Voids II+

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Void Nucleation Theory: Graphical depiction

$$\Delta G_n^0 = \underbrace{-nkT \cdot \ln(S_v)}_{\text{entropy of mixing (negative)}} + \underbrace{(36\pi\Omega^2)^{1/3}\gamma n^{2/3}}_{\text{formation of interfacial surface}}$$

$\sim 4\pi r^2 \gamma$

↑ as decrease

$\boxed{\uparrow S_v \propto 1/\text{nucl.}}$

↑

Temp

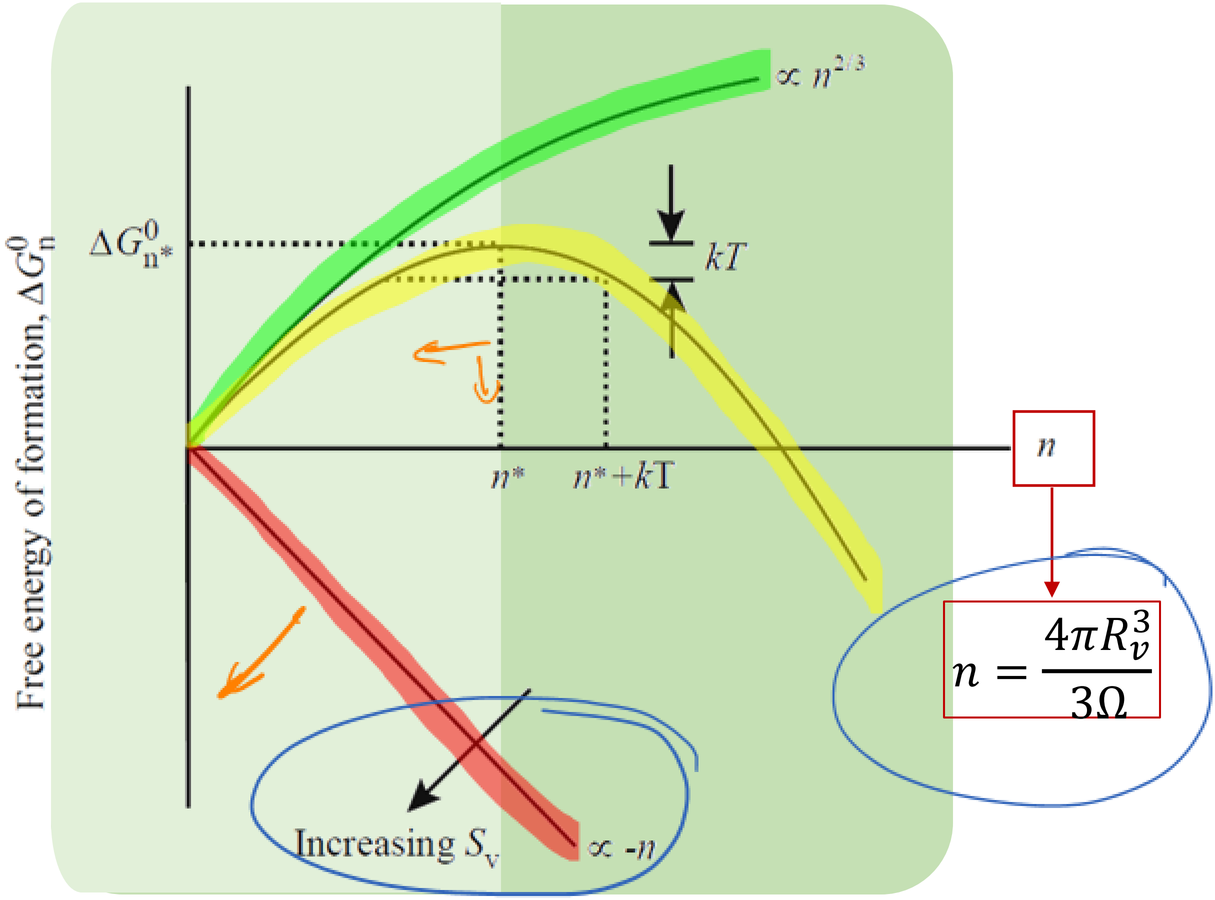
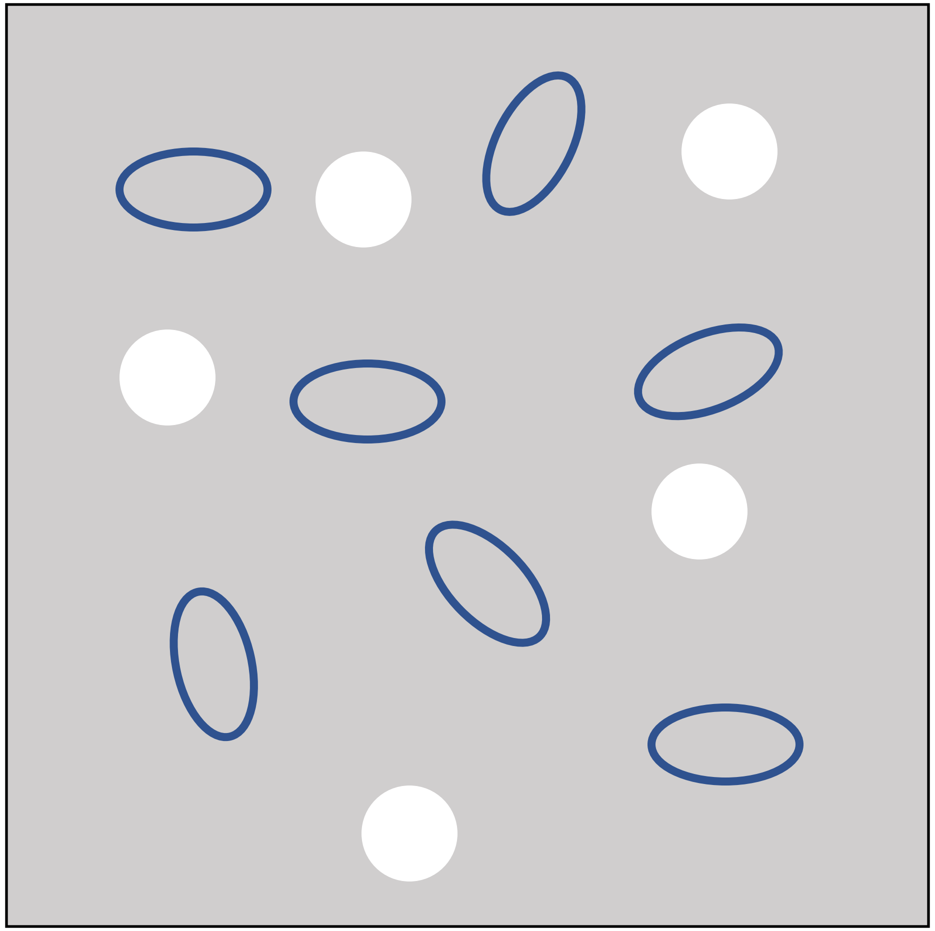


Fig. 8.2 Schematic illustration of ΔG_n^0, the free energy of formation of a spherical void consisting of n vacancies and the effect of thermal fluctuations on the critical size void embryo

Full derivations and discussion in Was 8.1

Void Growth – Simple Model



Let's assume a material with nucleated voids and dislocation loops only, then the total sink strength can be defined as (from Table 5.2 Was):

$$k_v^2 = z_v \rho_d + 4\pi R \rho_V$$

$$k_i^2 = z_i \rho_d + 4\pi R \rho_V$$

Void Growth

- For void growth, we need to know the net flux of vacancies to a void embryo. The net rate is thus a combination of the fluxes of interstitials and vacancies to a *nucleated* void, where:

$$J_{net}^{V} = J_{v}^{V} - J_{i}^{V} = 4\pi R\Omega D_{v}(C_{v} - C_{v}^{V}) - 4\pi R\Omega D_{i}\left(C_{i} - C_{i}^{V}\right)$$

$$J_{net}^{V} = {}^{dV}/_{dt} = 4\pi R\Omega(D_{v}C_{v} - D_{i}C_{i})$$

$${}^{dR}/_{dt} = \dot{R} = \frac{\Omega}{R}(D_{v}C_{v} - D_{i}C_{i})$$

$$C_{v}^{V} = C_{v}^{0} exp\left(\frac{2\gamma\Omega}{Rk_{b}T}\right)$$

Void Growth

$$\frac{\partial C_v}{\partial t} = K_0 - K_{iv}C_iC_v - K_{vs}C_vC_s = 0$$

$$C_v = \frac{-K_{is}C_s}{2K_{iv}} + \left[\frac{K_0K_{is}}{K_{iv}K_{vs}} + \frac{K_{is}^2C_s^2}{4K_{iv}}\right]^{1/2}$$

Next slide

$$dR/dt = \dot{R} = \frac{\Omega}{R}(D_vC_v - D_iC_i)$$

$$C_i = \frac{-K_{vs}C_s}{2K_{iv}} + \left[\frac{K_0K_{vs}}{K_{iv}K_{is}} + \frac{K_{vs}^2C_s^2}{4K_{iv}}\right]^{1/2}$$

Next slide

$$\frac{\partial C_i}{\partial t} = K_0 - K_{iv}C_iC_v - K_{vs}C_iC_s = 0$$

Void Growth

Remember:

$$C_v = \frac{-K_{is}C_s}{2K_{iv}} + \left[\frac{K_0 K_{is}}{K_{iv}K_{vs}} + \frac{K_{is}^2 C_s^2}{4K_{iv}}\right]^{1/2}$$

$$C_i = \frac{-K_{vs}C_s}{2K_{iv}} + \left[\frac{K_0 K_{vs}}{K_{iv}K_{is}} + \frac{K_{vs}^2 C_s^2}{4K_{iv}}\right]^{1/2}$$

and

$$k_{jx}^2 = \frac{K_{jx}C_x}{D_j}$$

and

$$k_v^2 = z_v\rho_d + 4\pi R\rho_V$$

$$k_i^2 = z_i\rho_d + 4\pi R\rho_V$$

You can now pull all three equations above together to get:

$$C_v = \frac{D_v(4\pi R\rho_v + z_v p_d)}{2K_{iv}}\left(\sqrt{1+\eta} - 1\right)$$

$$C_i = \frac{D_i(4\pi R\rho_v + z_i p_d)}{2K_{iv}}\left(\sqrt{1+\eta} - 1\right)$$

Where:

$$\eta = \frac{4K_0K_{iv}}{D_iD_v(4\pi R\rho_v + z_v p_d)^2}$$

Void Growth

- With everything defined,

$$C_v = \frac{D_v(4\pi R\rho_v + z_v p_d)}{2K_{iv}}\left(\sqrt{1+\eta} - 1\right)$$

$$C_i = \frac{D_i(4\pi R\rho_v + z_i p_d)}{2K_{iv}}\left(\sqrt{1+\eta} - 1\right)$$

$$\eta = \frac{4K_0 K_{iv}}{D_i D_v(4\pi R\rho_v + z_v p_d)^2}$$

$$dR/_{dt} = \dot{R} = \frac{\Omega}{R}\left(D_v(C_v - C_v^V) - D_i C_i\right)$$

- We can now rewrite the growth law as:

$$\dot{R}R = \frac{\Omega}{2K_{iv}} D_i D_v(z_i\rho_d - z_v\rho_d)\left(\sqrt{1+\eta} - 1\right)$$

Void Growth

$$R\dot{R} = K_o\Omega\left(\frac{z_i - z_v}{z_v}\right)\frac{z_v\rho_d}{(4\pi R\rho_v + z_v\rho_d)(4\pi R\rho_v + z_i\rho_d)}F(\eta)$$

- The first term is the main dpa-rate effect on void growth
- The second term is the "bias" term: if $Z_I = Z_V$, *void growth is impossible*
- The third term is the sink-strength balance term. Void growth is eliminated if there are too many or too few dislocations. Optimum growth occurs when the void sink term $(4\pi R\rho_v)$ and the dislocation sink term $(z_v\rho_d)$ are equal.
- The fourth term contains the effect of point defect recombination:

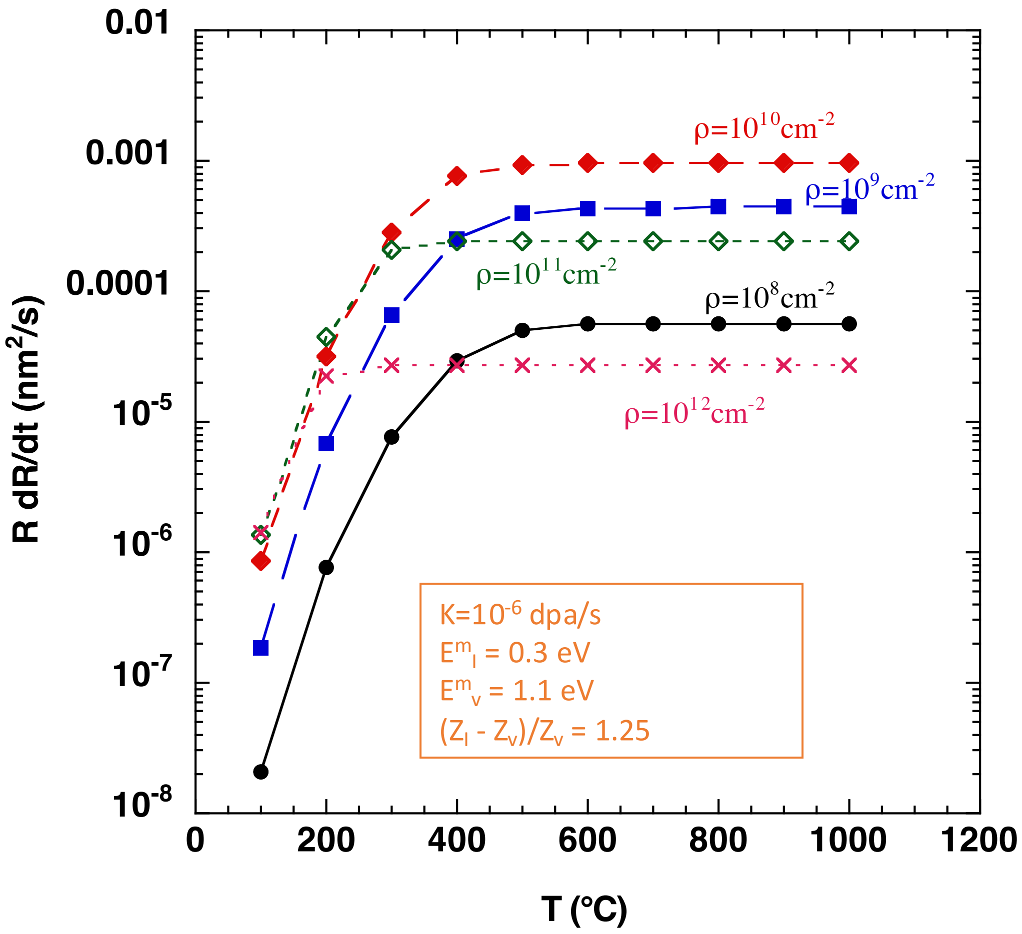
$$F(\eta) = 2\left(\sqrt{1+\eta} - 1\right)/\eta$$

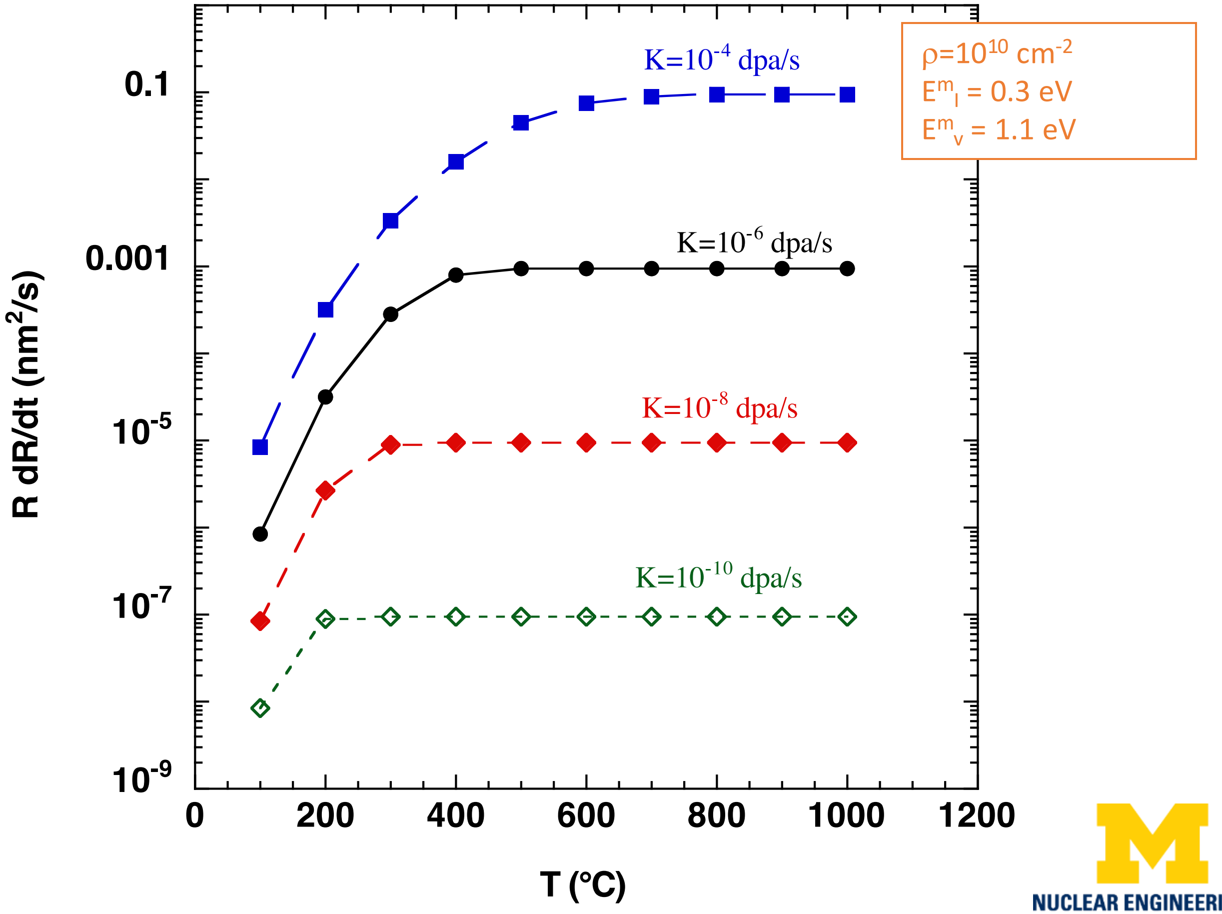
Since h decreases with increasing temperature and F decreases with increasing η:

- At high temperature, F $\rightarrow$ 1 and recombination does not effect void growth
- At low temperature, F $\rightarrow$ 0 and recombination prevents void growth.

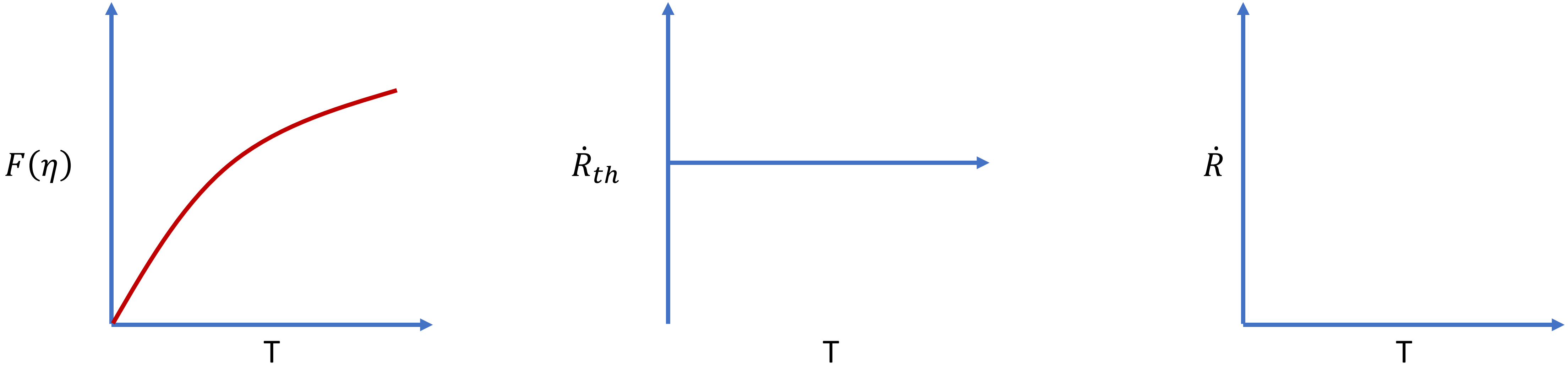
Void Growth

$$R\dot{R} = K_o\Omega\left(\frac{z_i - z_v}{z_v}\right)\frac{z_v\rho_d}{(4\pi R\rho_v + z_v\rho_d)(4\pi R\rho_v + z_i\rho_d)}F(\eta)$$





Void Swelling Temperature Dependence



At very high temperatures, void growth ceases because the vacancies "boil off" the voids. Repeating the previous derivation without neglecting C_v^0 gives the following shrinkage rate that competes with the growth rate:

$$R\dot{R}_{th} = -\frac{D_v C_v^0 \Omega^2 z_v \rho_d}{kT(4\pi RN + z_v \rho_d)}(e^y - 1)$$

Effect of Structure on Void Swelling

- Ferritic steels swell at rates ~0.2%/dpa
- Structure alone is not sufficient to explain the difference between α-Fe (BCC) and γ-Fe (FCC)
- BCC vanadium alloys can swell at rates more like austenitic steels
- Difference is likely in the relative bias for point defects at sinks
- If the bias is removed: $z_i = z_v$, void growth is impossible
- Recall the 3rd term, put simply:

$$\left.\frac{z_v \rho_d}{(4\pi R \rho_v + z_v \rho_d)(4\pi R \rho_v + z_i \rho_d)}\right\} \quad \frac{Q}{(1+Q)^2}$$

Where: $Q = \dfrac{\rho_d z_i}{z_v 4\pi R \rho_v}$

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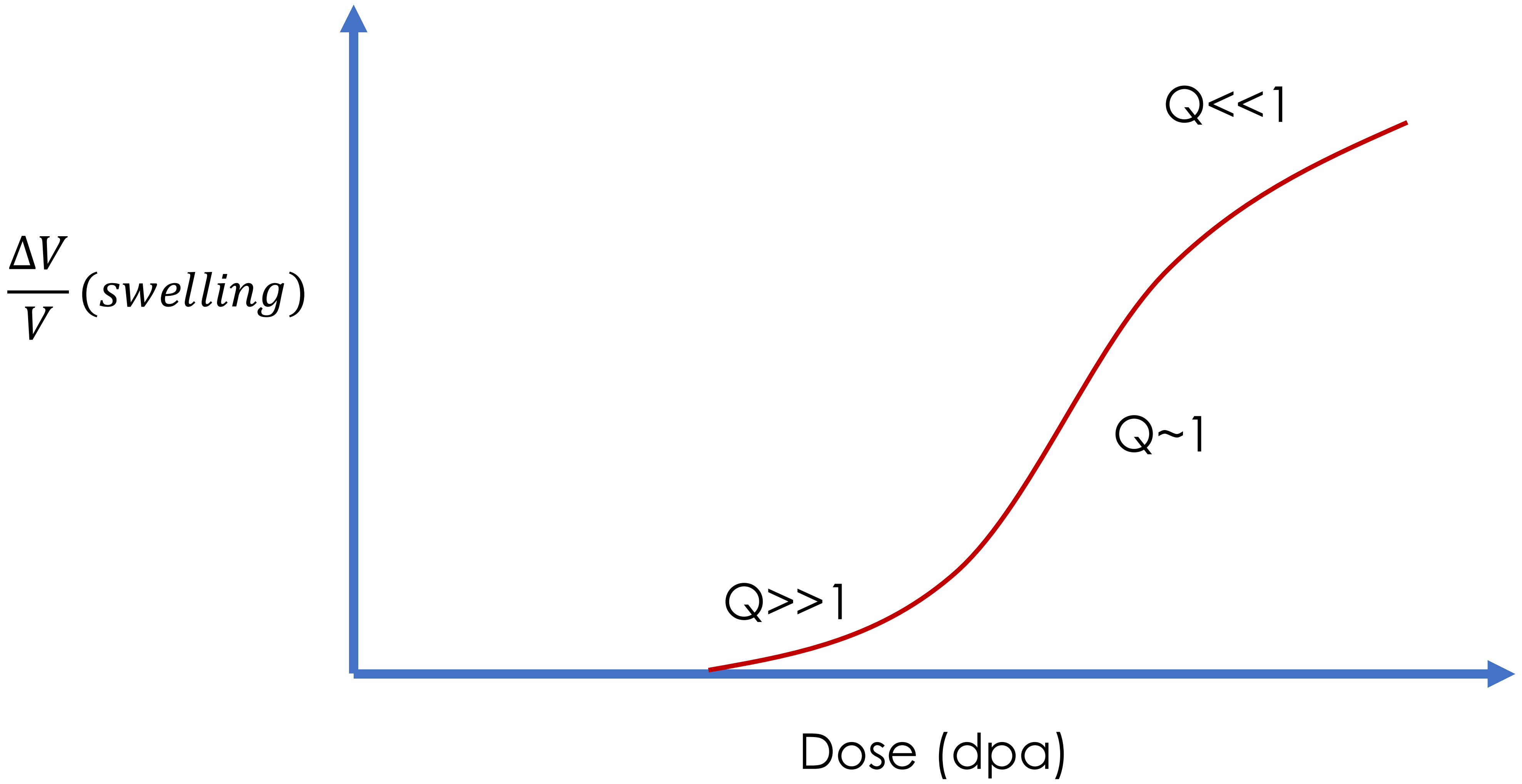
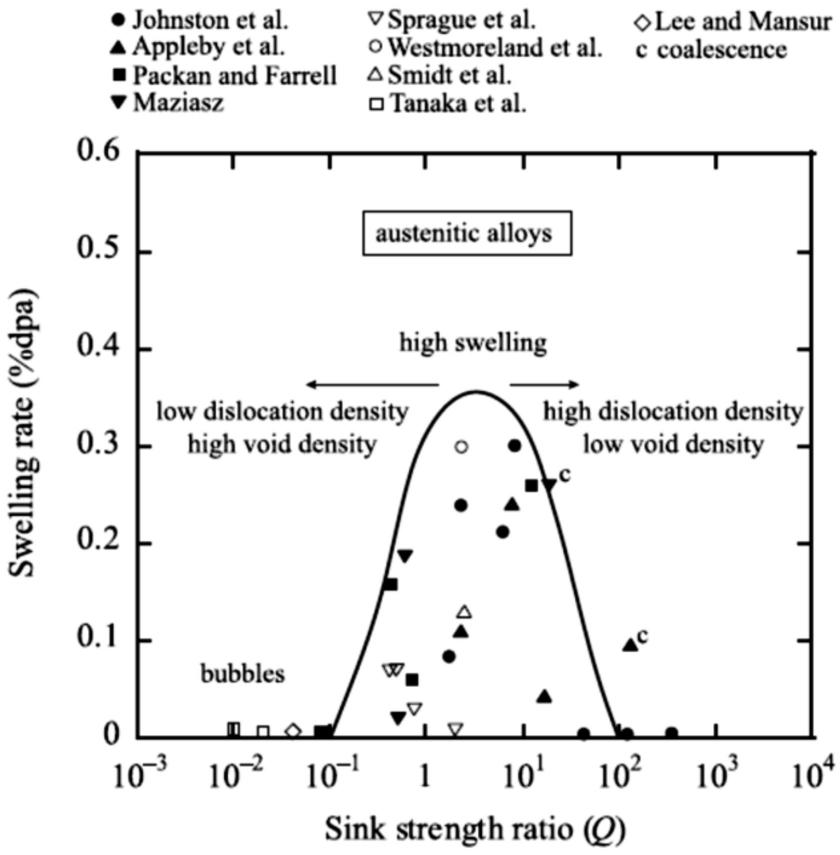
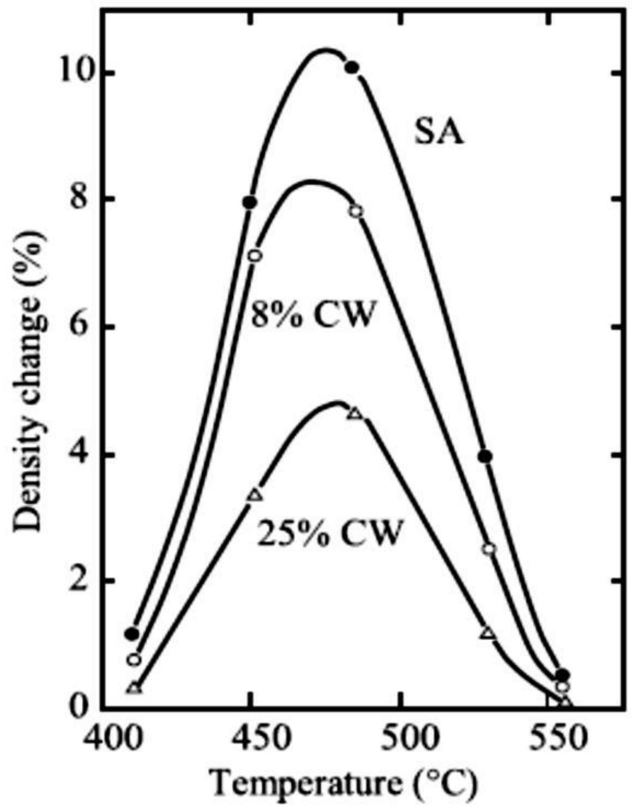
The Q-factor for structure dependence on dose



Effect of Structure on Void Swelling



Experimentally observed swelling rates as a function of Q for austenitic stainless steels (Mansur LK (1994) J NuclMater 216:97–123)



Dependence of swelling on cold-work for various temperatures for 316 stainless steel irradiated in the RAPSODIE reactor to doses of 20–71dpa (DupouyJM, Lehmann J, BoutardJL (1978) In: Proceedings of the Conference on Reactor Materials Science, vol. 5, Alushta, USSR. Moscow, USSR Government, pp 280–296)

- Growth rate is maximum when Q~1
- Growth decreases for Q != 1
- Observed experimentally
- CW reduces swelling because Q>>1

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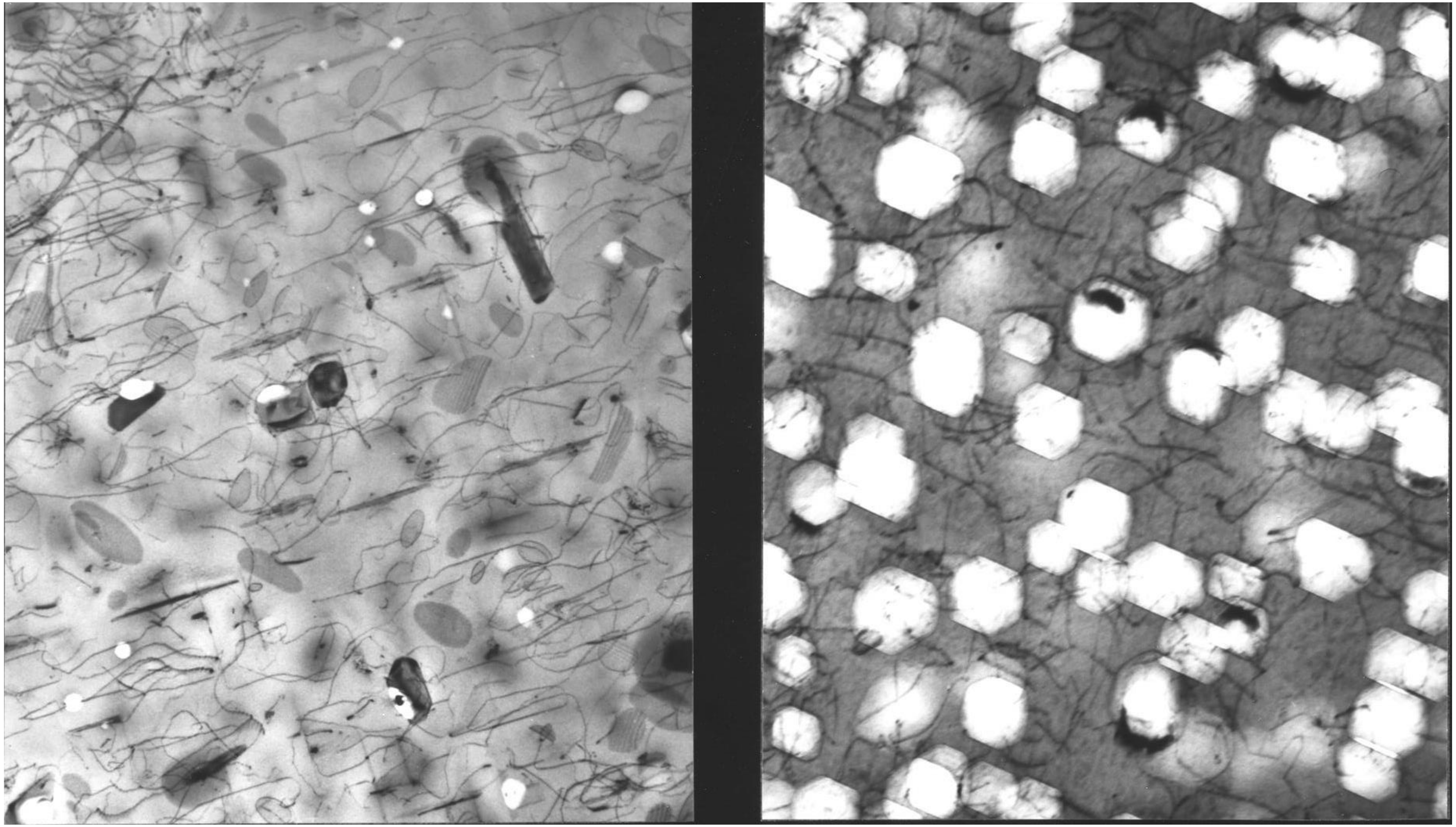
Based in Zeeland, Michigan, the iconic furniture company Herman Miller revolutionized office design forever when employee Robert Propst invented a modular, customizable system designed to give employees privacy and space. What year did Herman Miller introduce this invention, now famously known as the cubicle?

Image of voids + effect of sink strength

Commercial 316 SS (high sink strength) High-purity (low sink strength)



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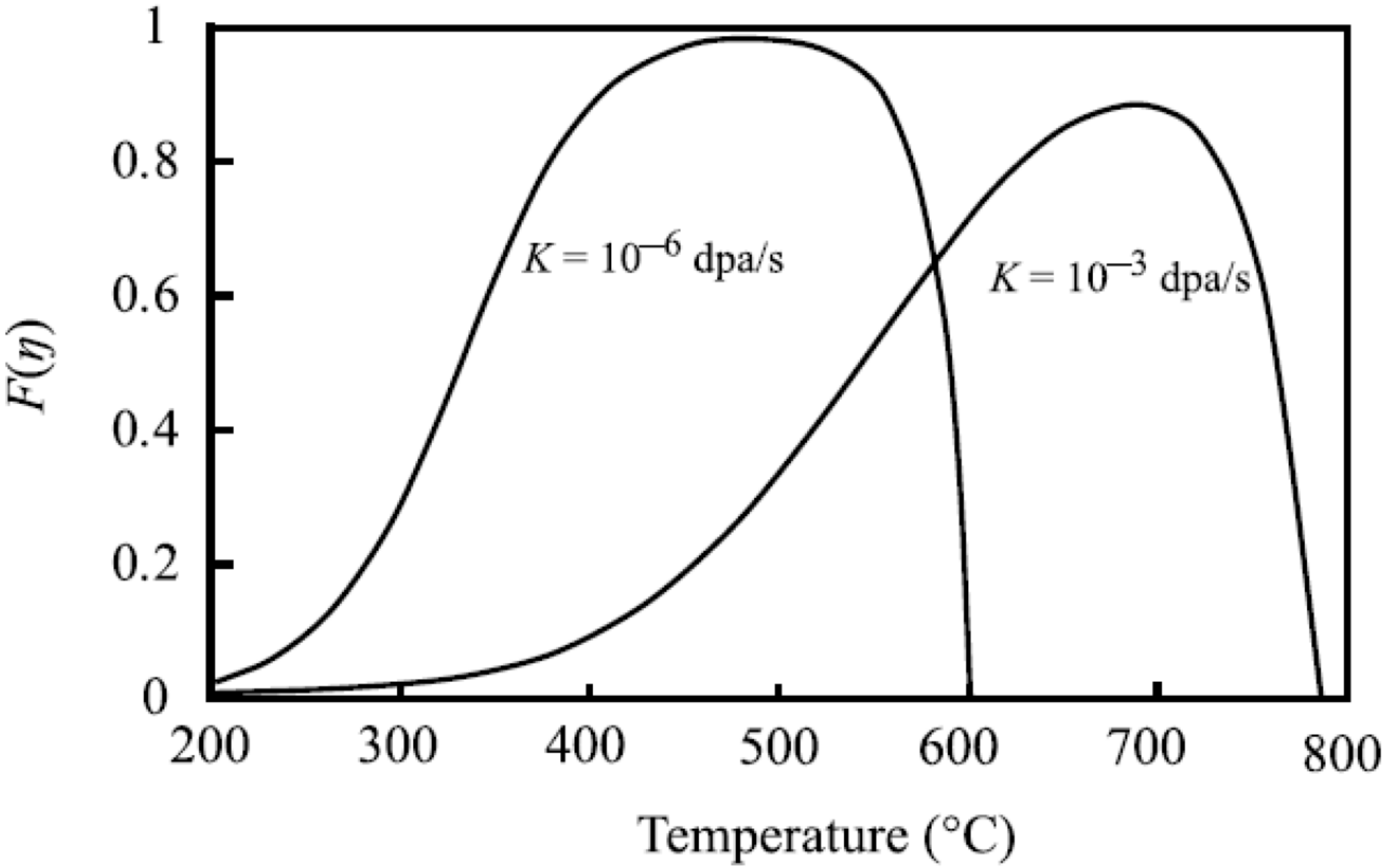
Effect of Dose Rate



Dose rate is captured in the **fourth term** where:

$$F(\eta) = 2\left(\sqrt{1+\eta} - 1\right)/\eta$$

And:

$$\eta = \frac{4K_0 K_{iv}}{D_i D_v (4\pi R \rho_v + z_v p_d)^2}$$

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Effect of Dose Rate

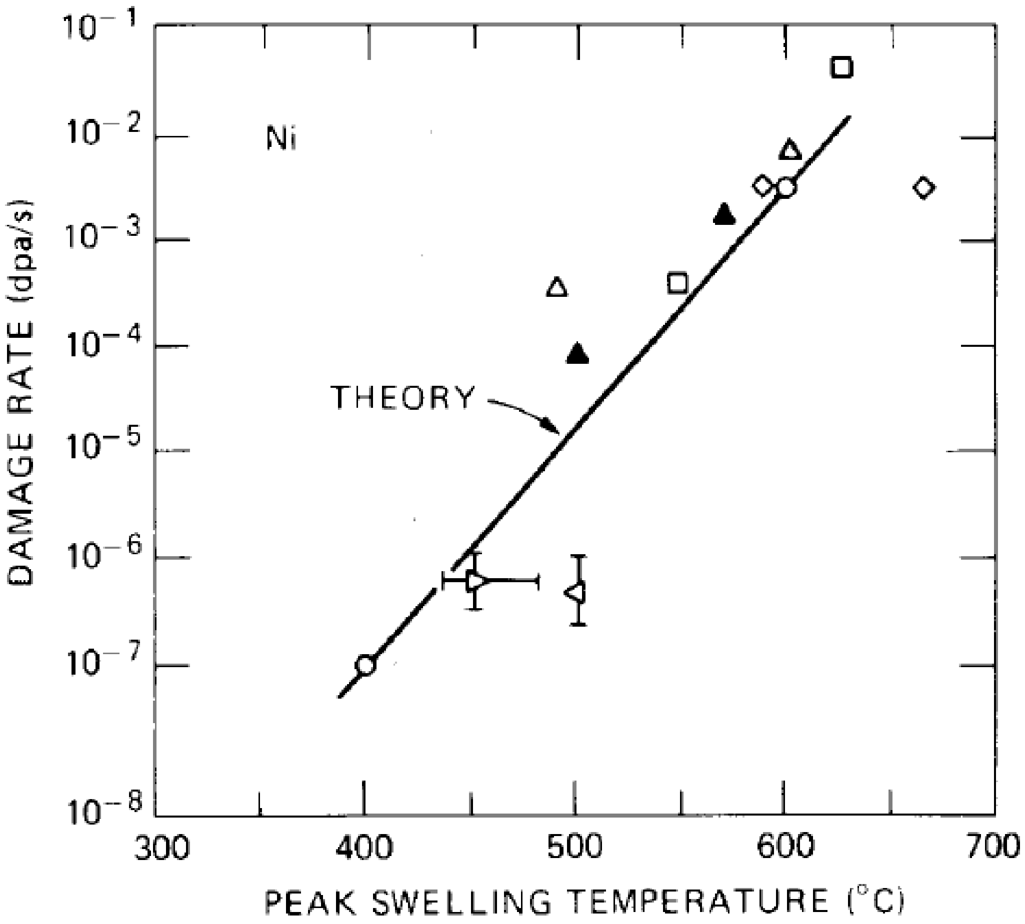

Figure 8.29. Compilation of experimental results for peak swelling temperature as a function of dose rate. Theoretically predicted trend is shown as the line. After Refs. 140 and 141.

Dose rate is captured in the **fourth term** where:

$$F(\eta) = 2\left(\sqrt{1+\eta} - 1\right)/\eta$$

And:

$$\eta = \frac{4K_0 K_{iv}}{D_i D_v (4\pi R \rho_v + z_v p_d)^2}$$

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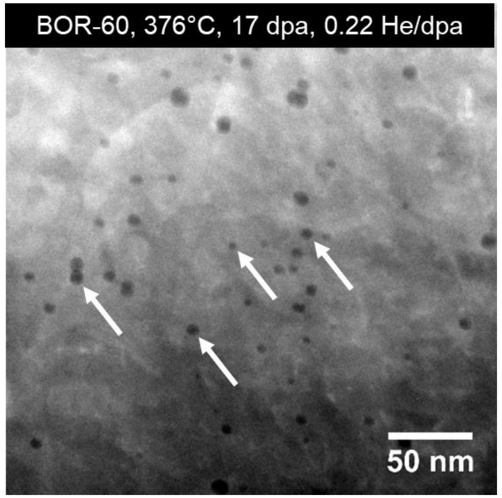
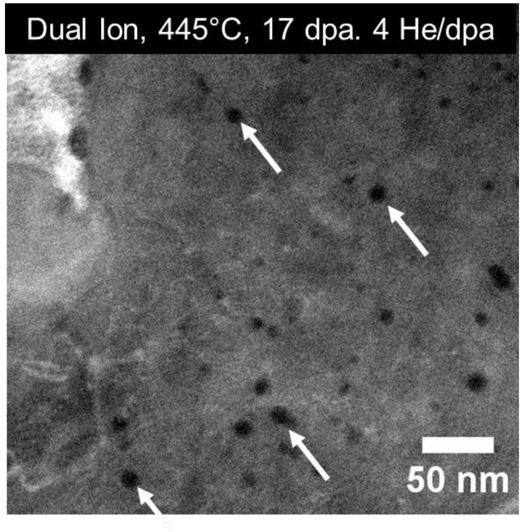
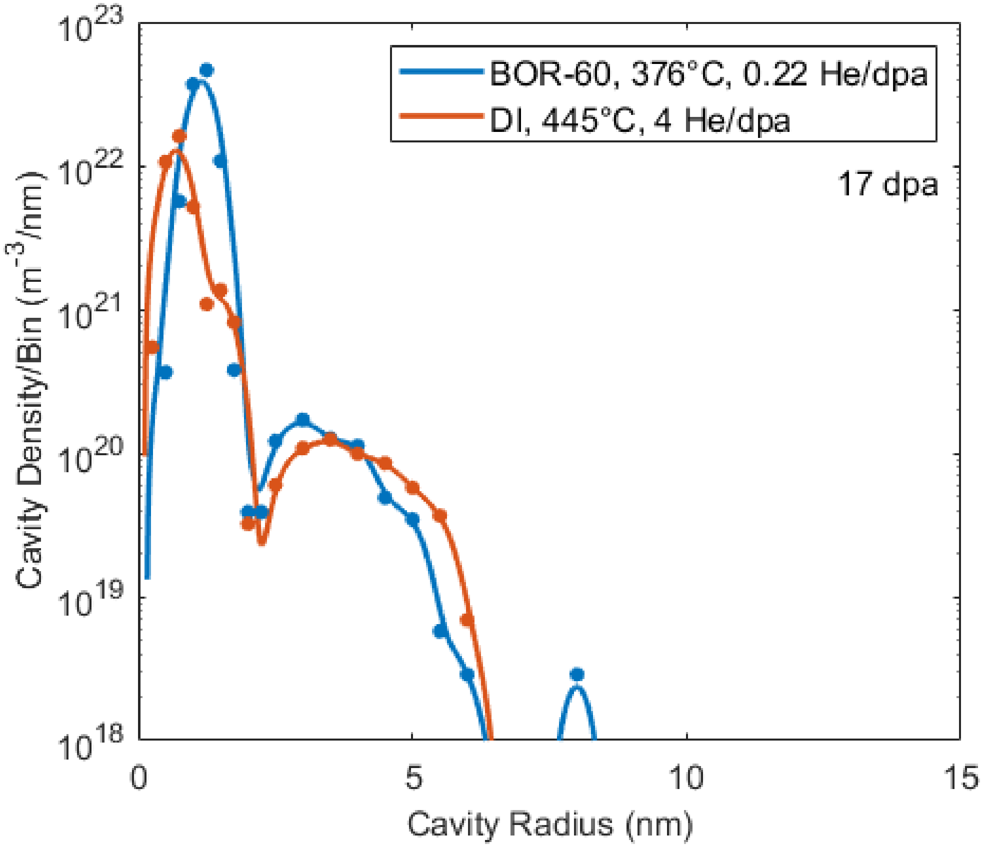
Effect of Dose Rate – Real World Example

STEM HAADF





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



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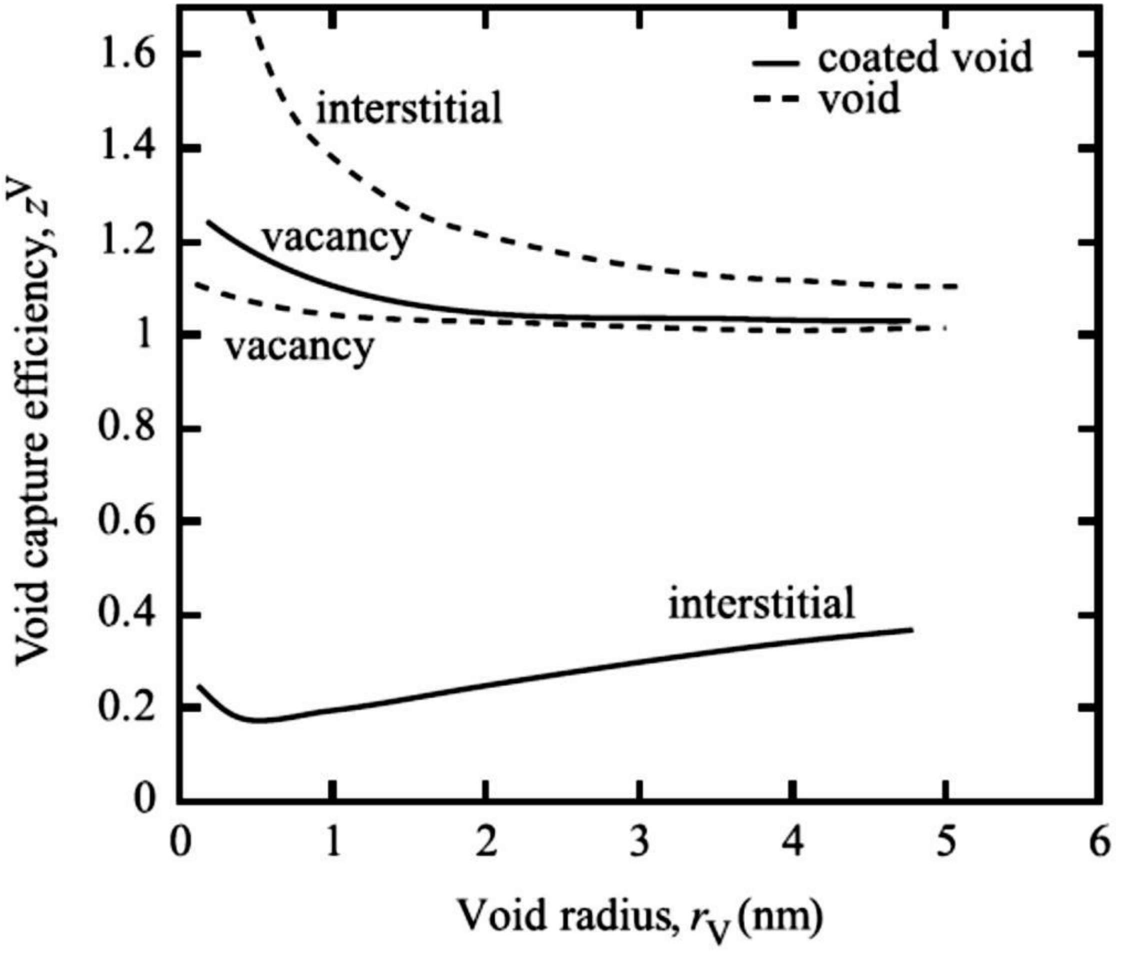
Effect of void surface segregation on defect bias



Capture efficiency for point defects diffusion to a void and a coated void as a function of void radius R_V. (W.G. Wolfer, L.K. Mansur, The capture efficiency of coated voids, Journal of Nuclear Materials, Volume 91, Issue 2, 1980, Pages 265-276)

- For a bare unpressurized void, interstitial bias is greater than vacancy bias. Voids will shrink
- If "shell" shear modulus or lattice parameter is greater than matrix shear modulus, vacancy bias becomes greater than interstitial bias
  - This effect can occur because of radiation induced segregation
- Thicker shells have a greater effect

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Effect of Inert Gas: Bubbles & Voids

- Inert gas atoms (H, He, etc.) are created by transmutation and interact with vacancies
  - Must be accounted for on bubble/void growth as:
    - Insoluble gas atoms can act as immobile nucleation sites to which vacancies and interstitials migrate to form voids
    - Inert gas atoms can stabilize a cavity and assist the nuclei during nucleation and growth
- First, let's assume the following:
  - No account taken of cascades or lattice imperfections
  - Gas atom association is stable and mobile

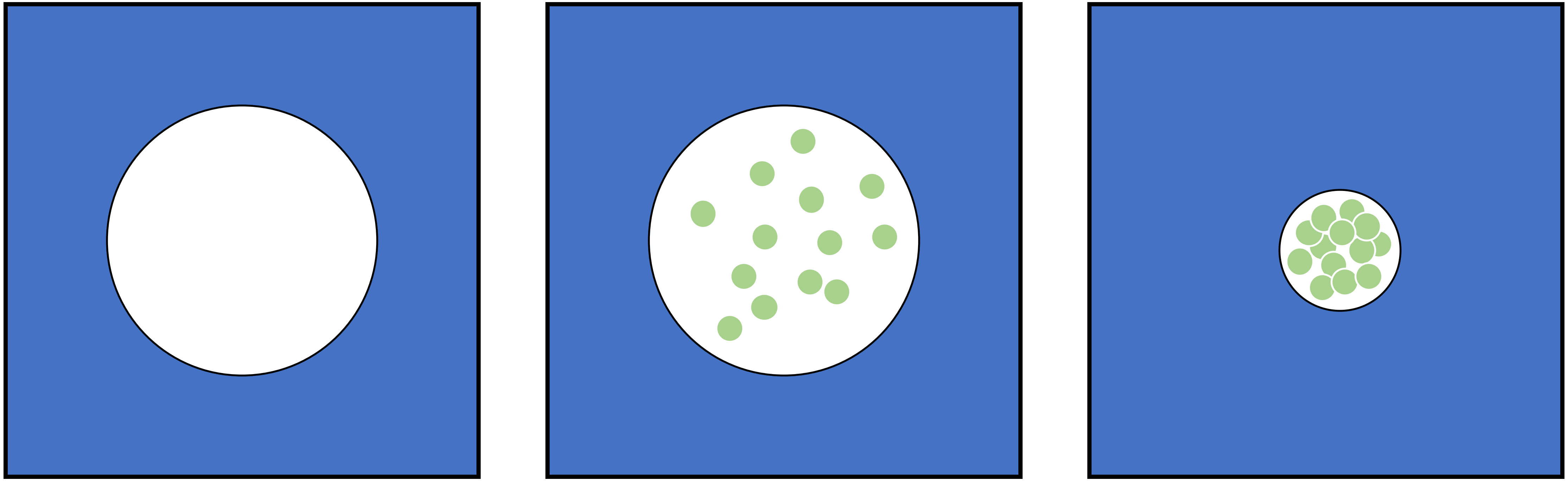
Side Note!
We generally define the following:

Void: open volume in a solid not pressurized by inert gas

Bubble: open volume in a solid that is pressurized by inert gas

Cavity: Generalization for open volume in a solid – can be a bubble or void

Effect of Inert Gas: Bubbles & Voids



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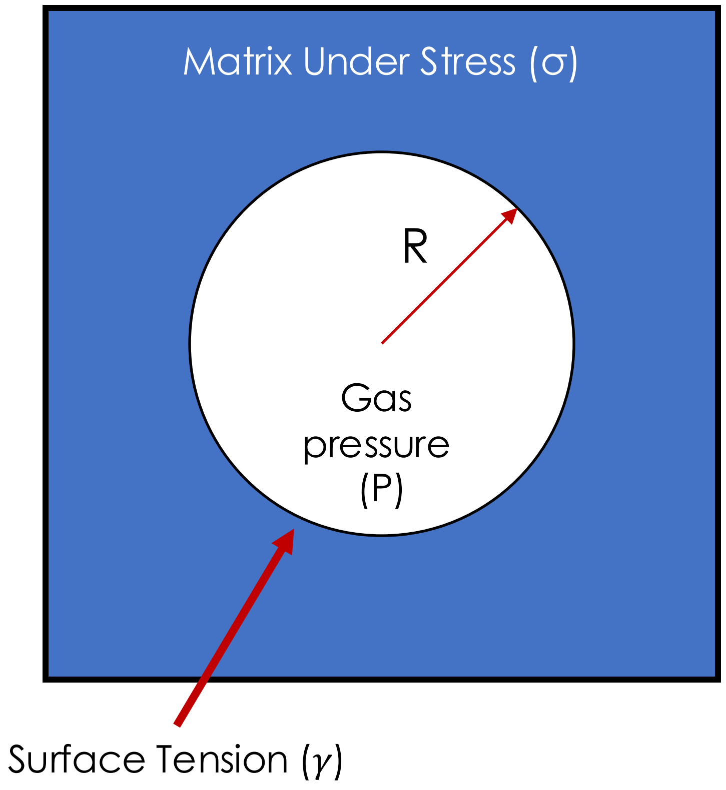
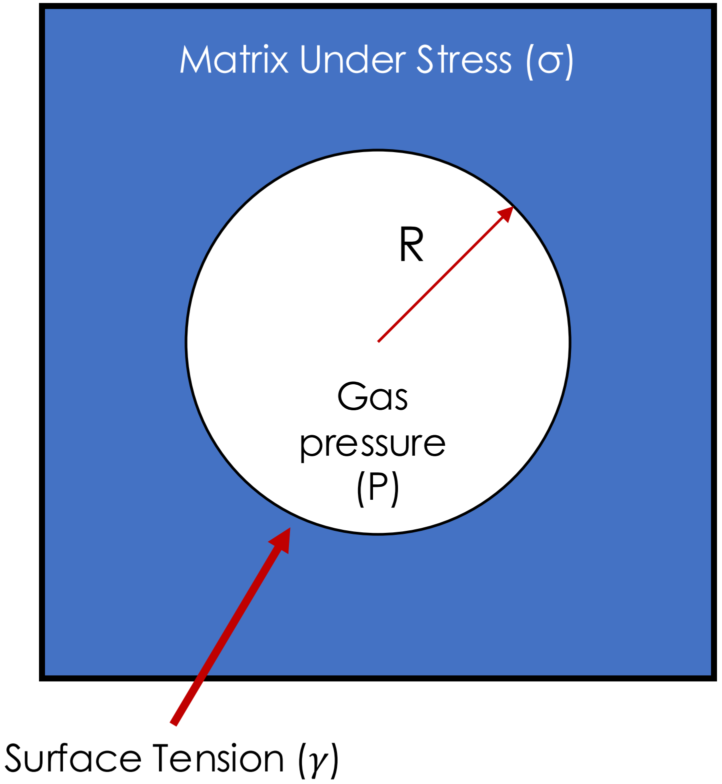
Effect of Inert Gas: Bubbles & Voids



- For a spherical cavity, the change in volume and surface area is:
- Under expansion (cavity growth) the pressure does work on $P\,dV$ and the surface energy increase by $\gamma\,dA$, or simply,
- If not at mechanical equilibrium, then:

Effect of Inert Gas: Bubbles & Voids



- Let's now calculate the number of gas atoms present in the bubble, using the ideal gas law:

- Remembering that $P = \frac{2\gamma}{r}$ and plugging in we get:

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Effect of Inert Gas: Bubbles & Voids

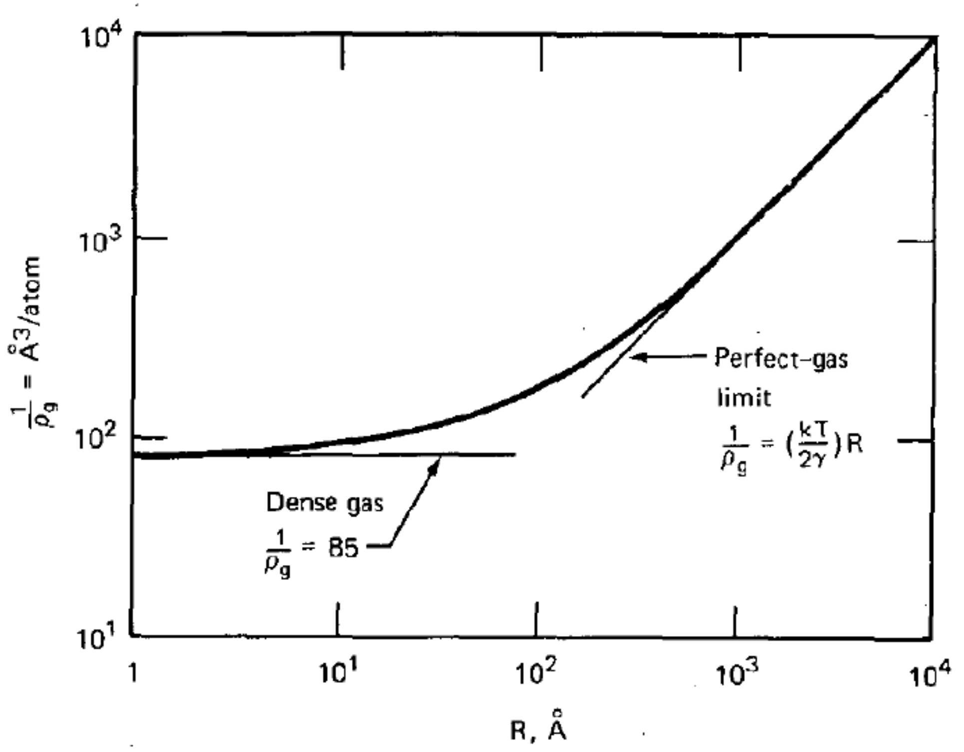


Fig. 13.3 Density of xenon gas in a spherical bubble imbedded in a stress-free solid of surface tension of 1000 dynes/cm.

For most applications we assume an ideal gas in mechanical equilibrium

- To account for non-ideal gas (e.g. high pressure in small bubbles) we need a different eq'n of state:
- We can then solve for n_x again using this relationship to get the number of gas atoms in the **dense gas limit:**

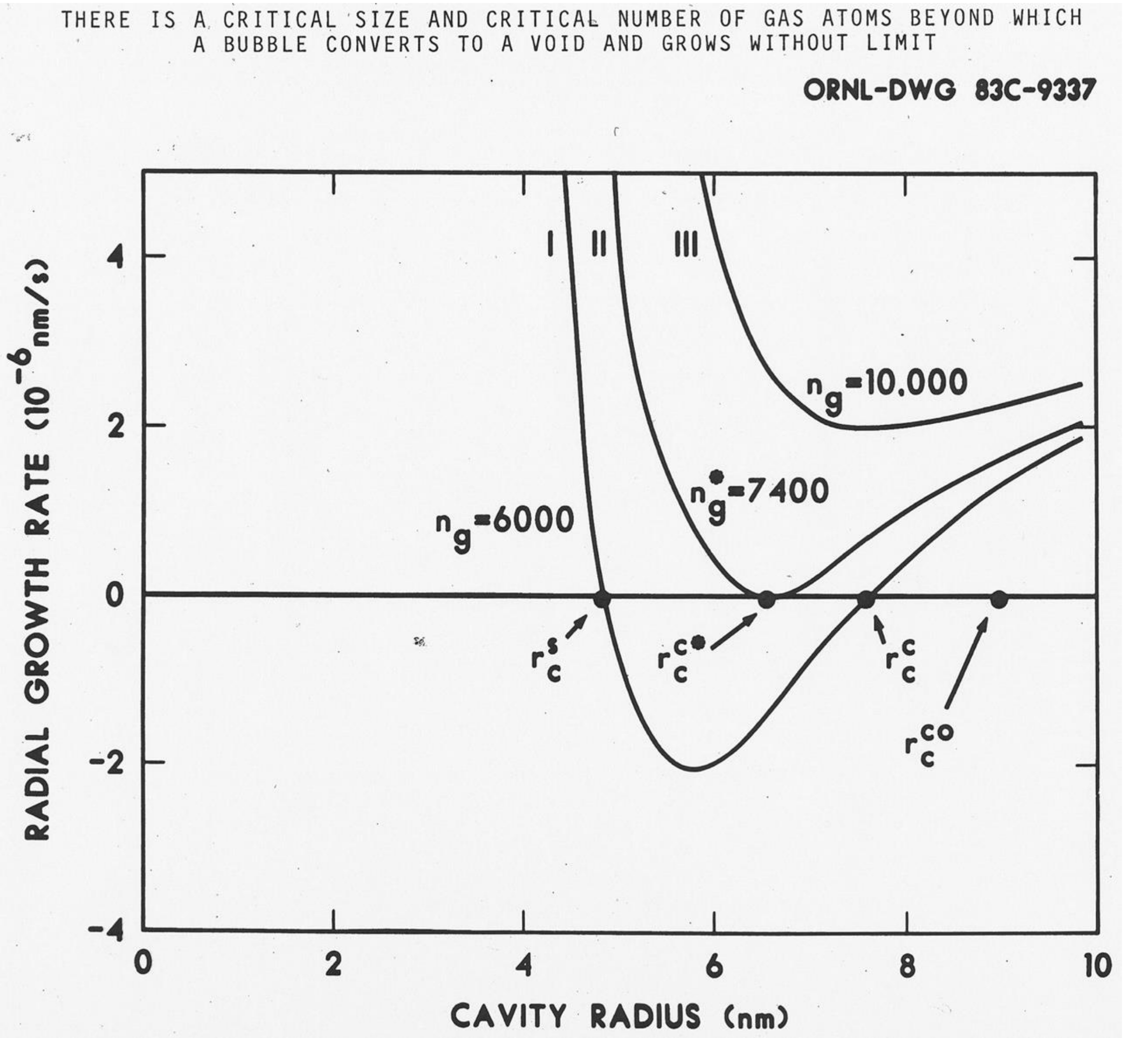
$$n_x = \frac{{}^{4}/_{3}\,\pi r^3}{B + \left({}^{k_b T}/_{2\gamma}\right) r}$$

And for non-equilibrium bubbles:

$$n_x = \frac{128\pi\gamma^3}{81\sigma_c^2 k_b T}$$

Now that we have an expression for n_x and P, we can add these into the terms for the growth rate law including thermal emission, we then get:

$$R\dot{R} = K_o\Omega\left(\frac{z_i - z_v}{z_v}\right)\frac{z_v\rho_d}{(4\pi R\rho_v + z_v\rho_d)^2}F(\eta) - \frac{D_v C_v^0 \Omega^2 z_v \rho_d}{kT(4\pi RN + z_v\rho_d)}\left(\frac{2\gamma}{R} - \frac{n_x kT}{{}^4/_3\,\pi R^3 - n_x B}\right)$$



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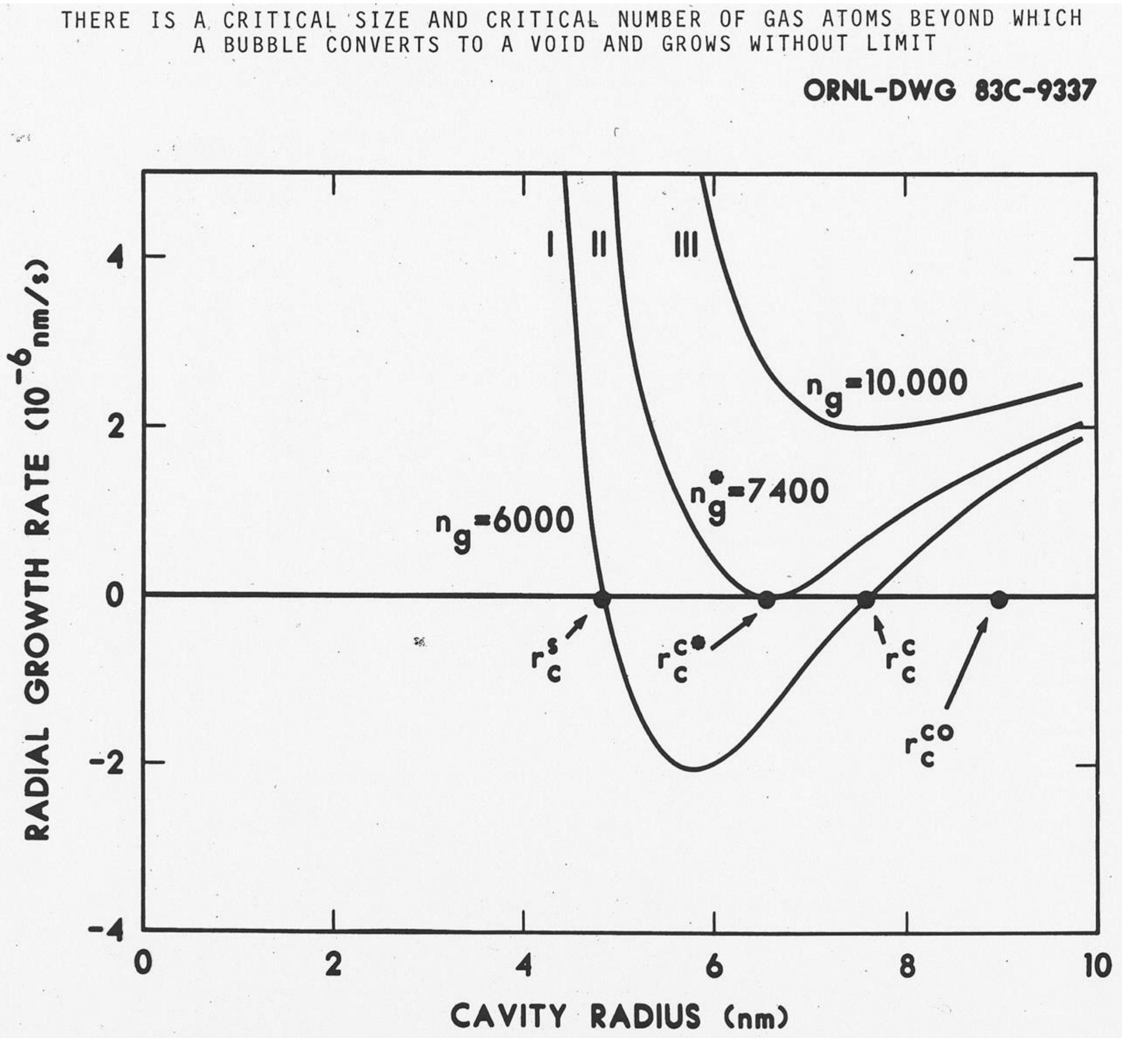
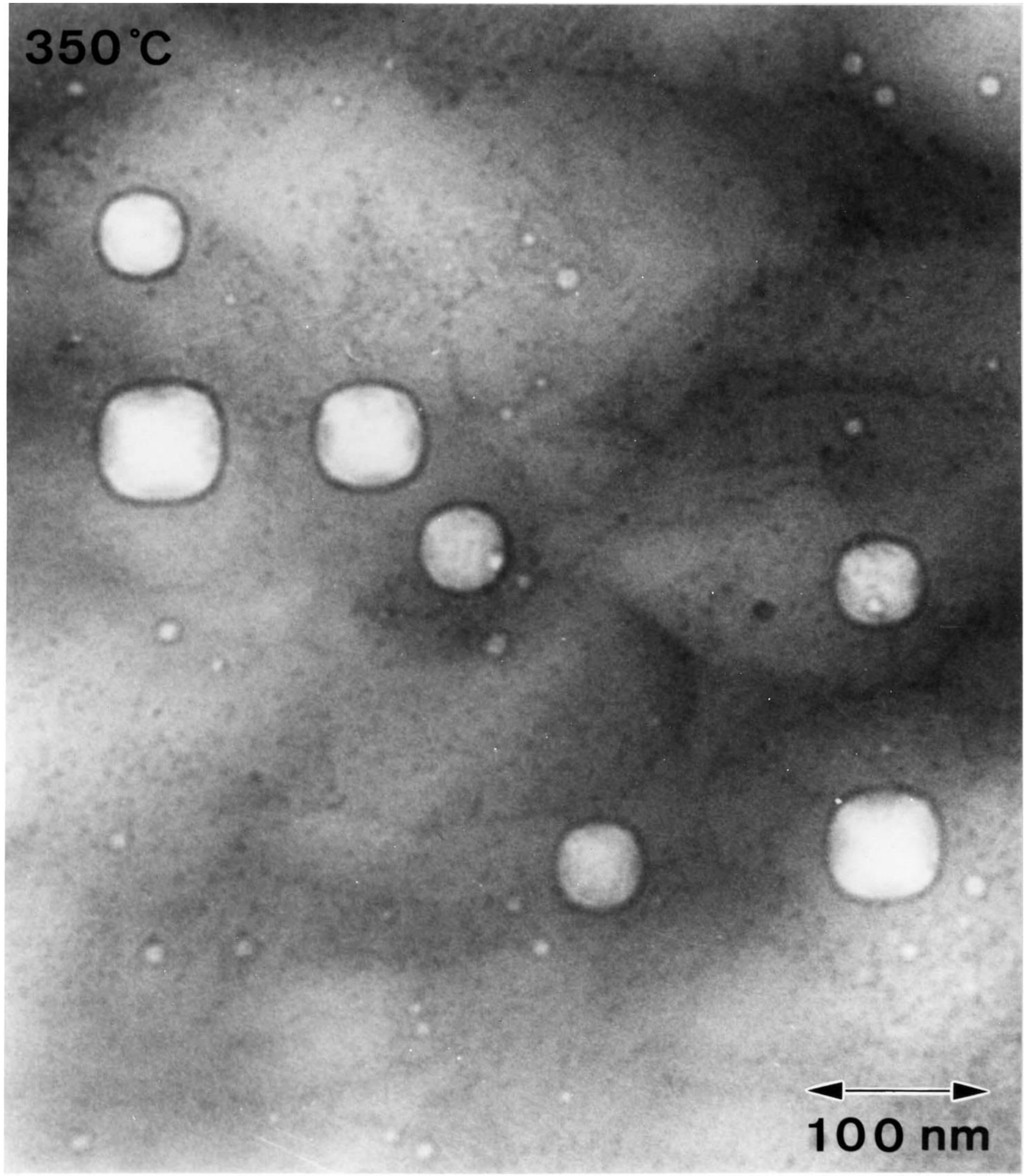
When gas is present, the current models predicts that cavities containing less than n_g* gas atoms remain at or below r_c*, but those with more than n_g*, this creates a bimodal distribution





Zinkle, Farrell and Kanazawa, J. Nucl. Mater. 179-191 (1991) 994

Void and He bubble formation in Cu-100 ppm B following fission neutron irradiation to 1.2 dpa at 350°C

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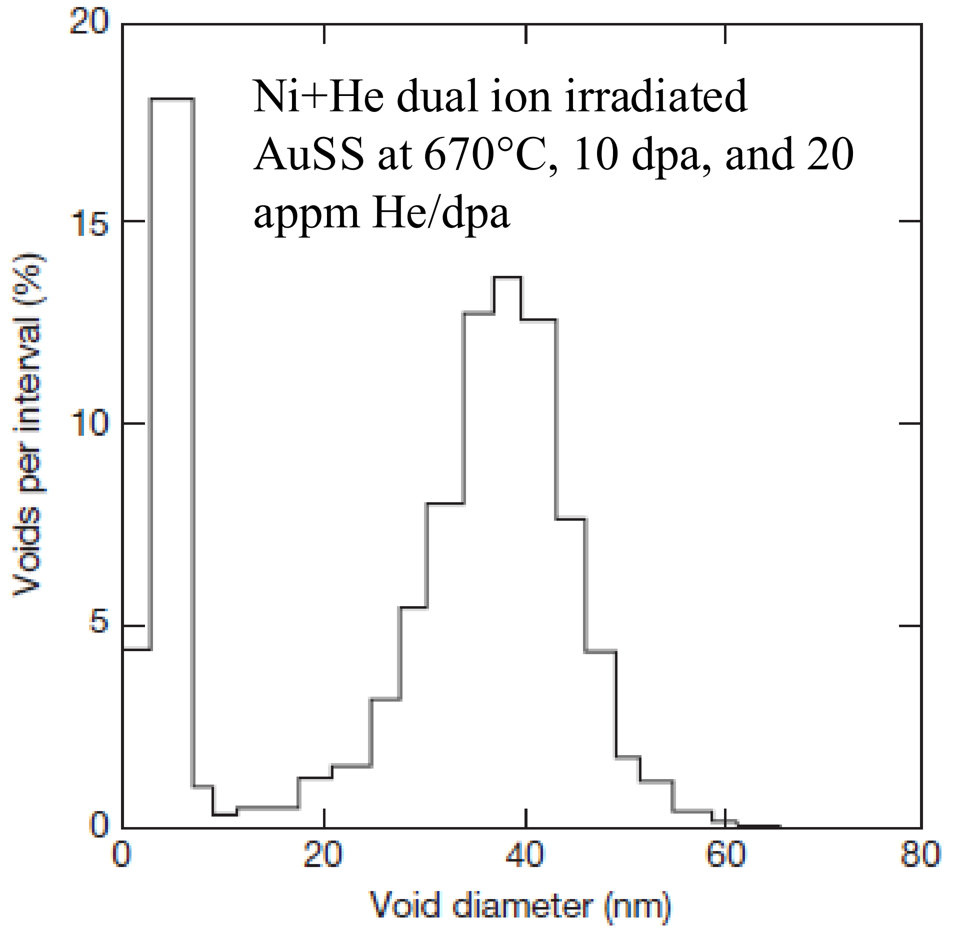
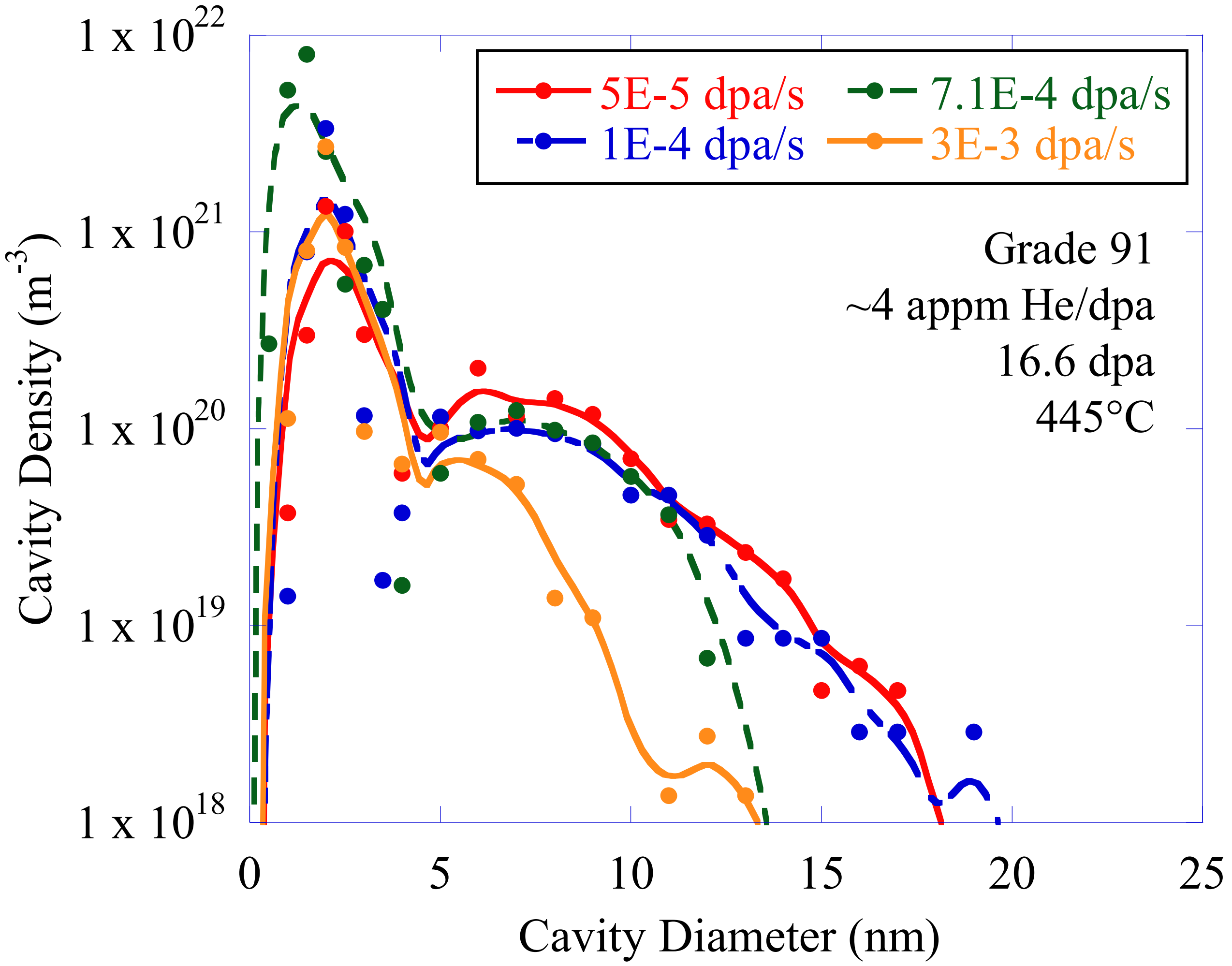
Experimental examples

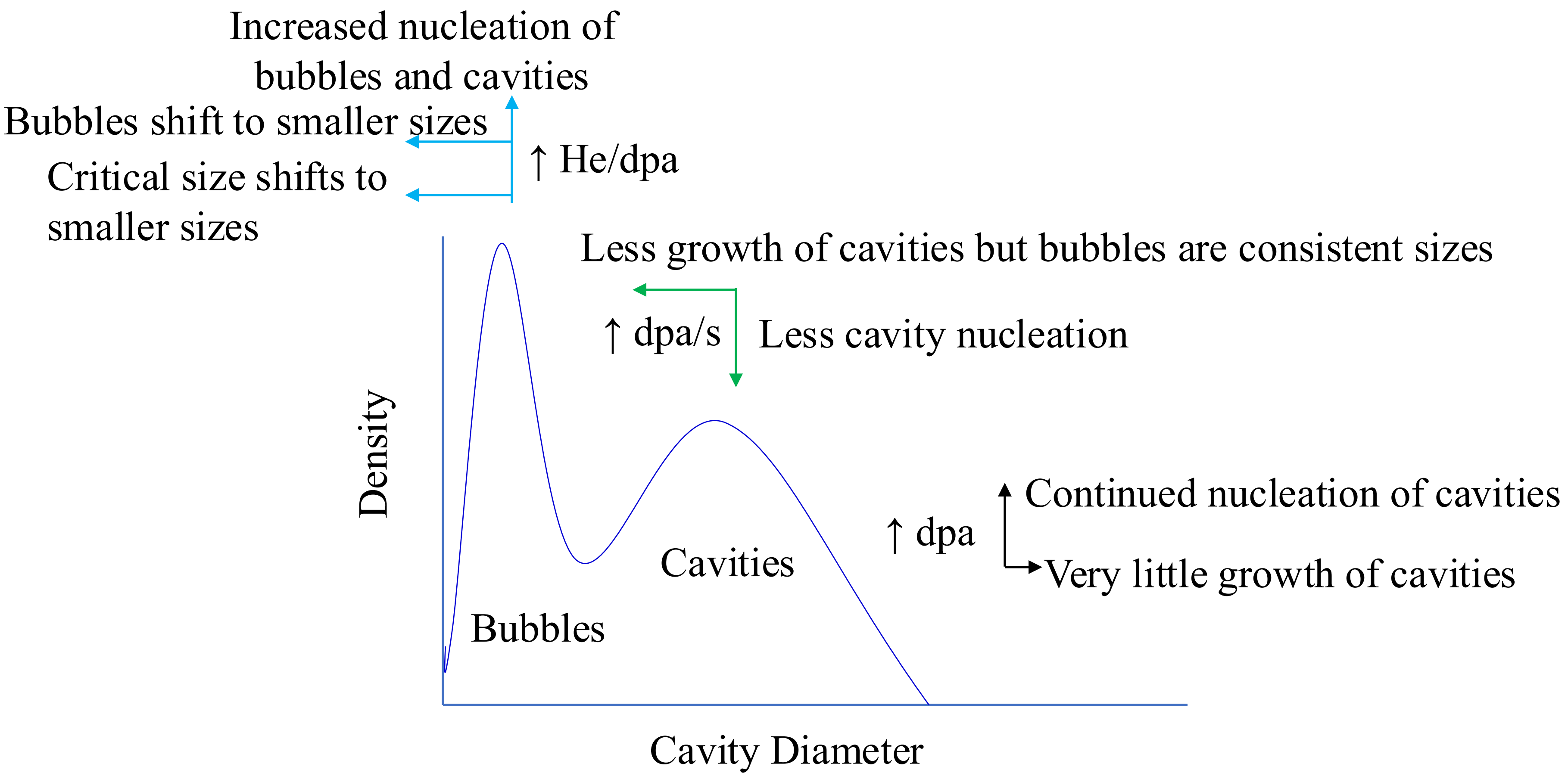


Mansur, Coghlan JNM 119 (1983)



Taller (2019)

Experimental examples



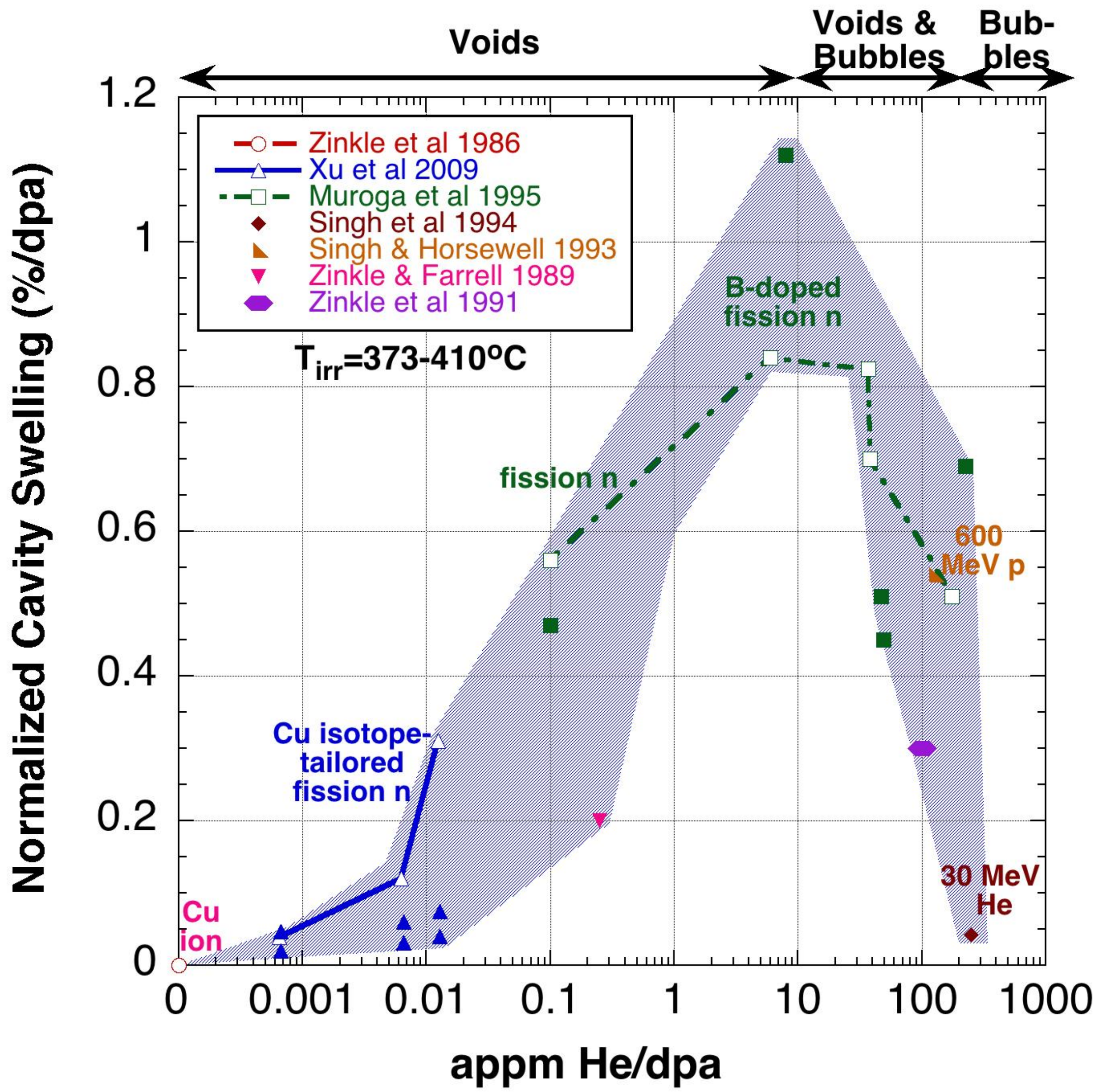
Fixed dpa/s – Variable He/dpa

Fixed He/dpa – Variable dpa/s

Increase in dpa for constant He/dpa and dpa/s

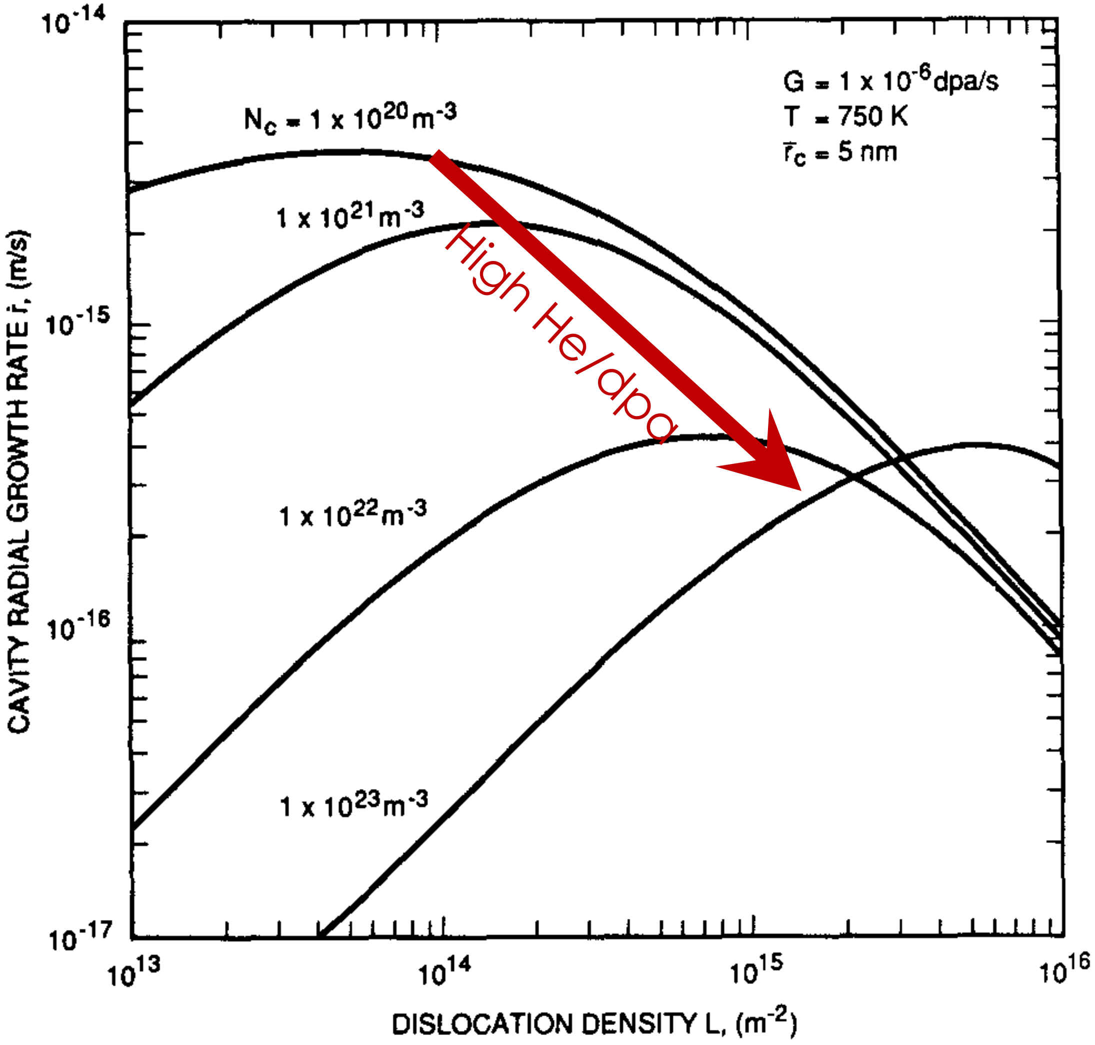
Taller (2019)

Cavity swelling vs. He/dpa ratio in irradiated copper



~1-10 dpa

Calculated void growth rate is typically reduced for high cavity and dislocation sink strengths



Over nucleation of cavities due to too high He/dpa can suppress void swelling

E.H. Lee & L.K. Mansur, Met. Trans. A 21 (1990) 1021

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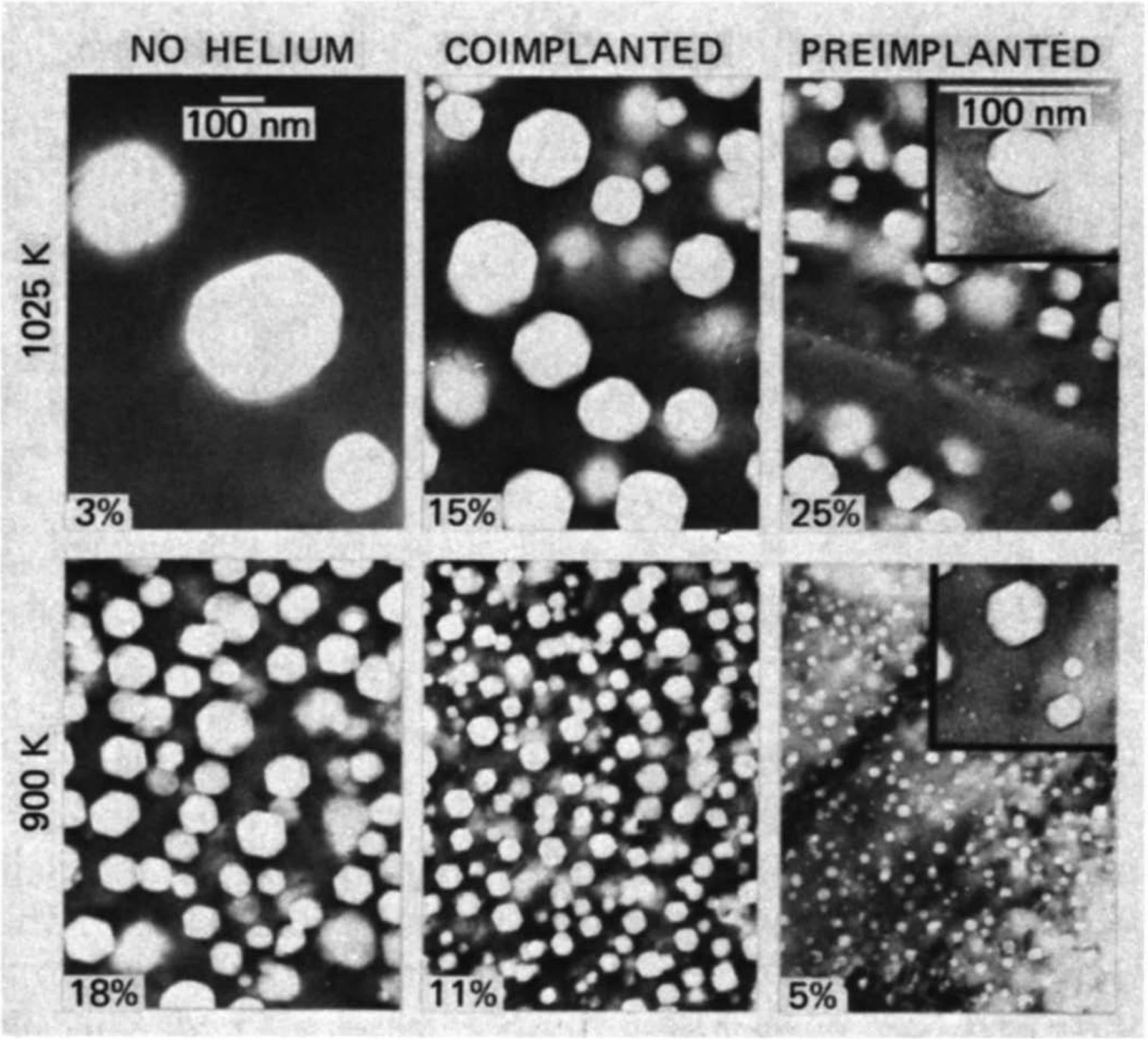
Effect of He in ion irradiations



Implantation method of He can drastically effect swelling in ion irradiated materials

Image of Fe-17Cr-16.7Ni-2.5Mo

Packan & Farrell, NT-Fusion, 1983

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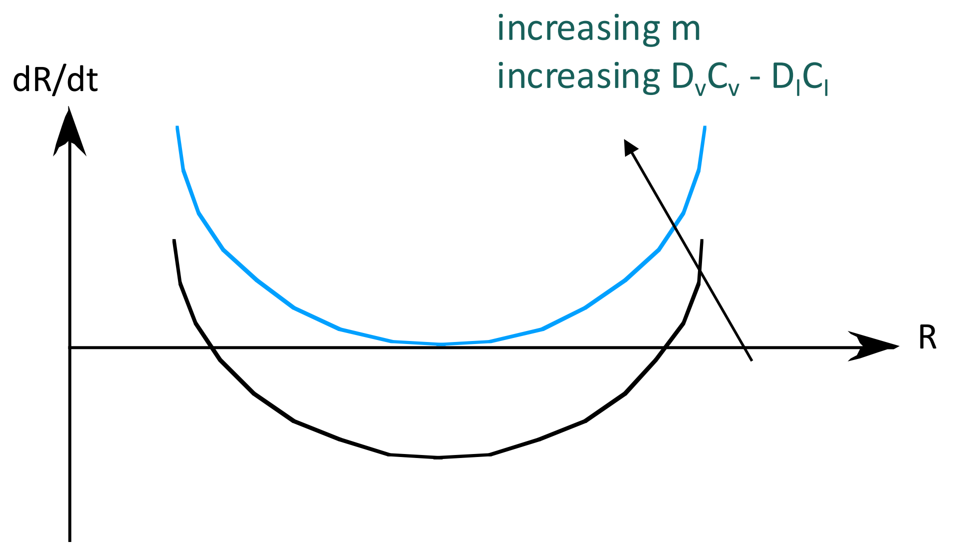
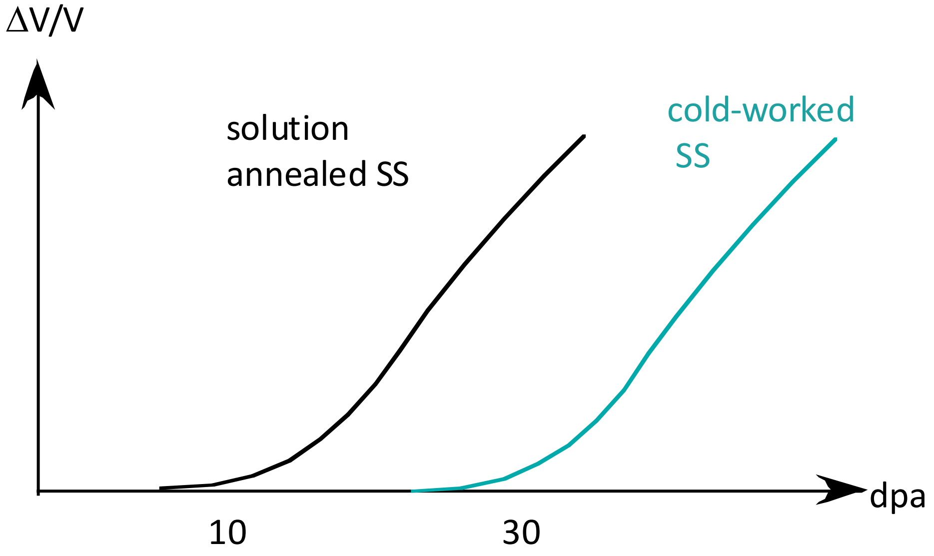
Remedies for void swelling?

Remedies for void swelling?

- Decrease $D_vC_v - D_IC_I$ arriving at cavity;
- Eliminate He gas production (expensive or impractical)

- Reduce C_v, C_I:
  - increase recombination
    - add precipitates or dispersoids (TiC/TiO_2) to act as recombination sink, trap He and stabilize dislocations
  - increase other sink strengths
    - add dislocations (cold-work); generally only effective for low to moderate doses
    - introduce nanoscale grain boundaries





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Questions?