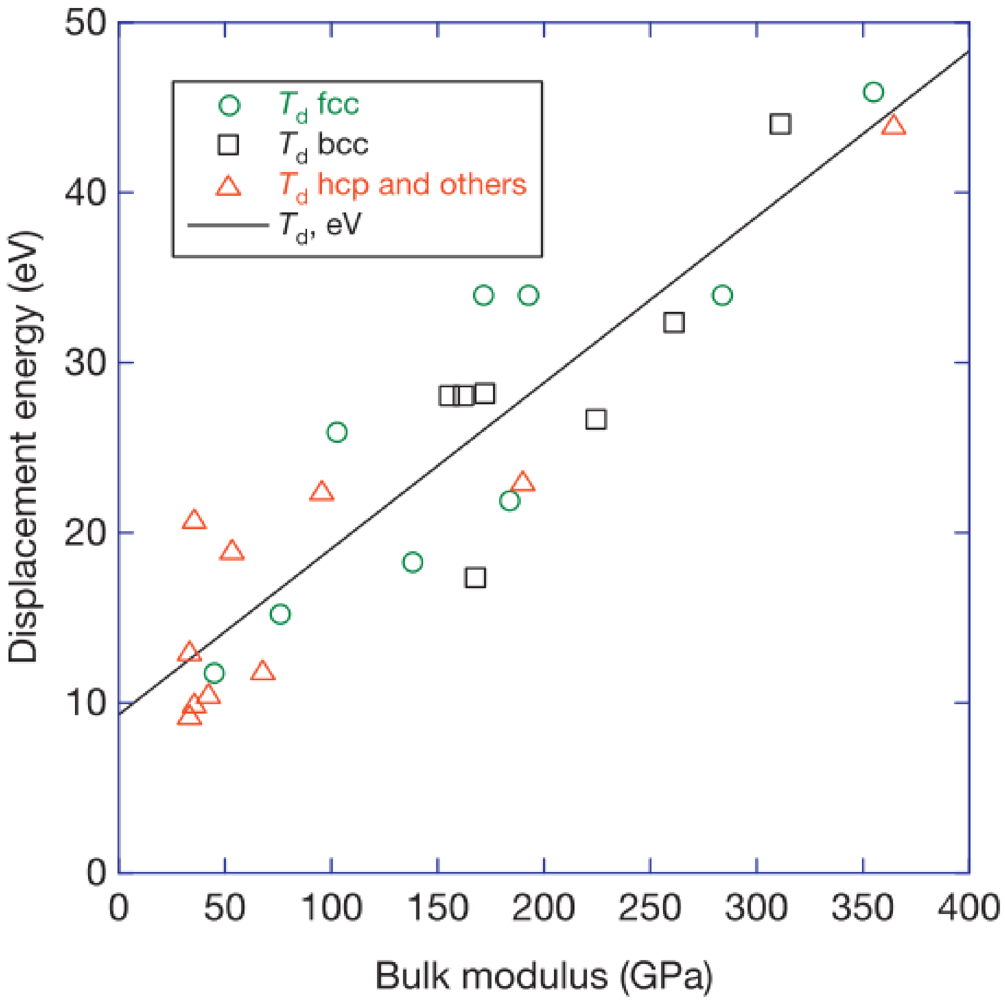
Radiation damage is a disruption of the crystal lattice and production of defects in the crystal



Displacements happen when enough energy is transferred to displace an atom



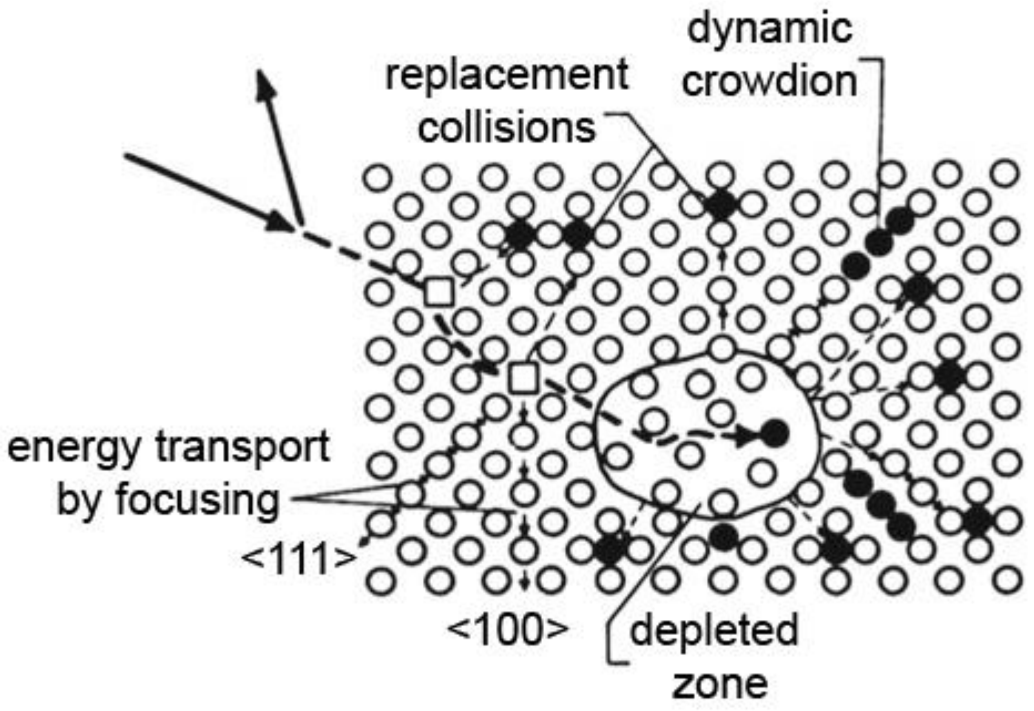
In reality, the threshold is blurred due to atomic vibrations, impurity atoms, scattering angle, etc.



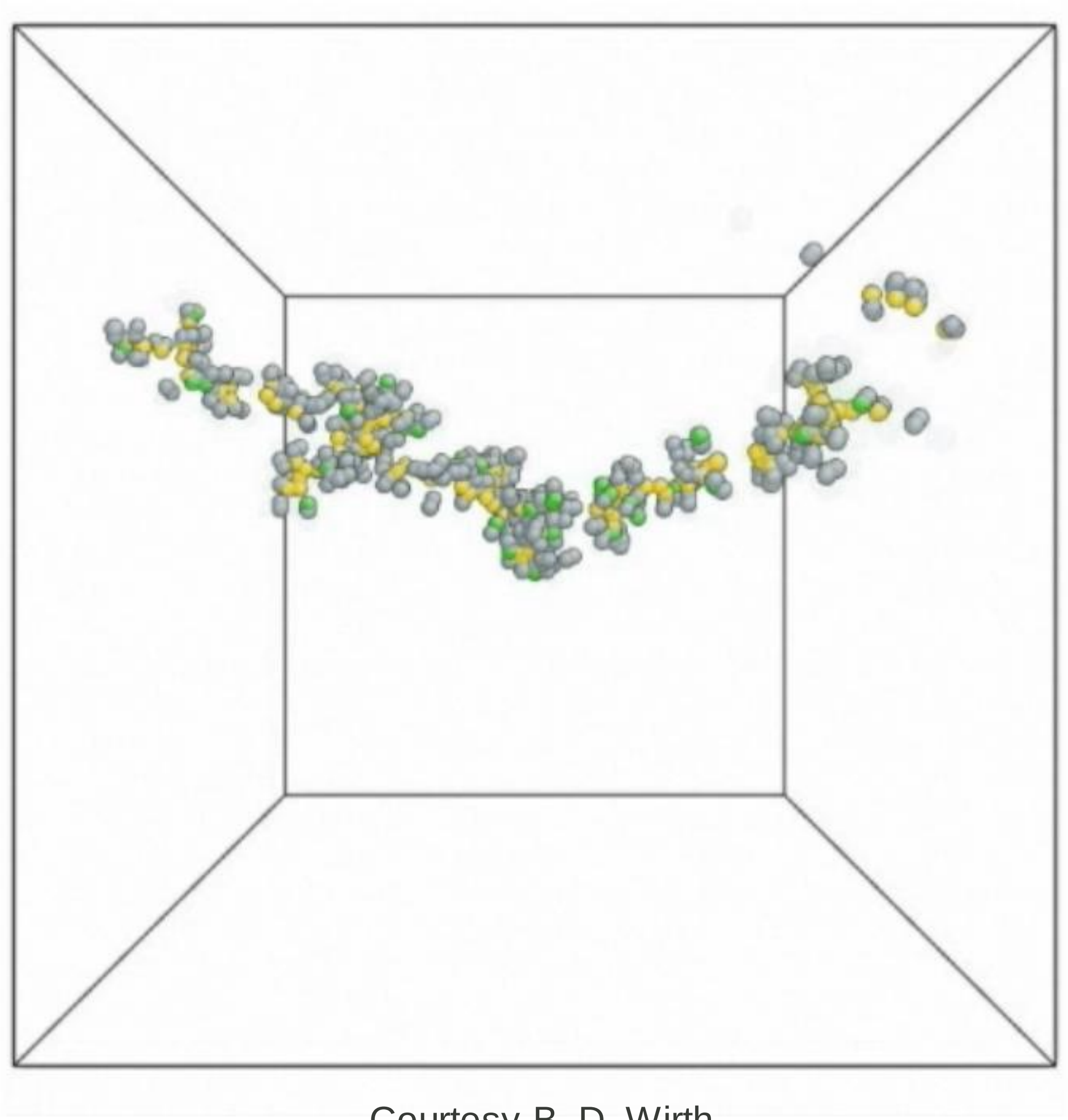
Wolfer, Comprehensive Nucl. Mat. (2020)

Adapted from *G. S. Was, Fund. of Rad. Mat. Sci., 2nd ed., 2017*.

Damage cascades and generates subcascades until all atoms are below a threshold energy

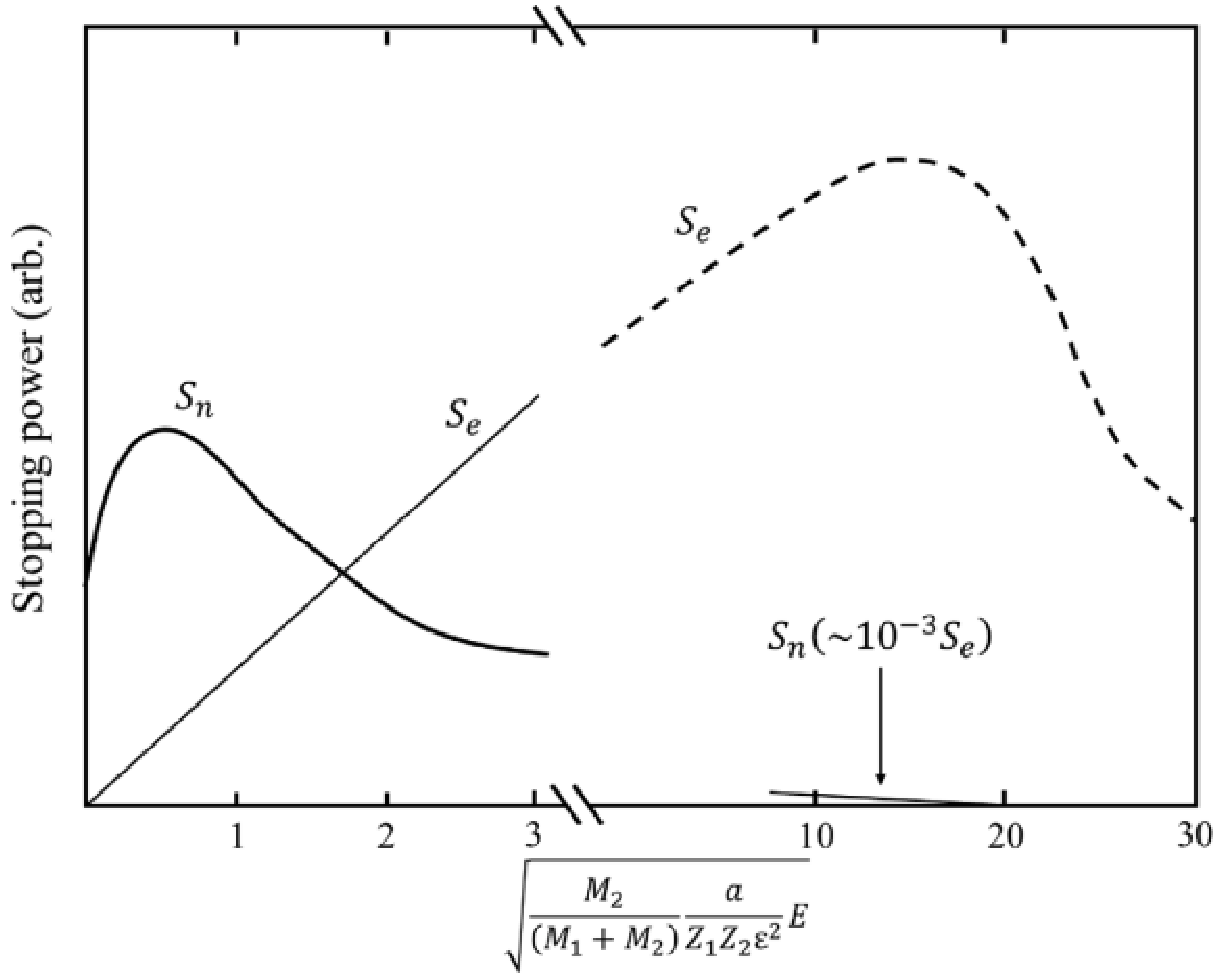


Adapted from *G. S. Was, Fund. of Rad. Mat. Sci., 2nd ed., 2017*.



Courtesy, B. D. Wirth

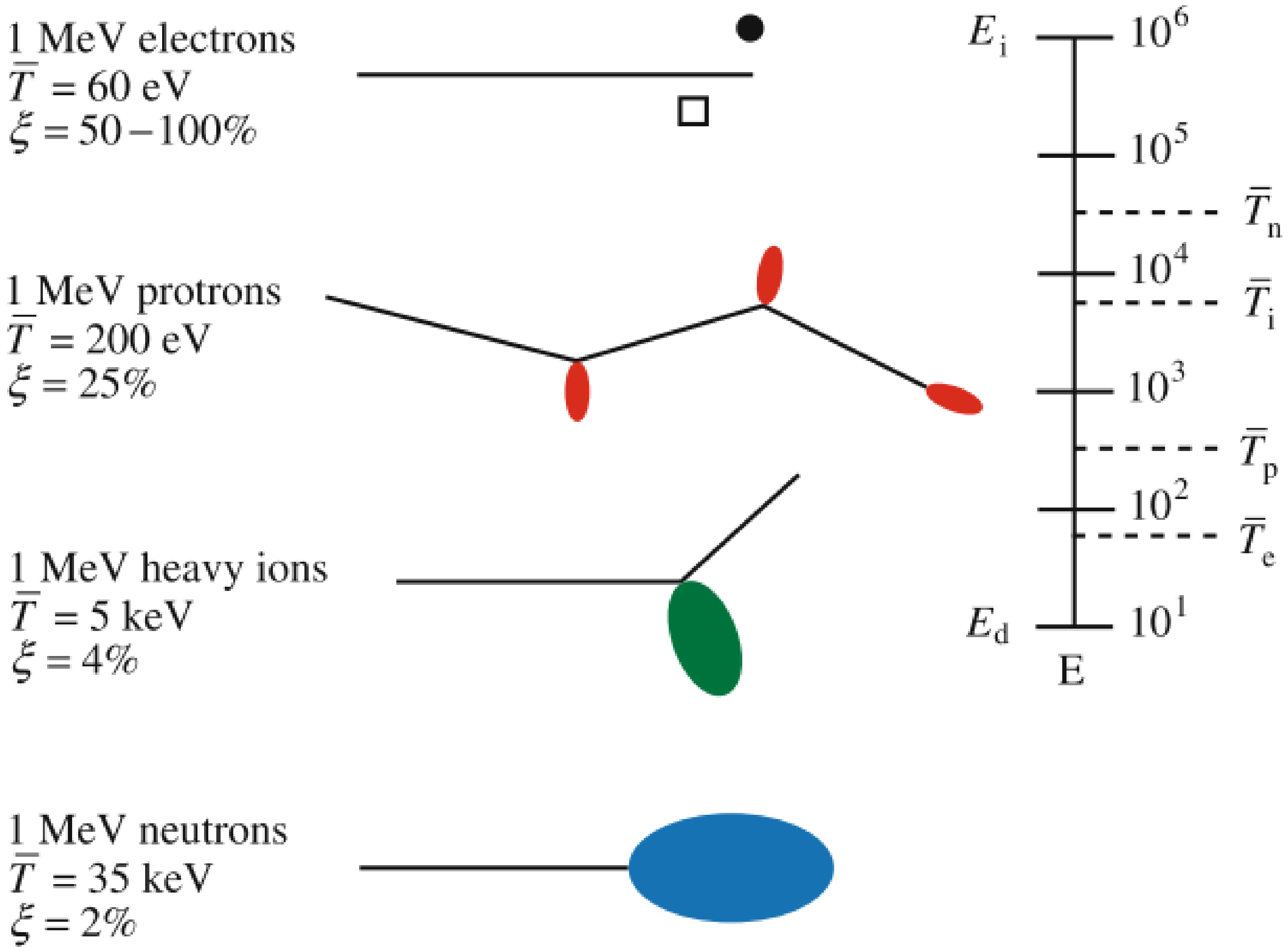
Differences in PKA and cascades occur via nuclear and electronic energy losses



Adapted from *G. S. Was, Fund. of Rad. Mat. Sci., 2nd ed., 2017*.

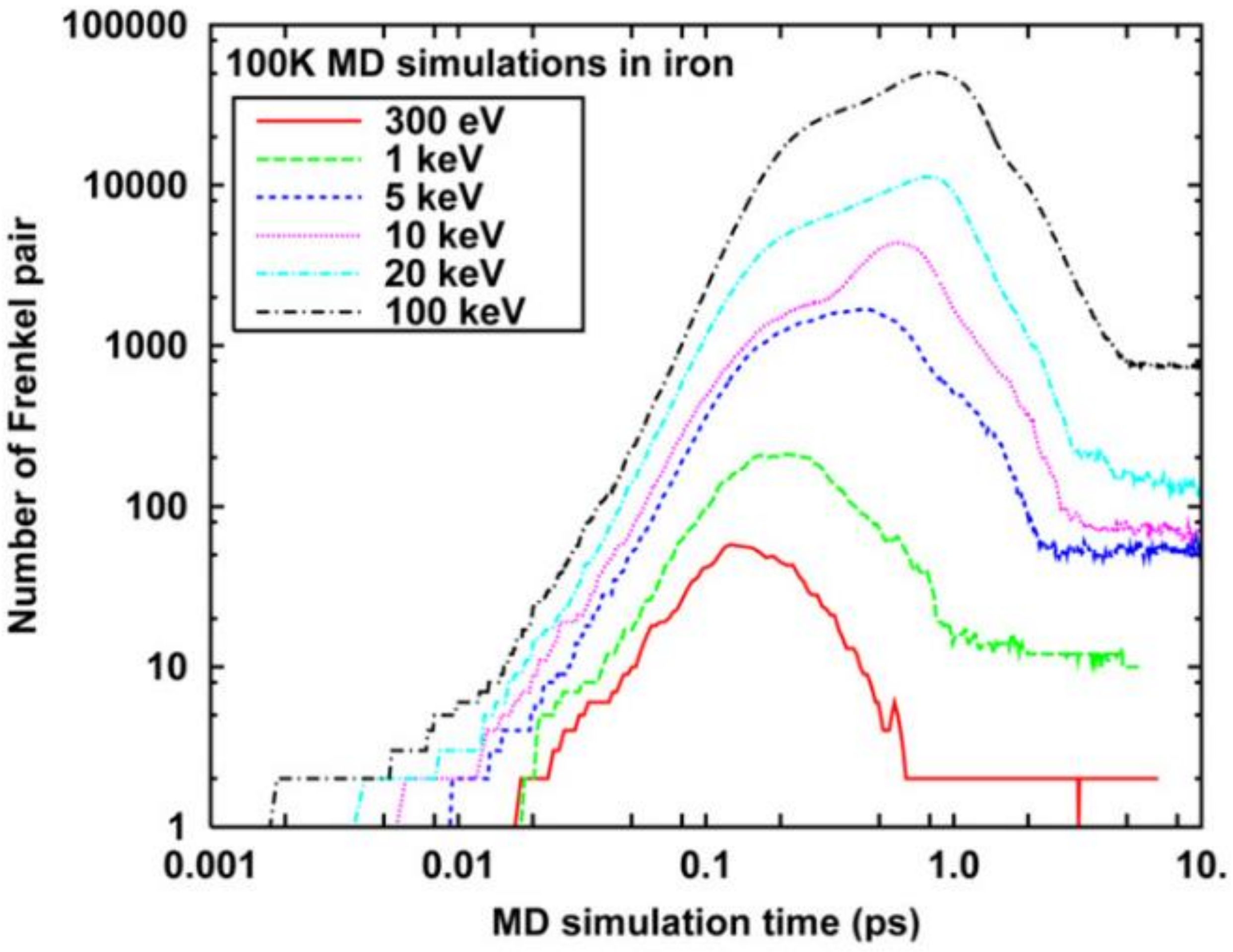
- Nuclear Stopping Power (S_n)
  - Primarily responsible for displacements
- Electronic Stopping Power (S_e)
  - Responsible for:
    - Phonon transport (heat)
    - Electron motion (chemical activity)

Radiation damage cascades are particle dependent

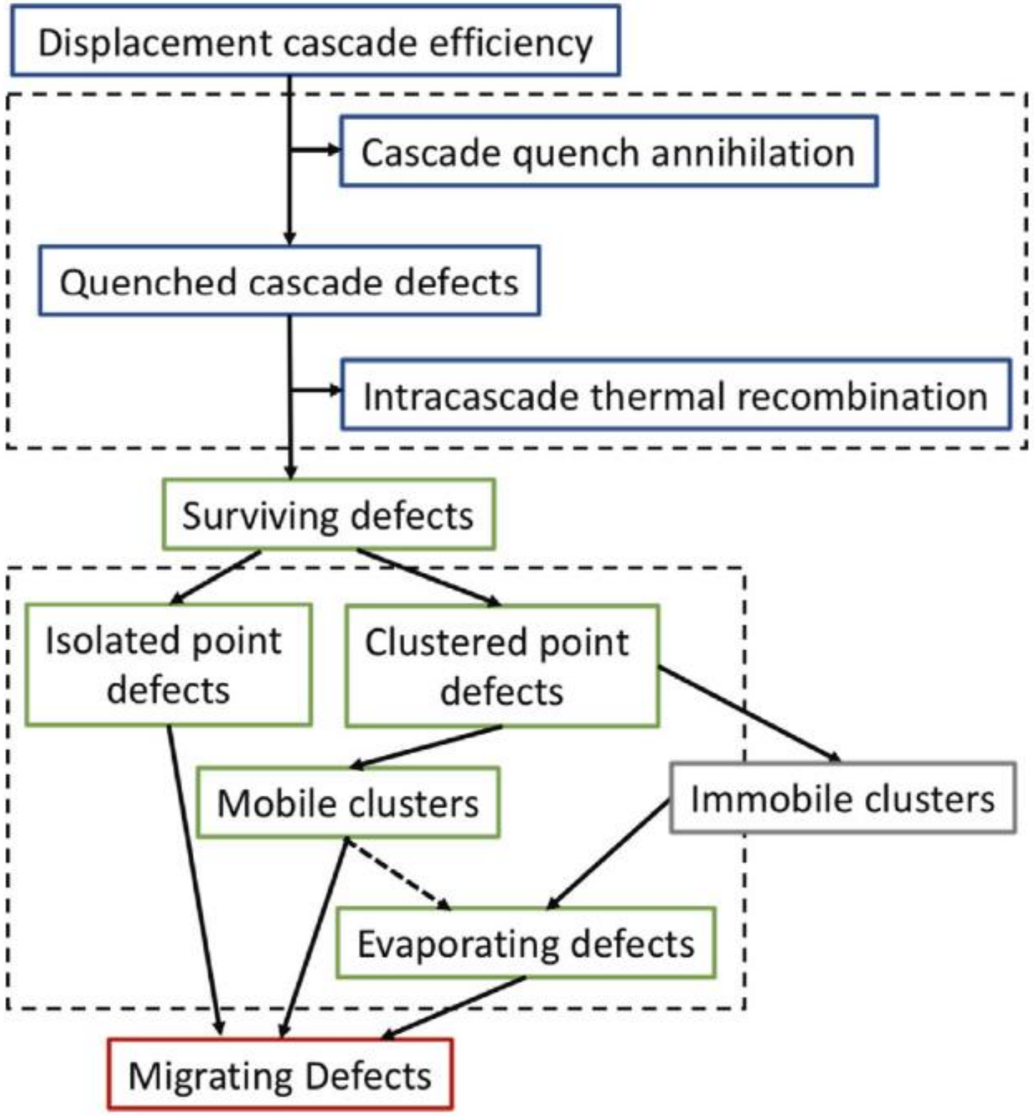


Adapted from *G. S. Was, Fund. of Rad. Mat. Sci., 2nd ed., 2017*.

Displacements are only one part of the process



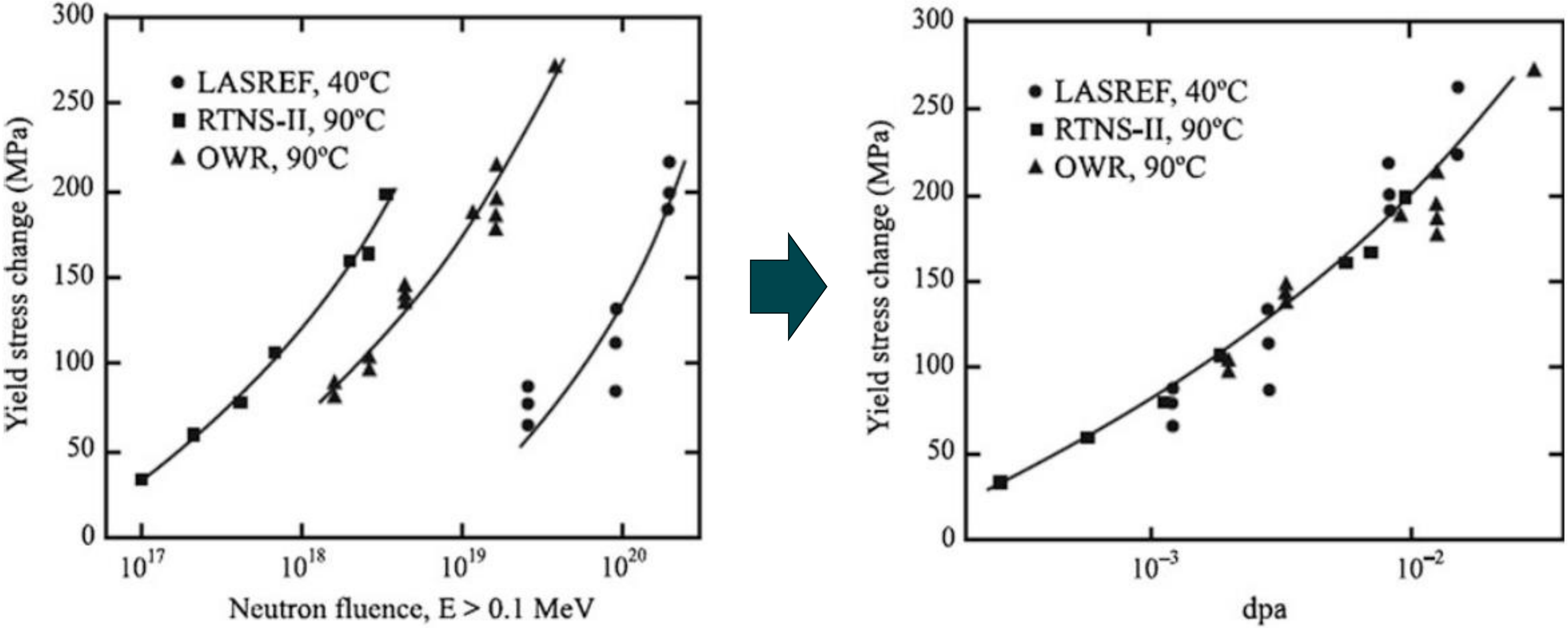
R. Stoller and E. Zarkadoula, Comp. Nuc. Mat. 2020



Adapted from *G. S. Was, Fund. of Rad. Mat. Sci., 2nd ed., 2017*.

What metric can define this process?

The metric of displacements per atom (DPA) is used to compare across environments



L. R. Greenwood, JNM 216, p. 29-44, (1994)

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The international standard is the Norgett-Robinson-Torrens model of displacements (NRT-dpa)

Kinchin-Pease model (1955)

$$N_d = \begin{cases} 0 & 0 < E < E_d \\ 1 & E_d < E < 2E_d \\ E/2E_d & 2E_d < E < E_1 \\ E_1/2E_d & E_1 < E < \infty \end{cases}.$$

Hard barrier for displacements
Linear rate of production at higher energies

Hard barrier for displacements
Non-linear rate of displacements based on additional factors

(a) The modified Kinchin–Pease formula of Torrens and Robinson [10] is used to calculate the number of Frenkel pairs N_d generated by a primary knock-on of initial kinetic energy E:

$$N_d = \kappa\hat{E}/2E_d, \quad (4)$$

where $\hat{E}$ is the energy available to generate atomic displacements by elastic collisions.

(b) The displacement efficiency κ is given the value 0.8, independent of the PKA energy, the target material, or its temperature.

(c) The inelastic energy loss is calculated according to the method of Lindhard et al. [13] using a numerical approximation [19] to the universal function $g(\epsilon)$:

$$\hat{E} = \frac{E}{[1 + k\, g(\epsilon)]}, \quad (5)$$

$$g(\epsilon) = 3.4008\,\epsilon^{1/6} + 0.40244\,\epsilon^{3/4} + \epsilon, \quad (6)$$

$$k = 0.1337\, Z_1^{1/6}\,(Z_1/A_1)^{1/2}, \quad (7)$$

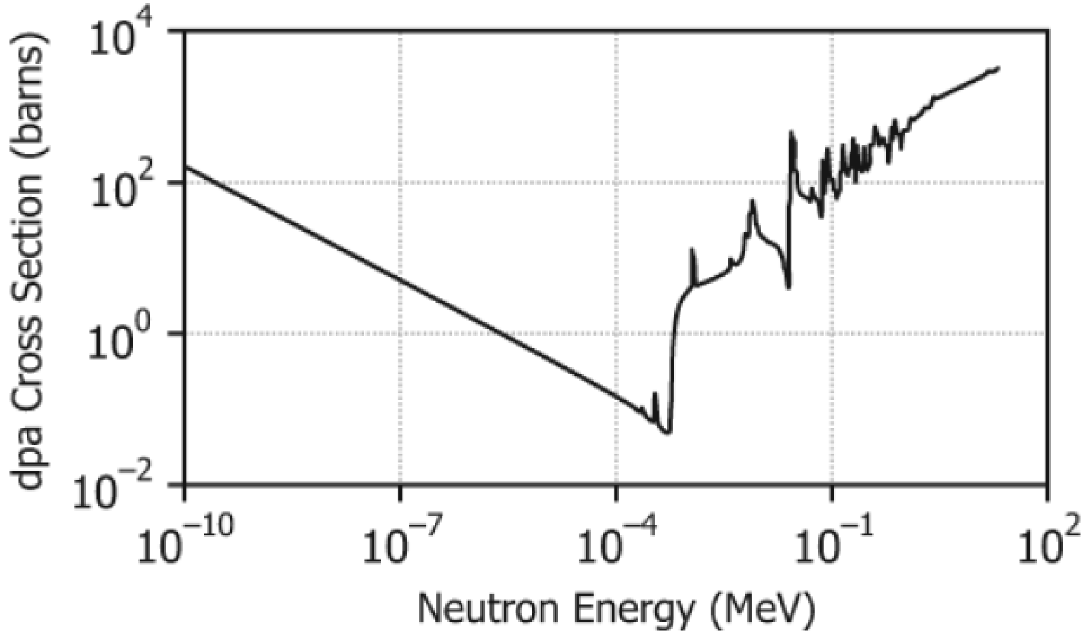
$$\epsilon = [A_2E/(A_1 + A_2)]\ [a/Z_1Z_2e^2], \quad (8)$$

$$a = (9\pi^2/128)^{1/3}\, a_0\, [Z_1^{2/3} + Z_2^{2/3}]^{-1/2}, \quad (9)$$

where a_0 is the Bohr radius, e the electronic charge, Z_1 and Z_2 are the atomic numbers of the projectile and target and A_1 and A_2 are the mass numbers of the two atoms.

Norgett et al, Nuc. Eng. and Design 33 (1975)

In practice, standardized methodologies are available to calculate displacements consistently across environments



ENDF/B-VI-based Iron Displacement Cross Section

Designation: E693 – 23

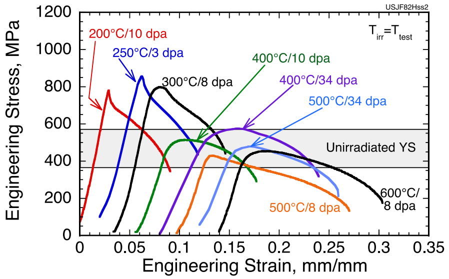
Standard Practice for Characterizing Neutron Exposures in Iron and Low Alloy Steels in Terms of Displacements Per Atom (DPA)[1]

Designation: E521 – 25

Standard Practice for Investigating the Effects of Neutron Radiation Damage Using Charged-Particle Irradiation[1]

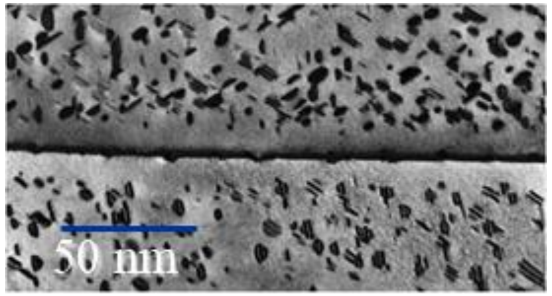
Several code packages exist to calculate displacements from cross section data using ASTM methods

Radiation Damage can Produce Large Changes in Materials



Radiation hardening and embrittlement (<0.4 T_M, >0.1 dpa)

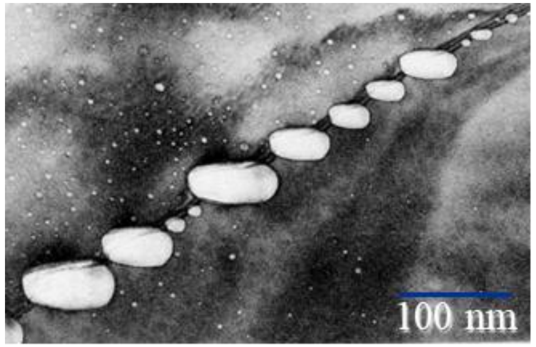
Phase instabilities from radiation-induced precipitation (0.3-0.6 T_M, >10 dpa)



Irradiation creep (<0.45 T_M, >10 dpa)

Volumetric swelling from void formation (0.3-0.6 T_M, >10 dpa)

High temperature He embrittlement (>0.5 T_M, >10-100 appm He)



All of these draw from microstructural evolution

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S.J. Zinkle, Phys. Plasmas 12 (2005) 058101; Zinkle & Busby, Mater. Today 12 (2009) 12

Fundamentally, rate theory equations provide a conceptual framework for understanding microstructure evolution

$$\frac{dC_i}{dt} = K_0 - K_{iv}C_iC_v - \sum^{sinks} k_i^2 D_i C_i$$

Rate of defect concentration change

Production from cascades

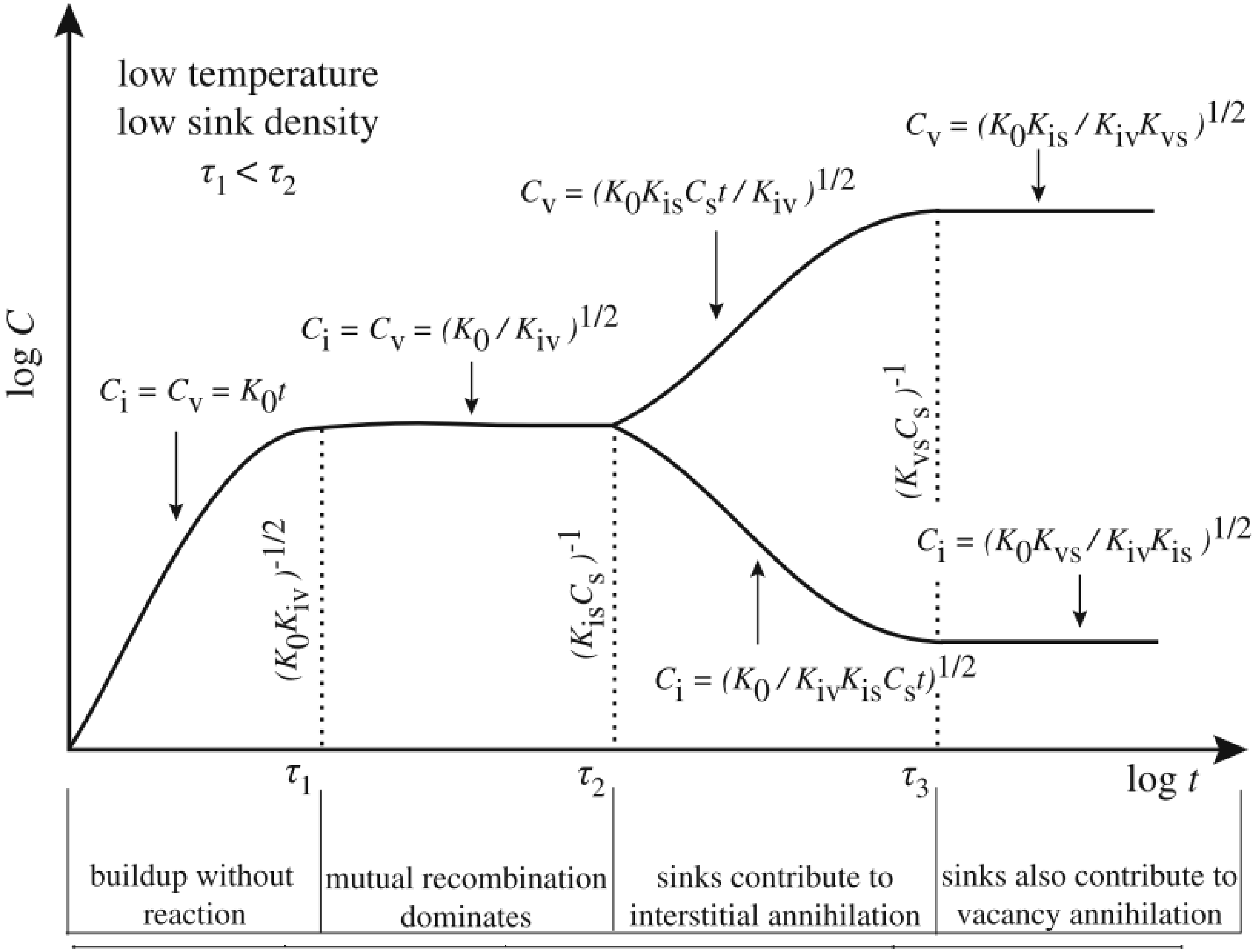
Mutual annihilation

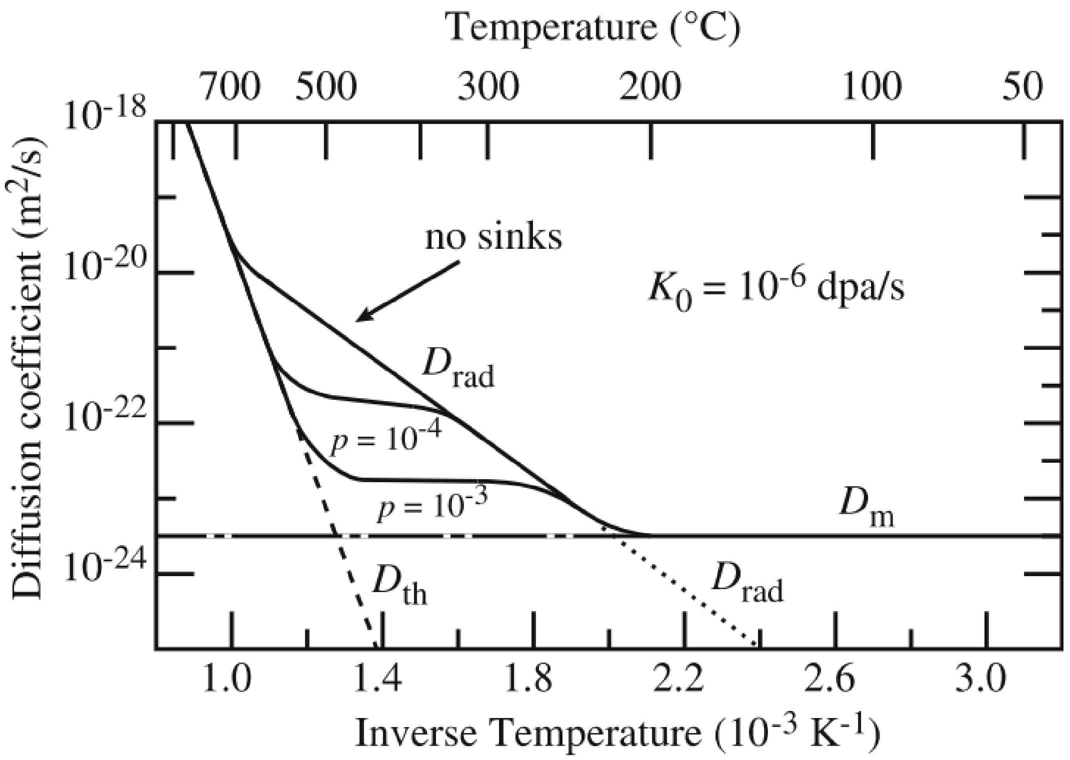
Loss to microstructure sinks

$$\frac{dC_v}{dt} = K_0 - K_{iv}C_iC_v - \sum^{sinks} k_v^2 D_v C_v$$

While microstructure evolution of an individual feature is more complex, they’re all interconnected.

Sink strength and temperature dictate timescales for evolution

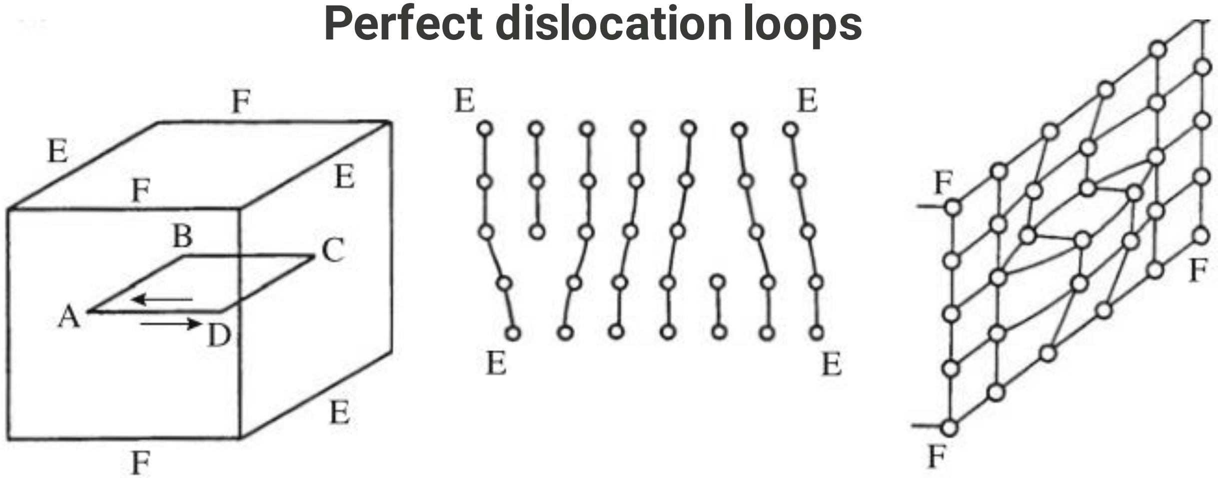


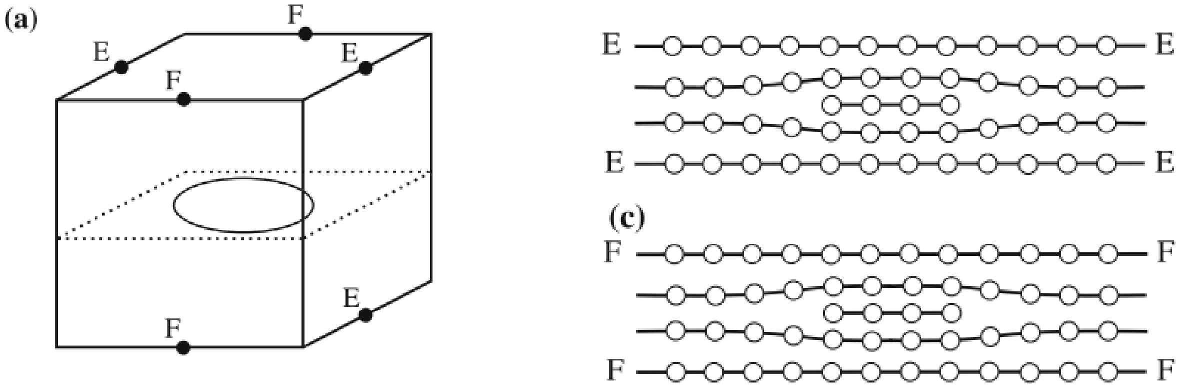


Adapted from *G. S. Was, Fund. of Rad. Mat. Sci., 2nd ed., 2017*.

Defects agglomerate into dislocation loops and lines depending on crystal structure, temperature, composition…

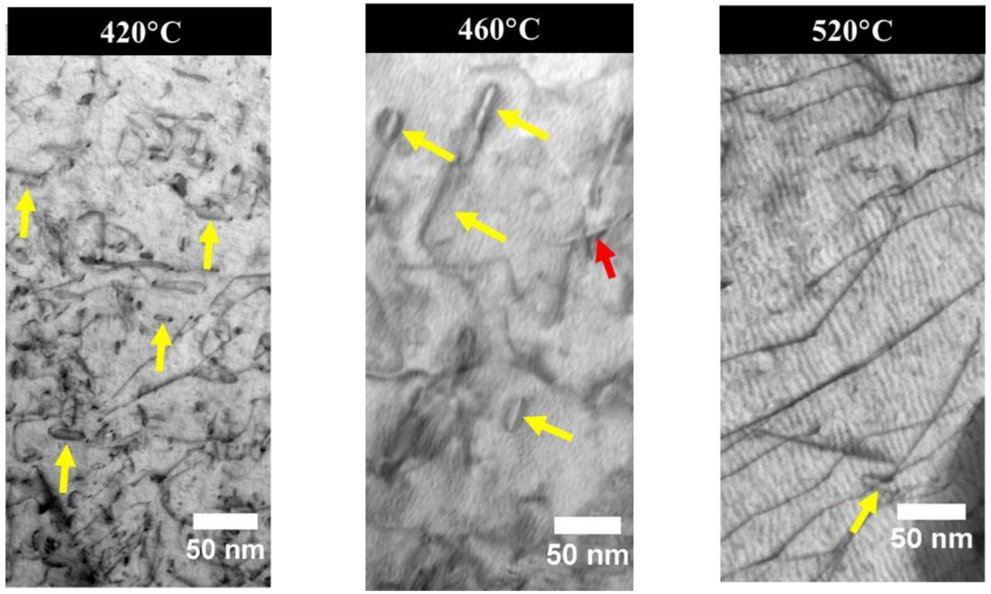
Perfect dislocation loops





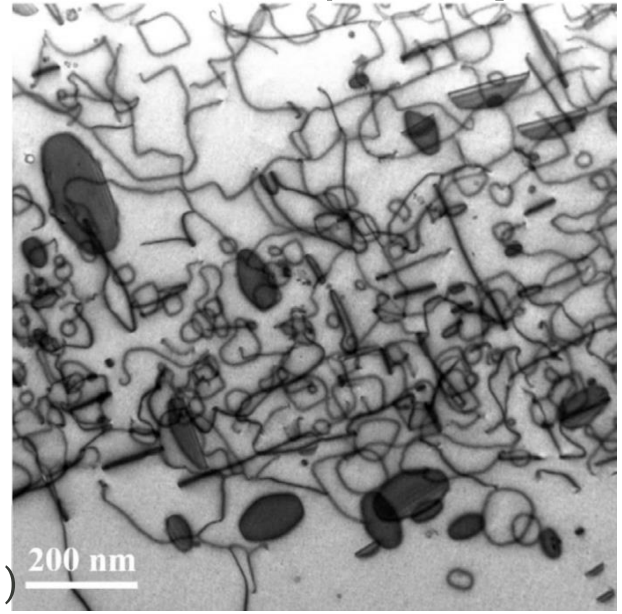
Adapted from *G. S. Was, Fund. of Rad. Mat. Sci., 2nd ed., 2017*.

Ion irradiated T91 steel



S. Taller, Dissertation, University of Michigan 2020

Ion irradiated $Ni_{40}Fe_{40}Cr_{20}$



P. Xiu et al, JNM 544 (2021)

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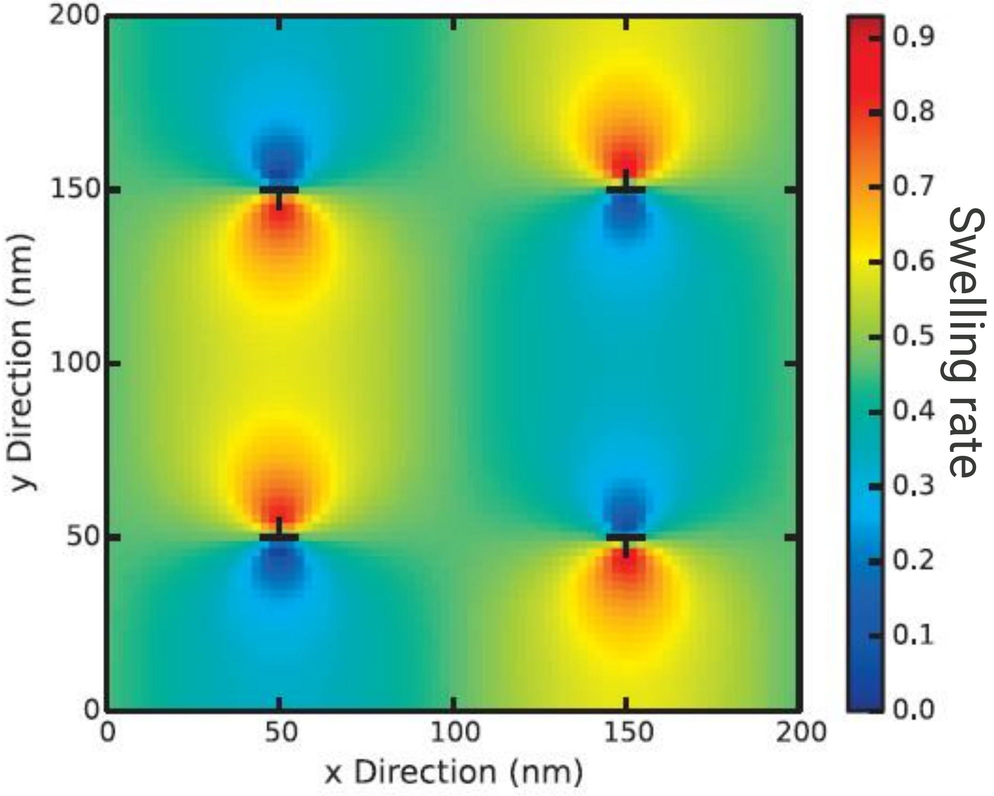
Dislocations generate local stress fields that impact defect migration and clustering



Kohnert, Capolungo J. Mech. Phys. Solids 122 (2019)

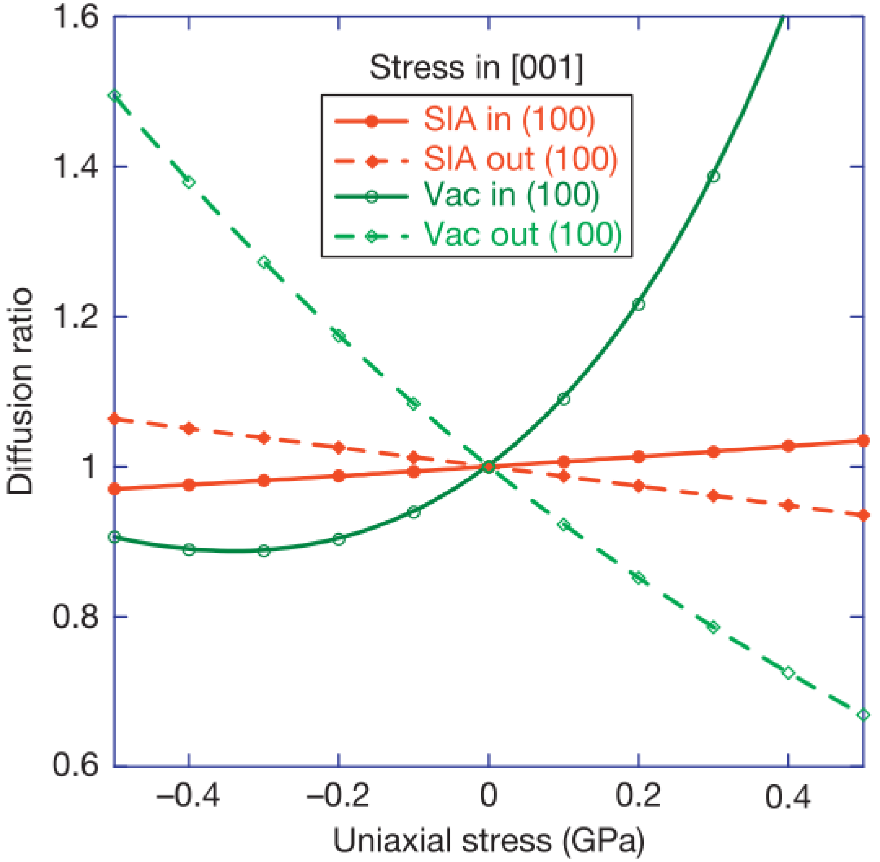


Figure 19 Change of the diffusion coefficients within and perpendicular to (001) crystal planes when a uniaxial stress is applied.

Wolfer, Comprehensive Nucl. Mat. (2020)

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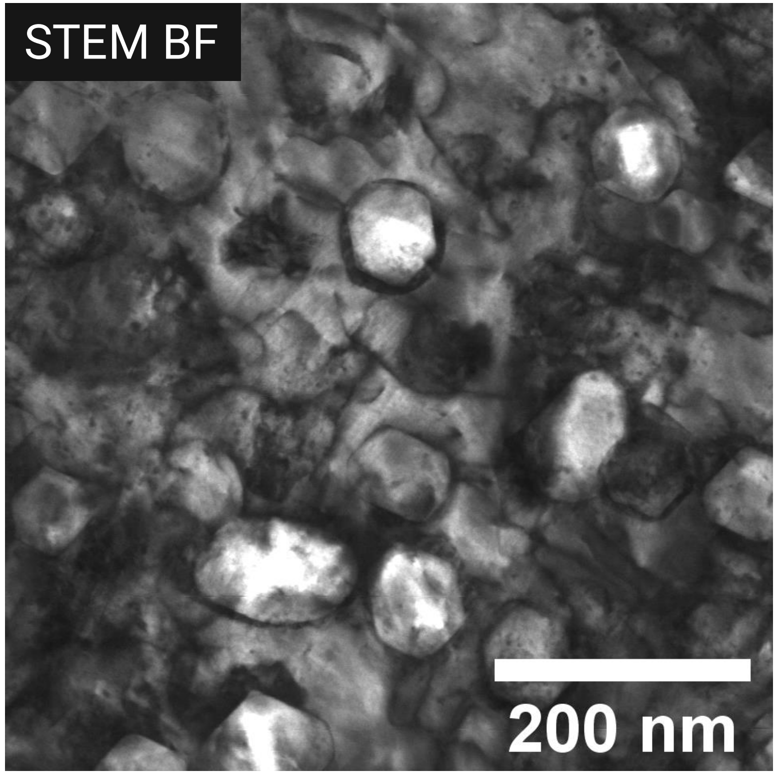
In the range of 0.3 < T_m < 0.6, cavities form from vacancy rich clusters

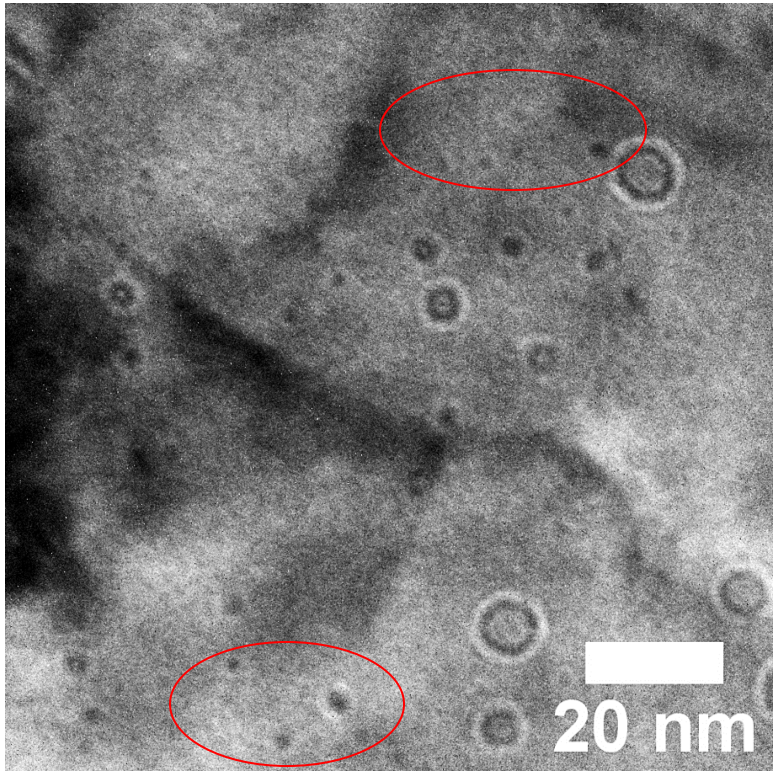
Stainless Steel 316 ion irradiated to 10 dpa near 600°C



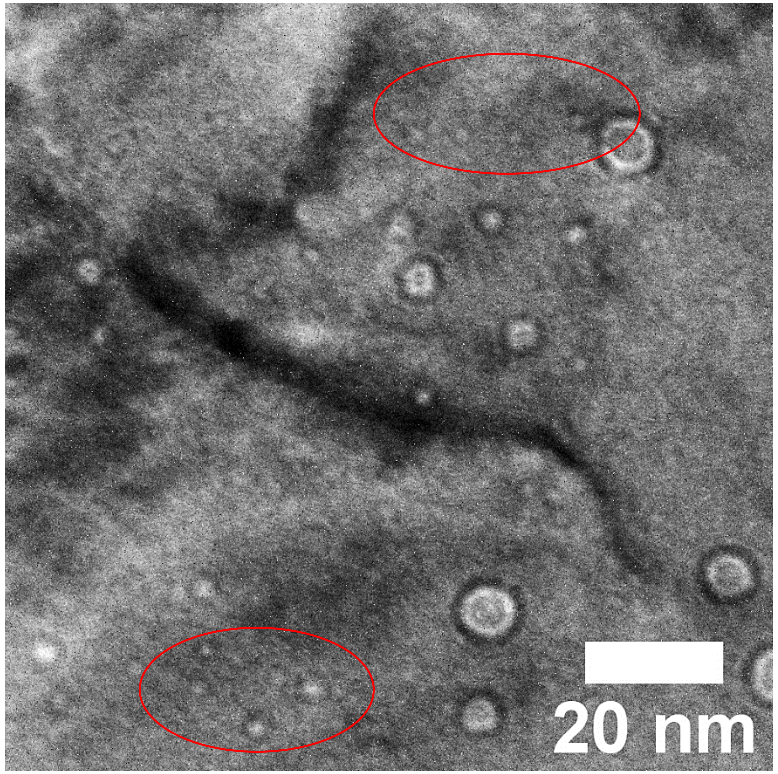
Stephen Taller, unpublished

T91 Steel ion irradiated to ~17 dpa near 445°C

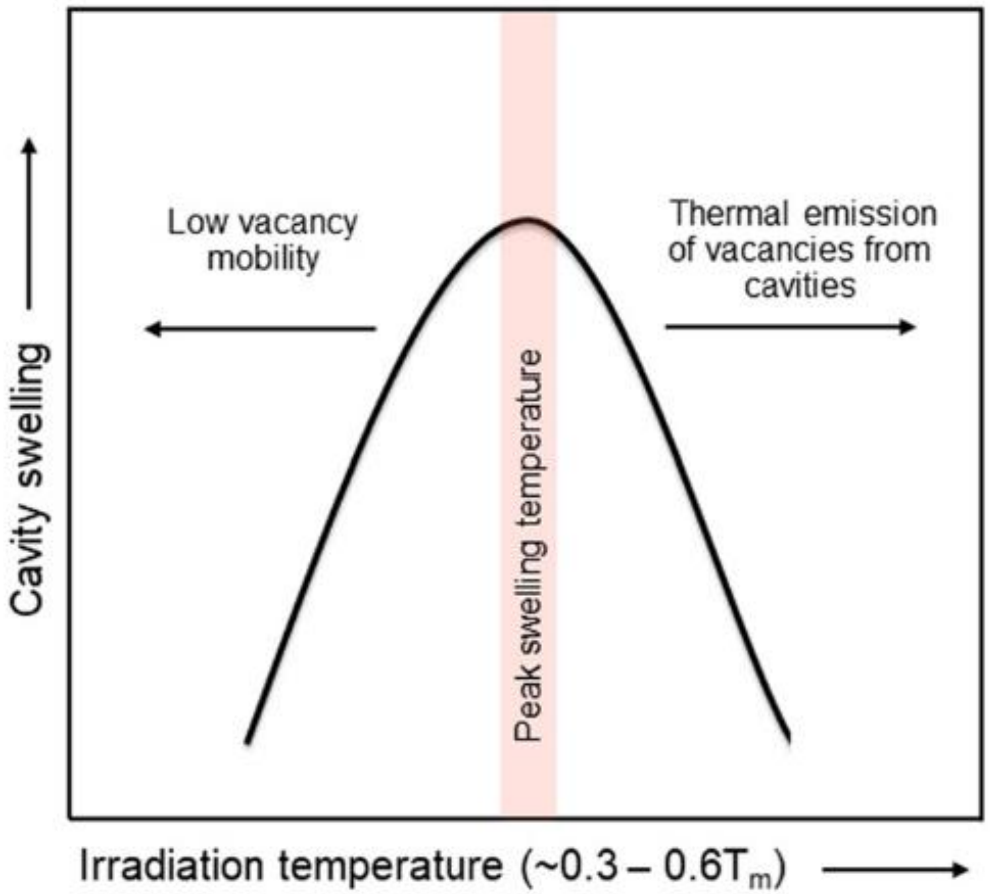
CTEM overfocus (+2μm)



CTEM underfocus (-2μm)



The cavity growth rate equation provides a framework to examine cavity formation



Bhattacharya and Zinkle, Comprehensive Nuclear Materials, 2020

Growth rate of cavity

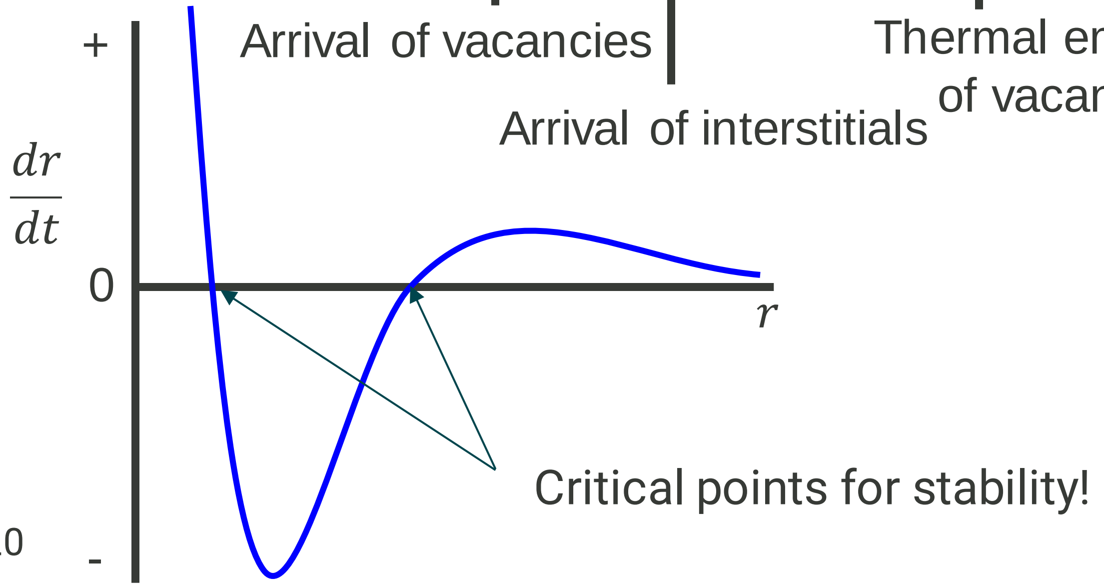
Pressure of He gas from Eq. of State

$$\frac{dr}{dt} = \frac{\Omega}{r}\left[D_v C_v - D_i C_i - D_v C_{v,T} \exp\left(\frac{2\gamma}{r} - p_g\right)\right]$$

Arrival of vacancies

Arrival of interstitials

Thermal emission of vacancies



Cavities have a dependence on temperature through diffusion, helium rate through the gas pressure, p_g and damage rate through the concentration of defects, C_v and C_i and their own size, r.

Helium from transmutation is a (near) uniquely nuclear problem and has its own behaviors and influence on materials

Trapping

Thermal Release

Helium in Lattice

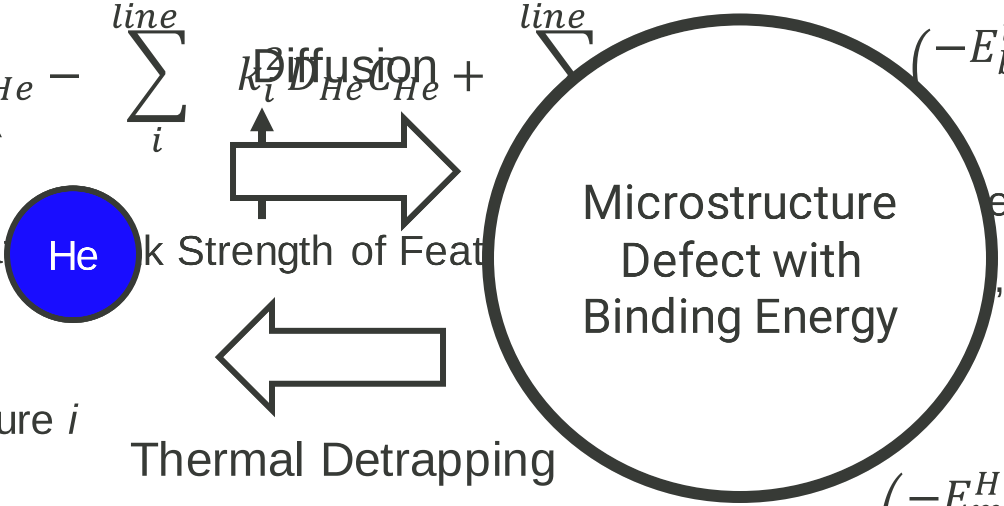
$$\frac{dC_{He}}{dt} = G_{He} - \sum_{i}^{loops,\ bubbles\ line} k_i^2 D_{He} C_{He} + \sum^{loops,\ bubbles,\ line} \ldots \left(-E_b^i/_{kT}\right)$$

He binding energy of *i*

Helium Injection Ra… …k Strength of Feat…

…equency

…, *i*



Helium trapped per feature *i*

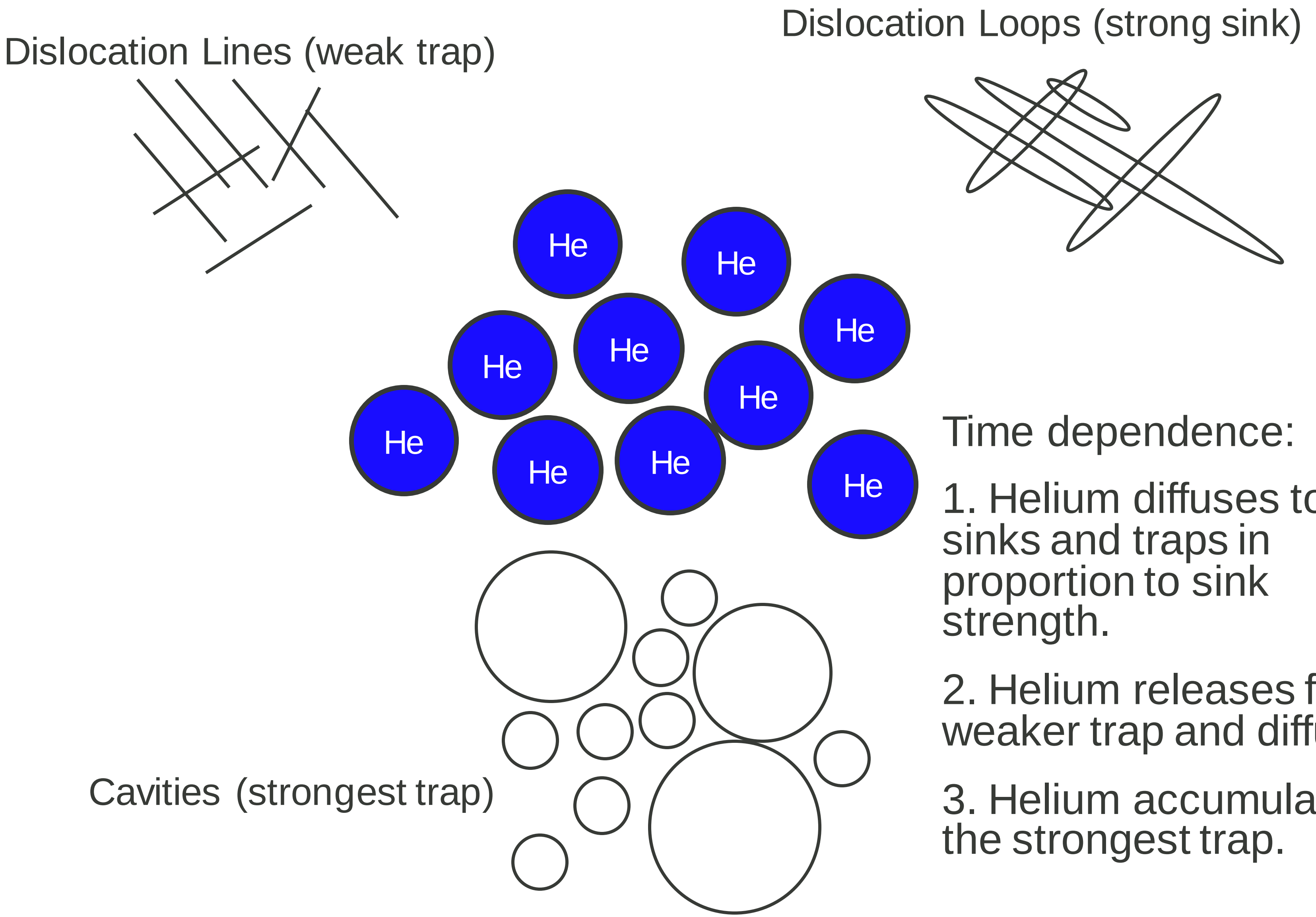
$$\frac{dn_g^i}{dt} = \frac{k_i^2 D_{He} C_{He}}{N_i \Omega} - n_g^i \nu_0 \exp\left(\frac{-E_b^i}{kT}\right)$$

$$D_{He} = D_0 exp\left(\frac{-E_m^{He}}{kT}\right)$$

E_m^{He}~0.064 eV

Stewart et al. JNM 417 (2011)

Helium "flows" from sink to sink based on sink strength and binding energy



Time dependence:

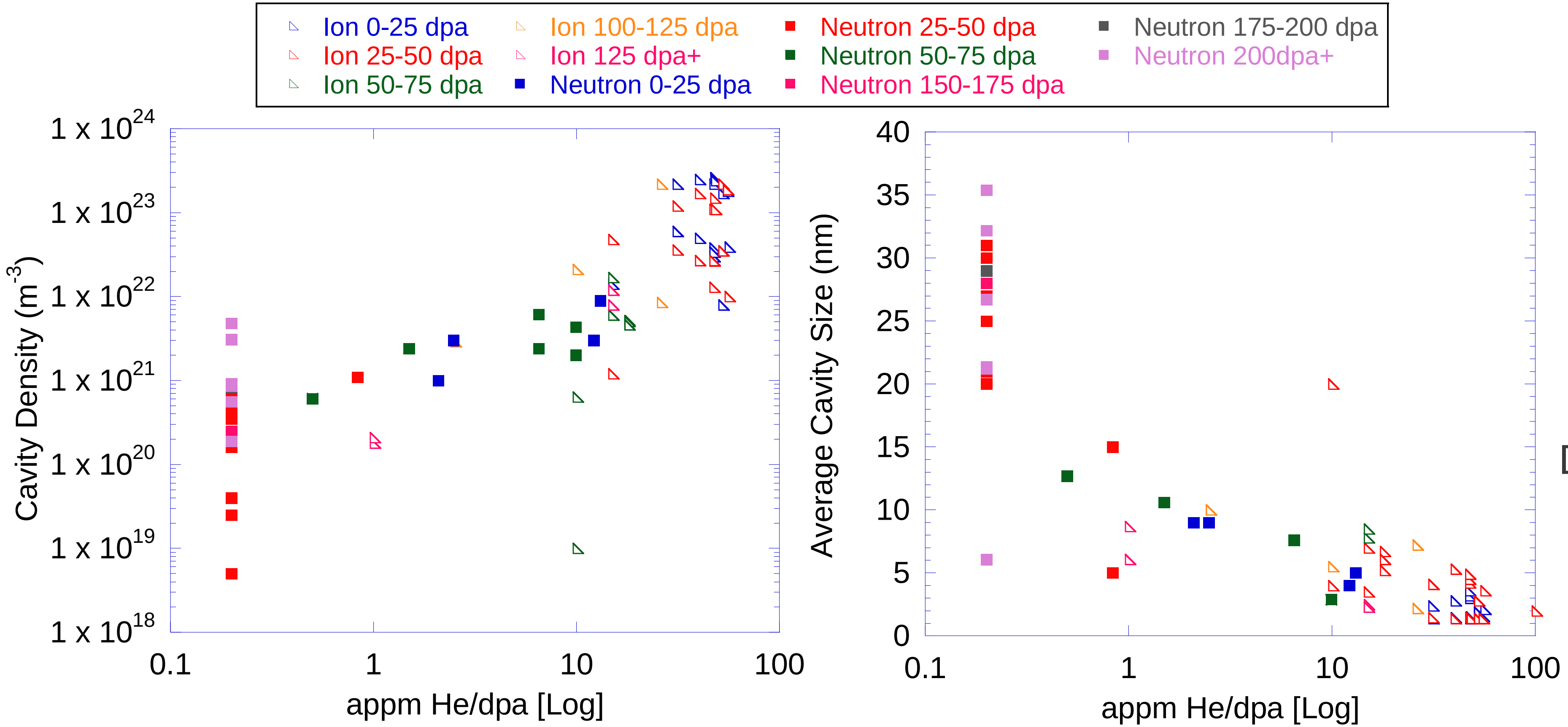
1. Helium diffuses to sinks and traps in proportion to sink strength.

2. Helium releases from a weaker trap and diffuses.

3. Helium accumulates at the strongest trap.

The role of helium is to increase cavity nucleation (primarily)

$T_{neutron}$ = 400-500°C
$T_{dual\ ion}$=360-600°C



S. Taller, Dissertation, University of Michigan, 2020

Reactor Irradiation Literature

J.M. Vitek, R.L. Klueh, JNM. 122–123 (1984)
N. Hashimoto, R.. Klueh, JNM. 305 (2002)
E. Wakai, et al, JNM. 283–287 (2000)
E. Wakai, et al, JNM. 307–311 (2002)
J.J. Kai, R.L. Klueh, JNM. 230 (1996)
A. Kimura, et al JNM. 191–194 (1992)
B.H. Sencer, et al, JNM. 414 (2011)
D.S. Gelles, JNM. 237 (1996)
J. Van Den Bosch, et al, JNM. 440 (2013)

Dual Ion Irradiation Literature

K. Asano, et al, JNM. 157 (1988)
Y.E. Kupriiyanova, et al, JNM. 468 (2015)
H. Ogiwara, et al, JNM. 307–311 (2002)
E. Wakai, et al, JNM. 318 (2003)
S. Hiwatashi, et al, JNM. 179–181 (1991)
T. Yamamoto, et al, JNM. 449 (2014)
A. Bhattacharya, et al, Acta Mater. 108 (2016)
D. Brimbal, et al, Acta Mater. 61 (2013)

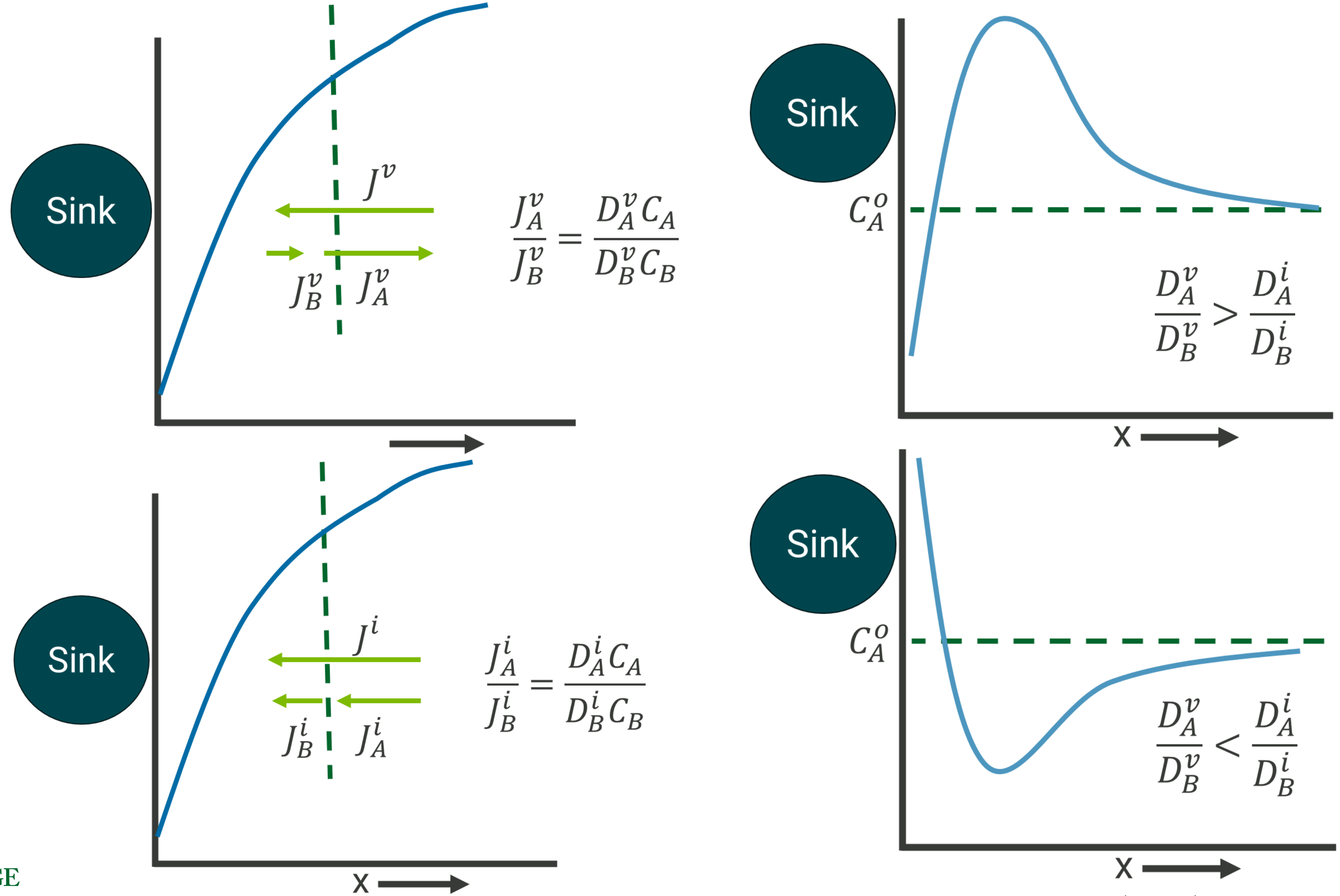
Cavity density increases with He/dpa.

Overall, average cavity diameter decreases with appm He/dpa. However, without the cavity size distributions, it is unclear what cavities are bubbles, stabilized by helium, or how they grow to larger sizes.

So far we've seen the basic defect clusters based on interstitials and vacancies.

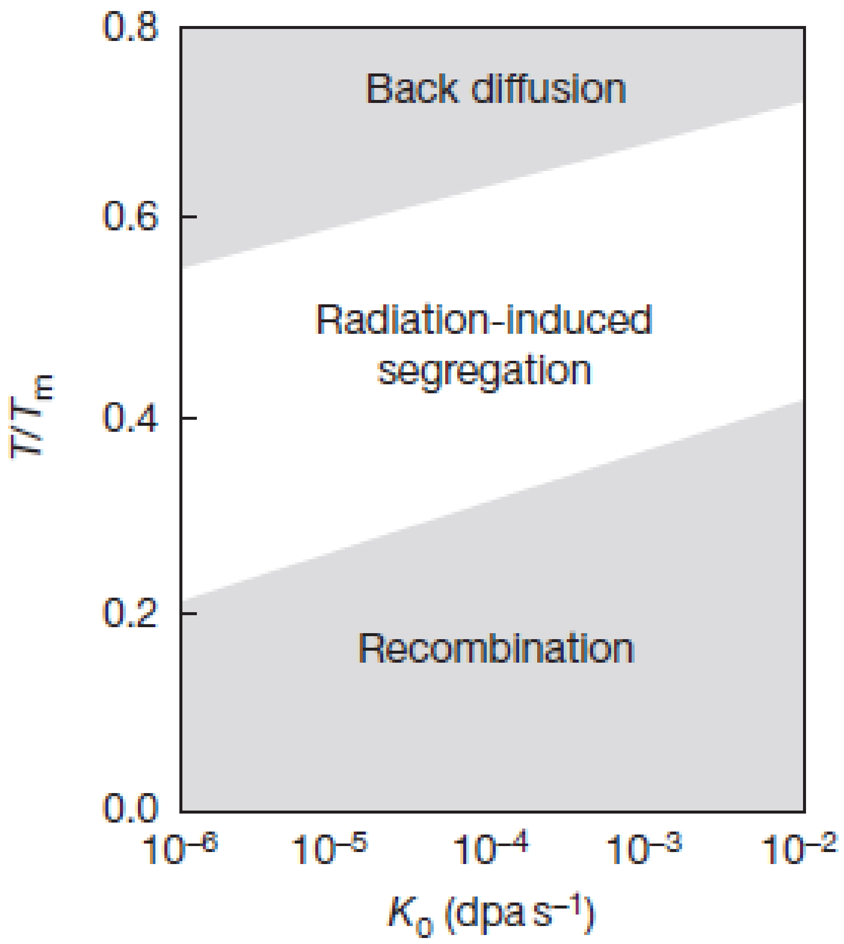
What happens when individual elements are considered?

Differences in atomic diffusivity with enhanced point defect concentrations leads to radiation induced segregation



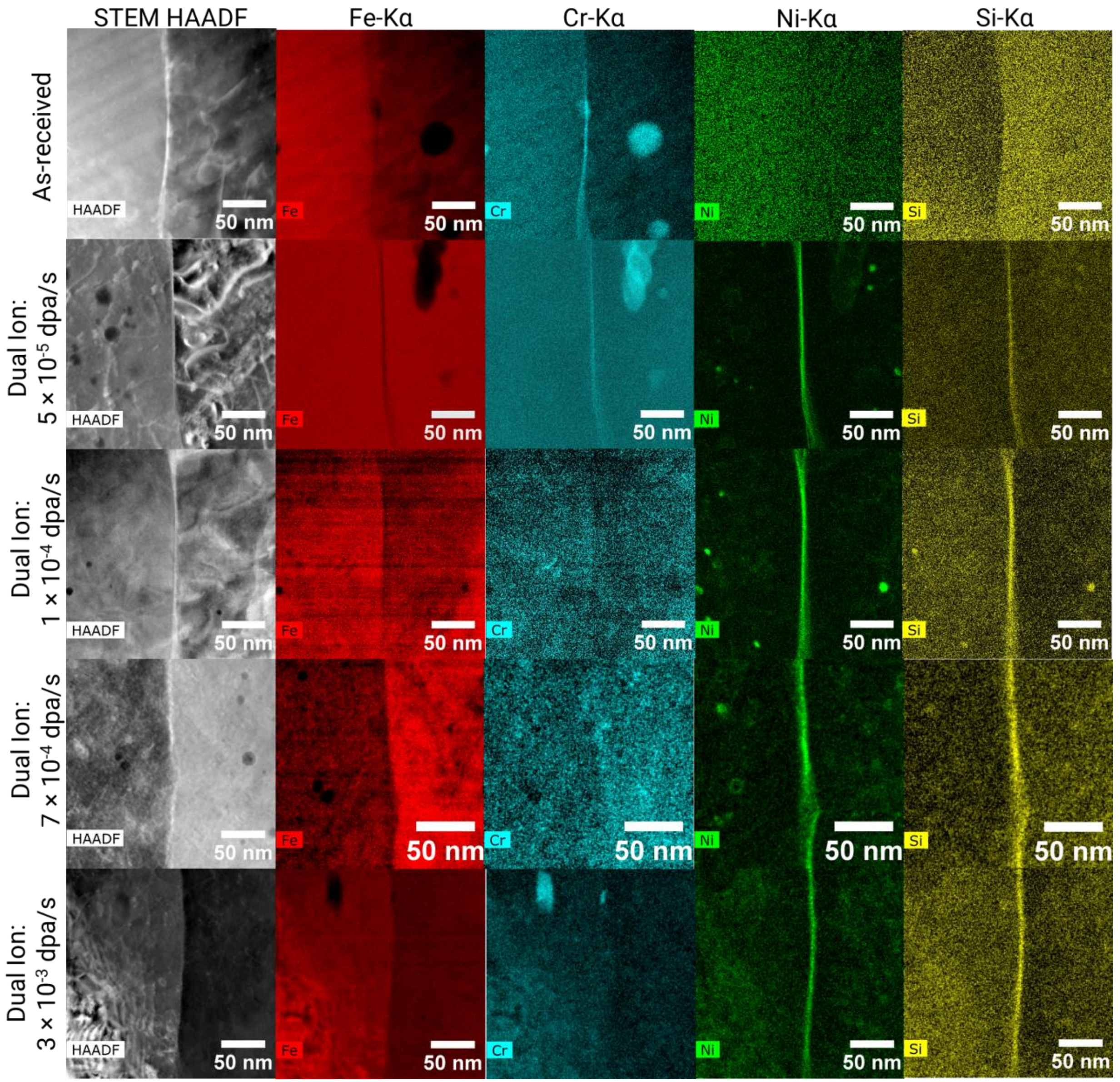
Adapted from P. R. Okamoto and L. E. Rehn, JNM 83 (1979)

Radiation induced segregation is temperature and dose rate dependent



Nastar, Soisson, Comp. Nuc. Mat. (2012)

T91 Steel, 445°C, ~17 dpa



Adapted from S. Taller et al, JNM 563 (2022)

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Precipitation is a complex mix of thermodynamics and kinetics

Phase Stability

Radiation Induced Precipitation

New phases form that otherwise are prohibited

Radiation Enhanced Precipitation

Enhanced point defect concentrations and mobility increase kinetics

Radiation Ordering/Disordering

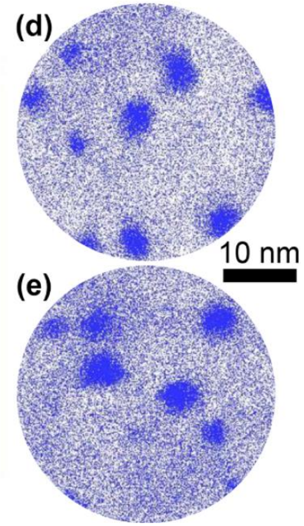
Loss of long range order (LRO) or formation of short range order (SRO) due to irradiation

Recoil Dissolution

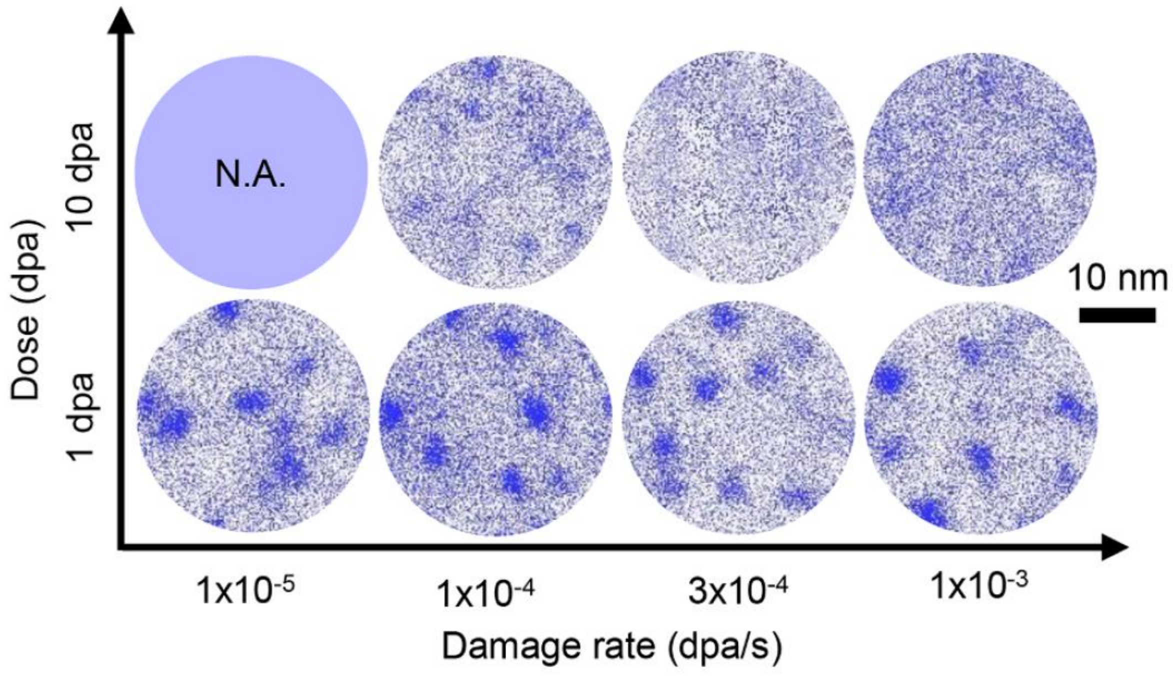
Damage cascades ballistically release solutes to the surrounding lattice

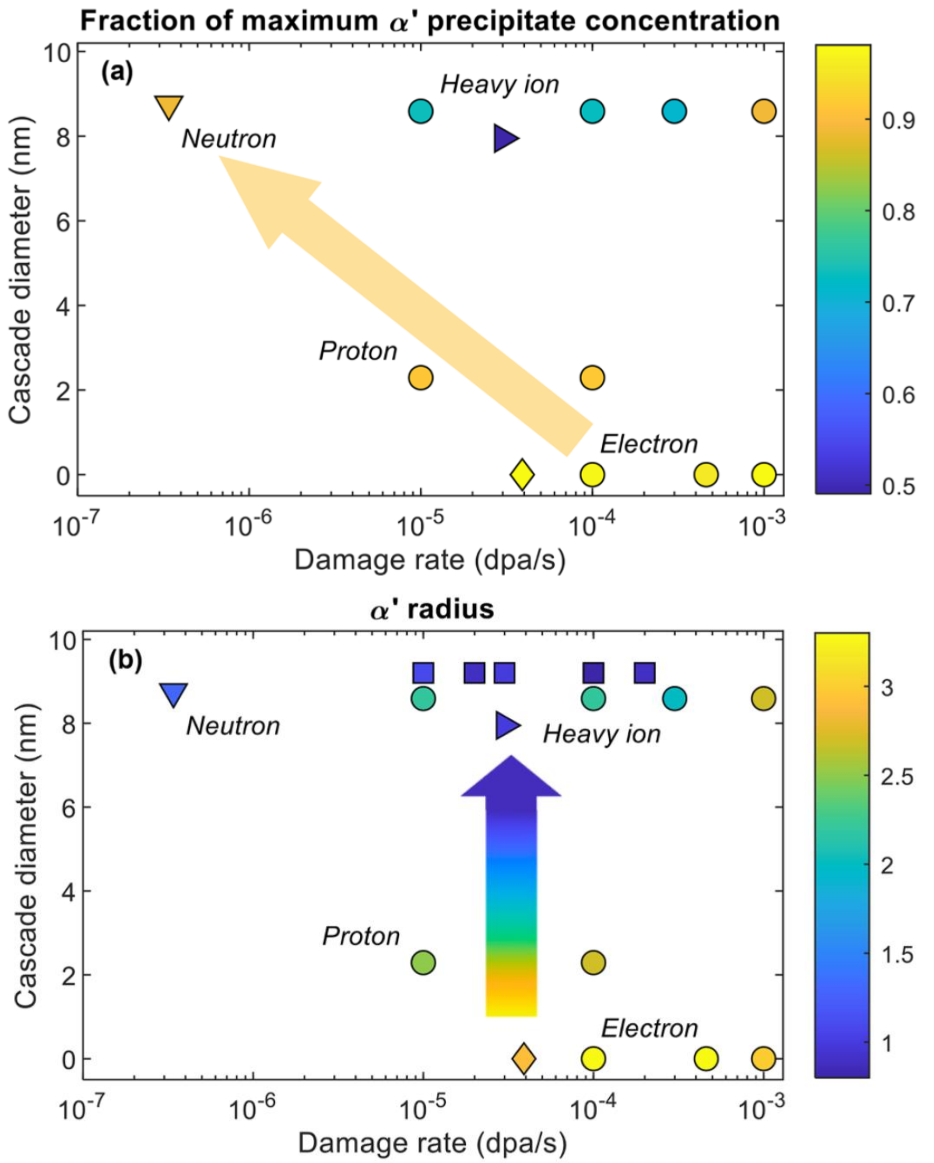
Case Study: α’ precipitates in Fe-Cr binary alloys

Initial conditions
Cr-rich precipitates
present



Heavy ion irradiation at 400°C





Temperature range: ~290-400°C, ~1 dpa
Fe-(15,18)Cr alloys

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K. Thomas and G. S. Was, JNM 585 (2023)

Incoherent precipitate stability can be strongly influenced by radiation

vacancies

Solute supersaturation

$$\Delta G_p^0 = -nkT \ln S_v + (36\pi\Omega^2)^{1/3}\gamma x^{2/3} - xkT \ln S_x + \frac{\Omega E x(\delta - n/x)^2}{9(1-v)}$$

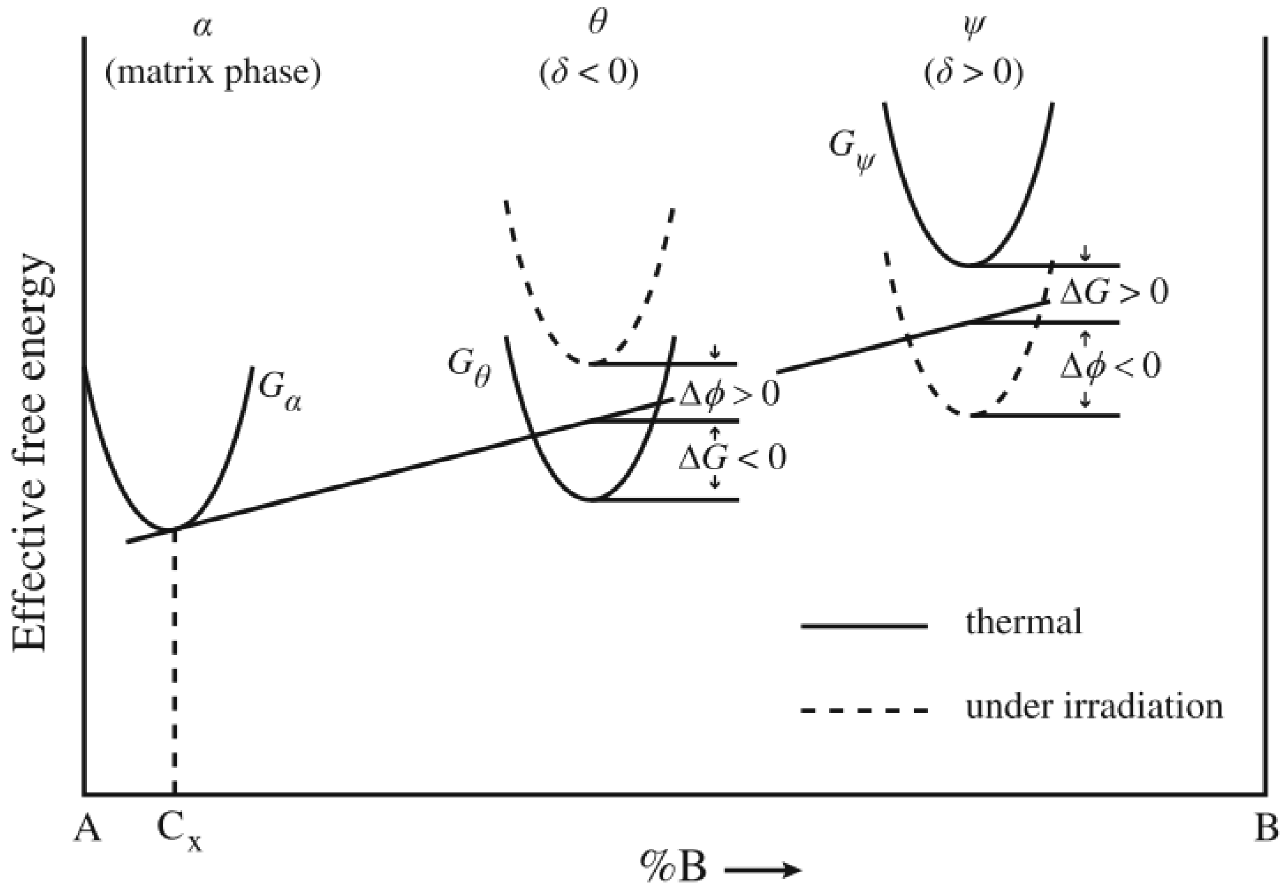
Vacancy supersaturation

solute atoms

$$\Delta\phi = -kT \ln S_x,$$

Thermally Stable

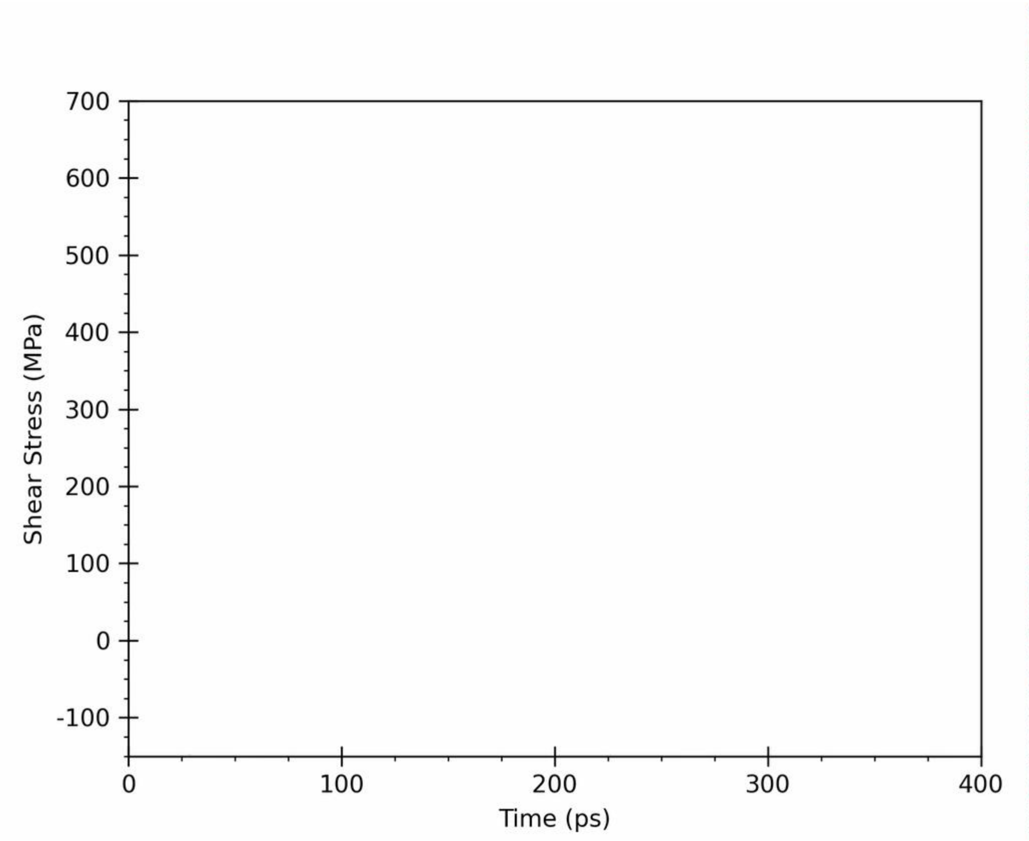
Thermally unstable

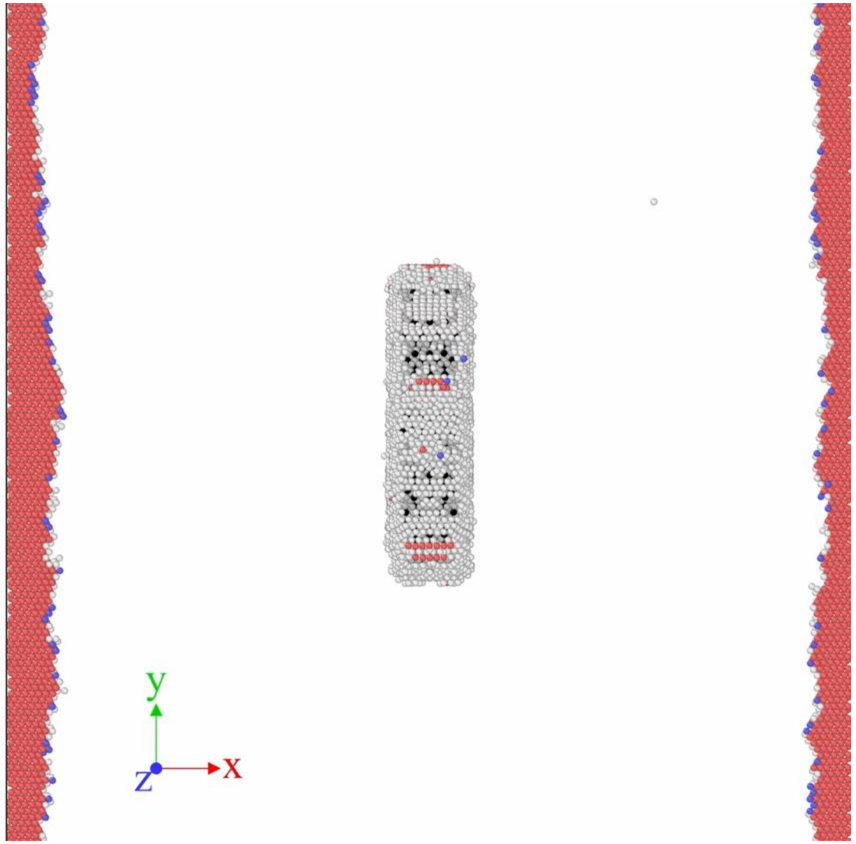


Adapted from *G. S. Was, Fund. of Rad. Mat. Sci., 2nd ed., 2017*.

Precipitates and defect clusters act as barriers to dislocation motion

γ″ precipitate in Nickel



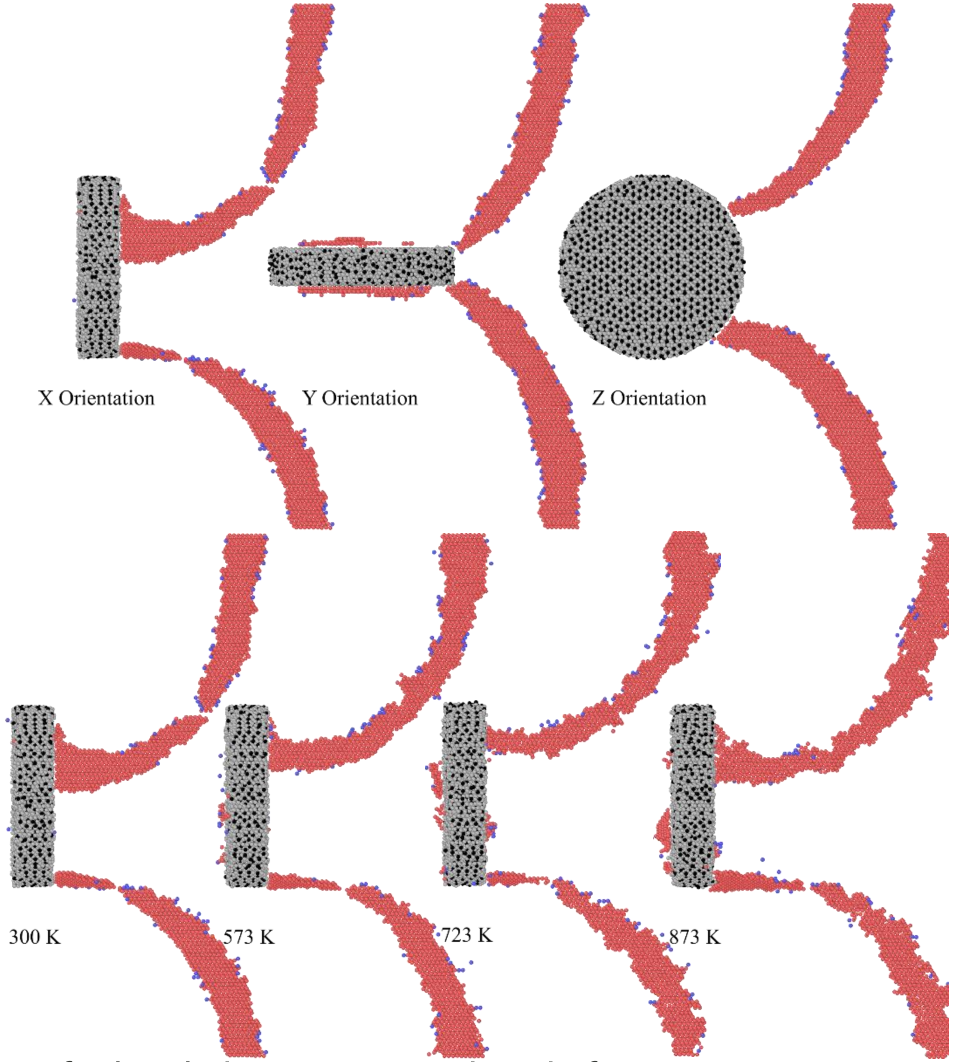


Visualization of edge dislocation structure mobility and shear stress behavior during dislocation MD simulation for X-oriented, 10nm, 300K simulation

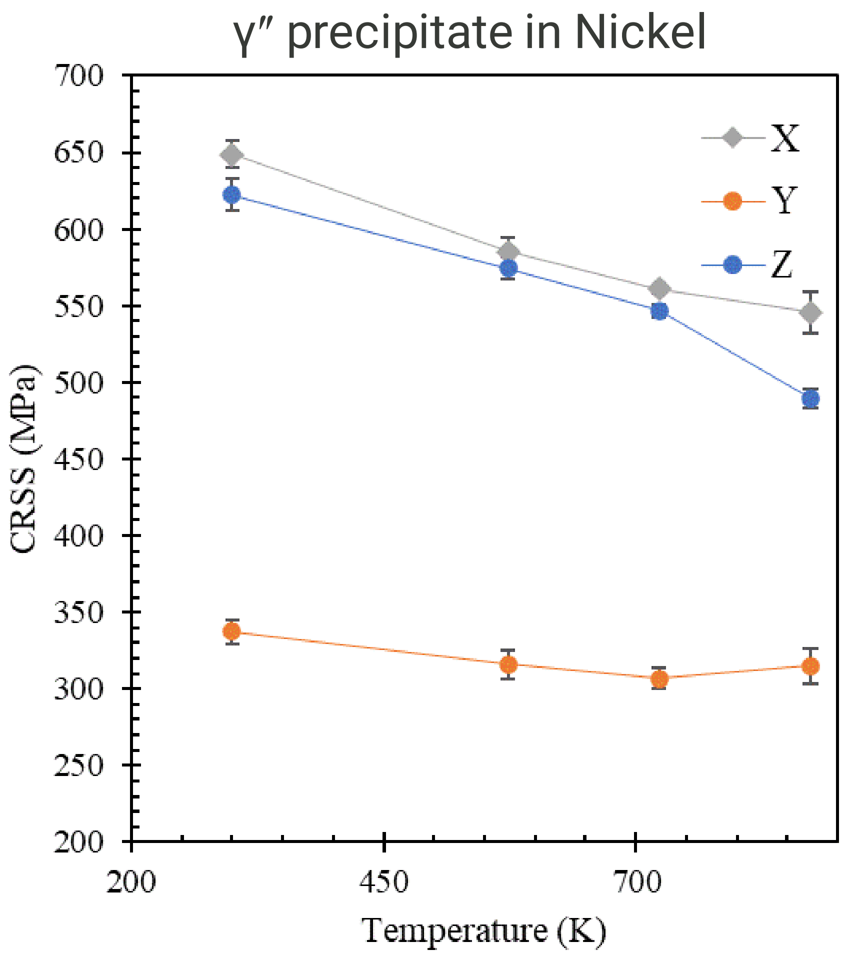
Provided by L. Metzger, Virginia Tech

Defect cluster barrier strengths can depend on orientation relationships and temperature

Provided by L. Metzger, Virginia Tech



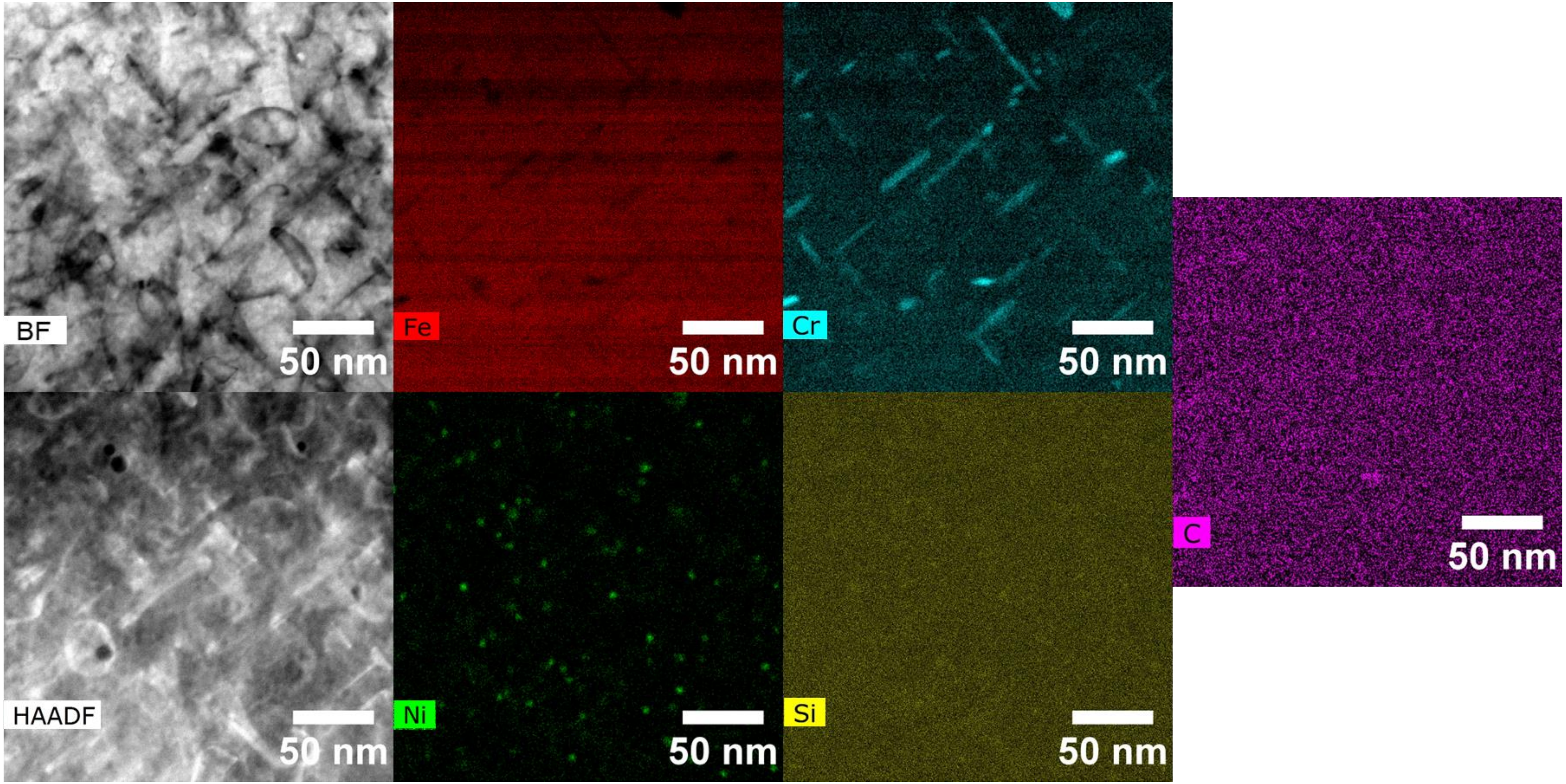
Visualization of edge dislocation critical angle for 10 nm precipitates. (*Top*) varying precipitate orientations ; (*Bottom*) varying temperatures for X-oriented precipitates



Comparison of simulation maximum shear stresses and simulation temperatures for varying precipitate orientations.

Segregation can lead to enrichment on dislocations

T91 Steel Ion Irradiated at 445°C to ~17 dpa



Stephen Taller, unpublished

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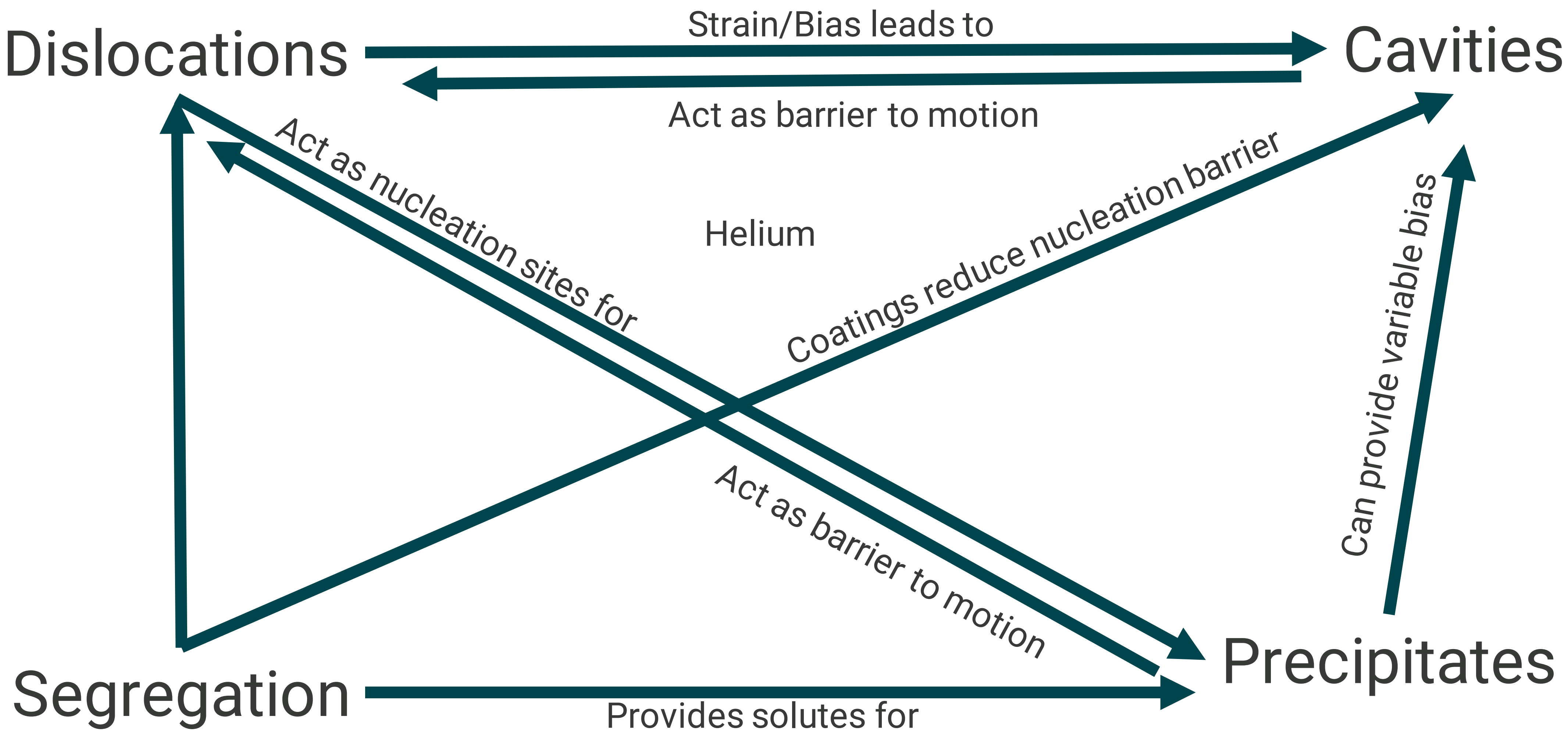
Microstructure evolution is holistic!

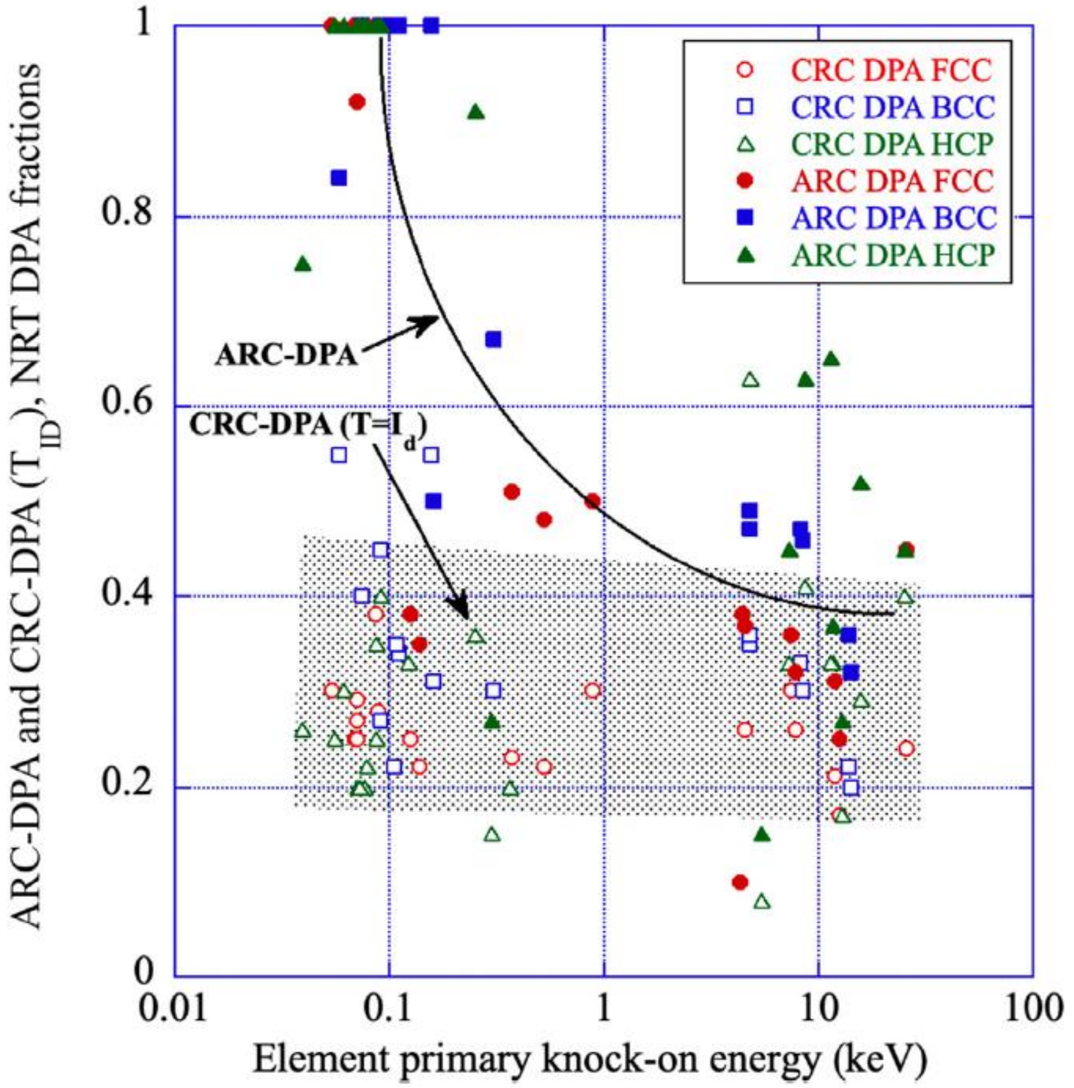


Which DPA measure is the right one?

- Recall NRT-dpa is primarily about the damage event, NOT the outcome of the damage.

- Two additional metrics are proposed:

- ARC-dpa (athermal recombination correlated)
- CRC-dpa (correlated recombination-corrected dpa, “core-dpa”)



S. Zinkle and R. Stoller, JNM 577 (2023)

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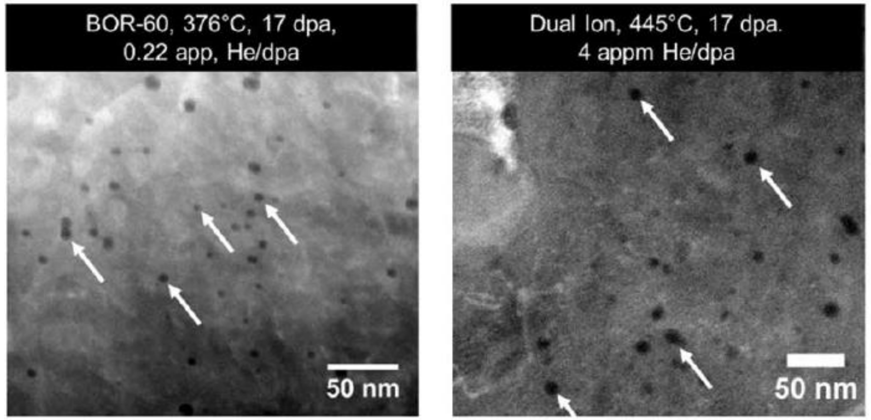
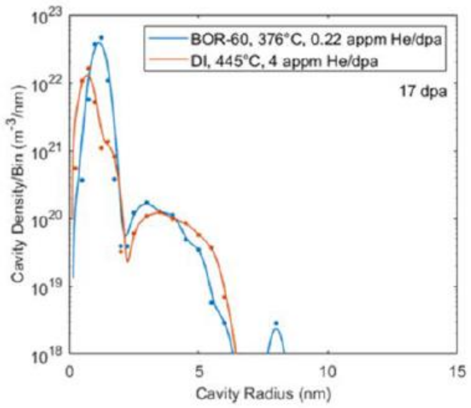
How can we simulate one environment with another?

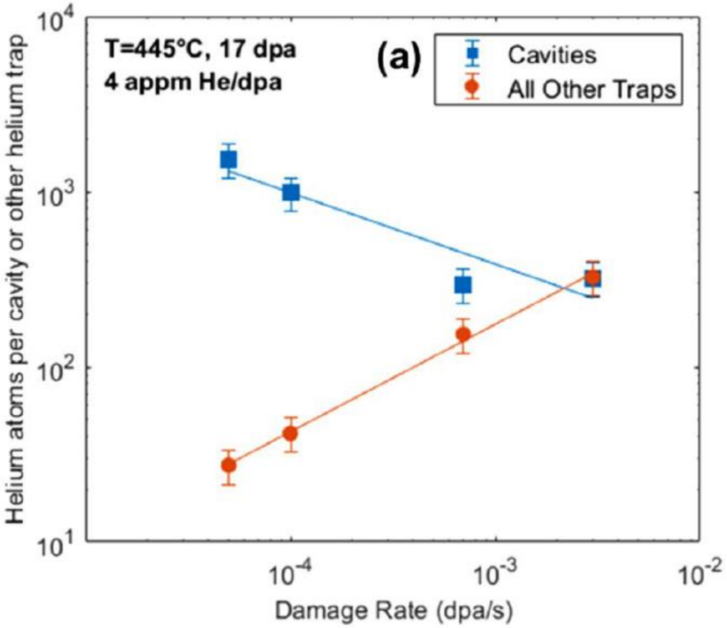
$$T_2 - T_1 = \frac{\dfrac{kT_1^2}{E_v^m + 2E_v^f}\ln\left(\dfrac{G_2}{G_1}\right)}{1 - \dfrac{kT_1}{E_v^m + 2E_v^f}\ln\left(\dfrac{G_2}{G_1}\right)},$$

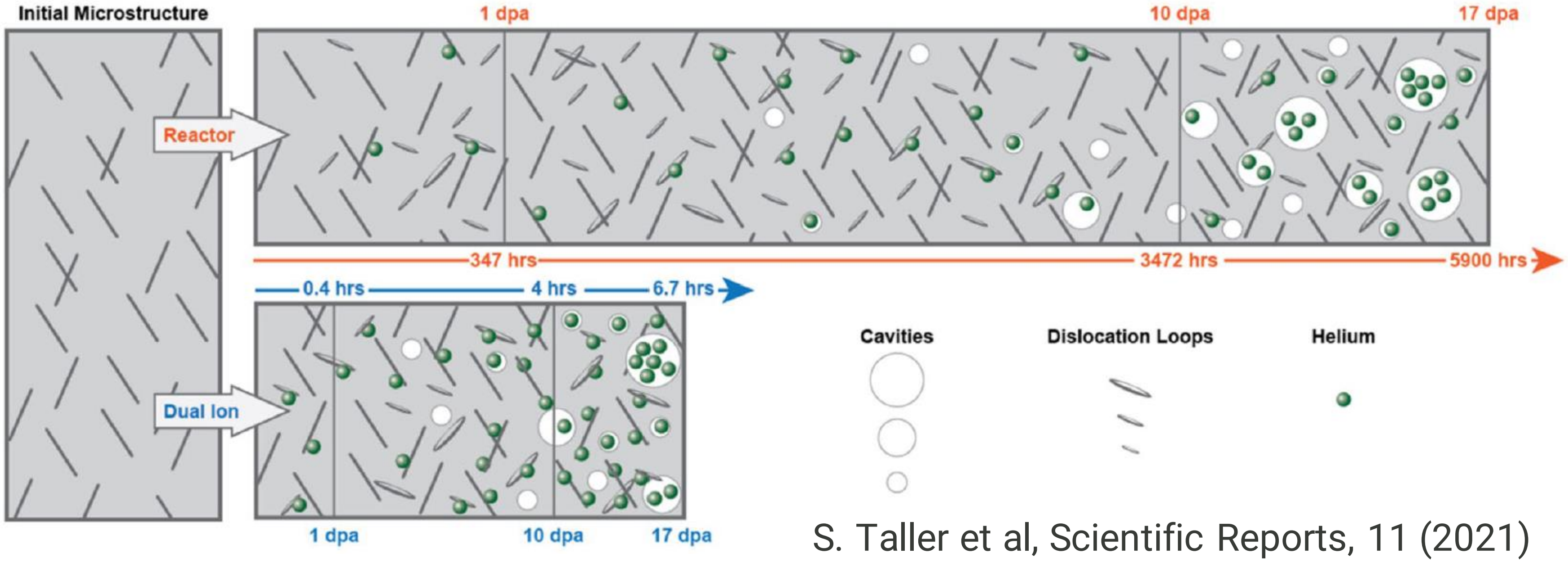
Mansur, JNM 206 (1993)





ΔT~60-70°C across ~10^3 in dpa rate





S. Taller et al, Scientific Reports, 11 (2021)

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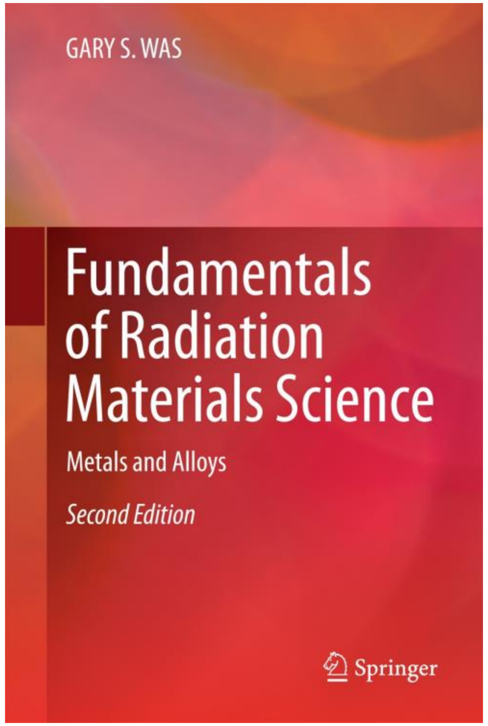
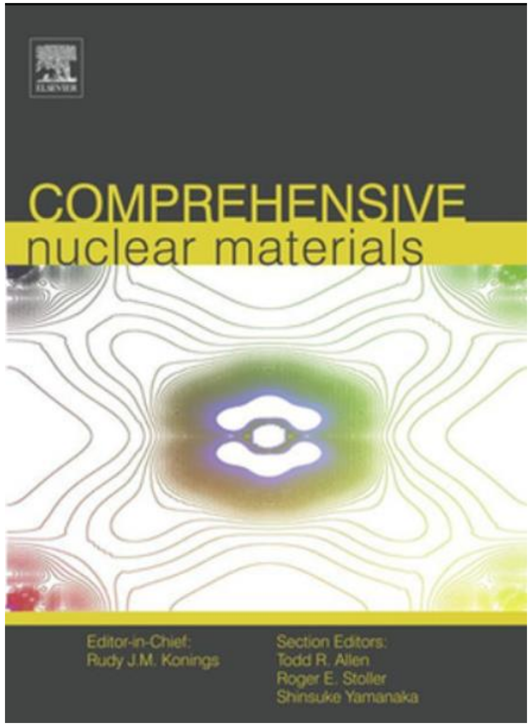
Short answer: It depends and it's complicated.

For more information there's great reference materials

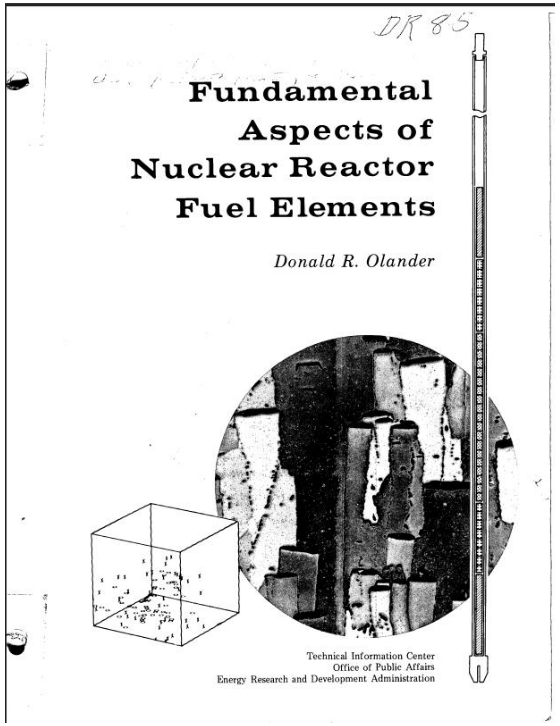
For theory and equations:



For material specific reviews:



For more fuels related items:
Also free on OSTI



https://doi.org/10.2172/7343826

Focused on LWR materials:

Summary

- Radiation damage is a complex set of interconnected phenomena (and that’s why its fun to research!)
- Picosecond (timescale) events influence large length scale properties!
- There are many independent and synergistic effects to consider when planning or designing irradiation test campaigns.

- NSUF scientists are available to help plan out experiments across the 20+ facilities.