

# Voids II

K.G. Field<sup>1,a</sup>,

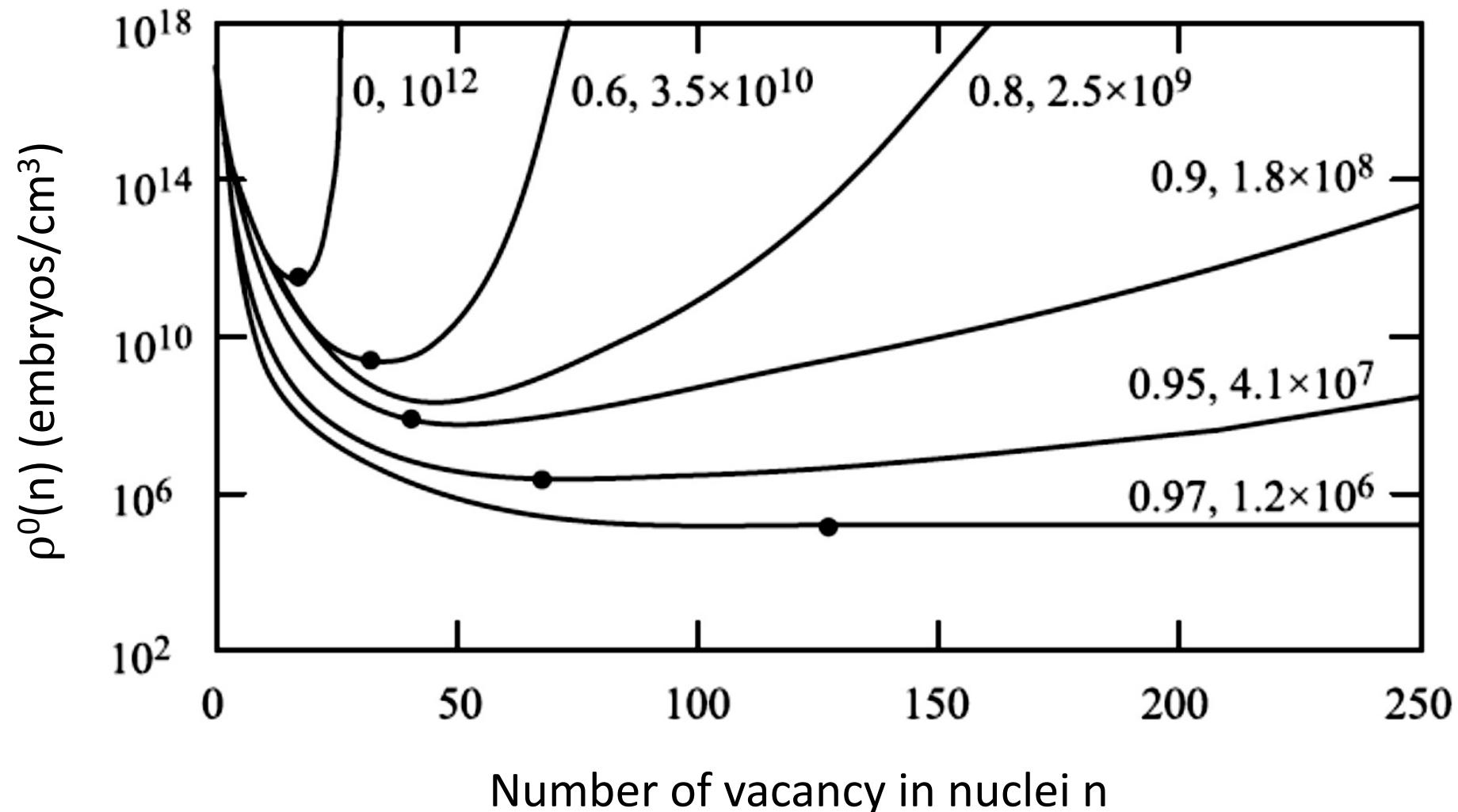
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# Concentration of void embryo sizes w/ nucleation rate



# Void Nucleation Rate

- To obtain void nucleation as  $(D_i C_i)/(D_v C_v)$  approaches 1 requires higher vacancy supersaturation
- Strong dependence of nucleation on vacancy supersaturation

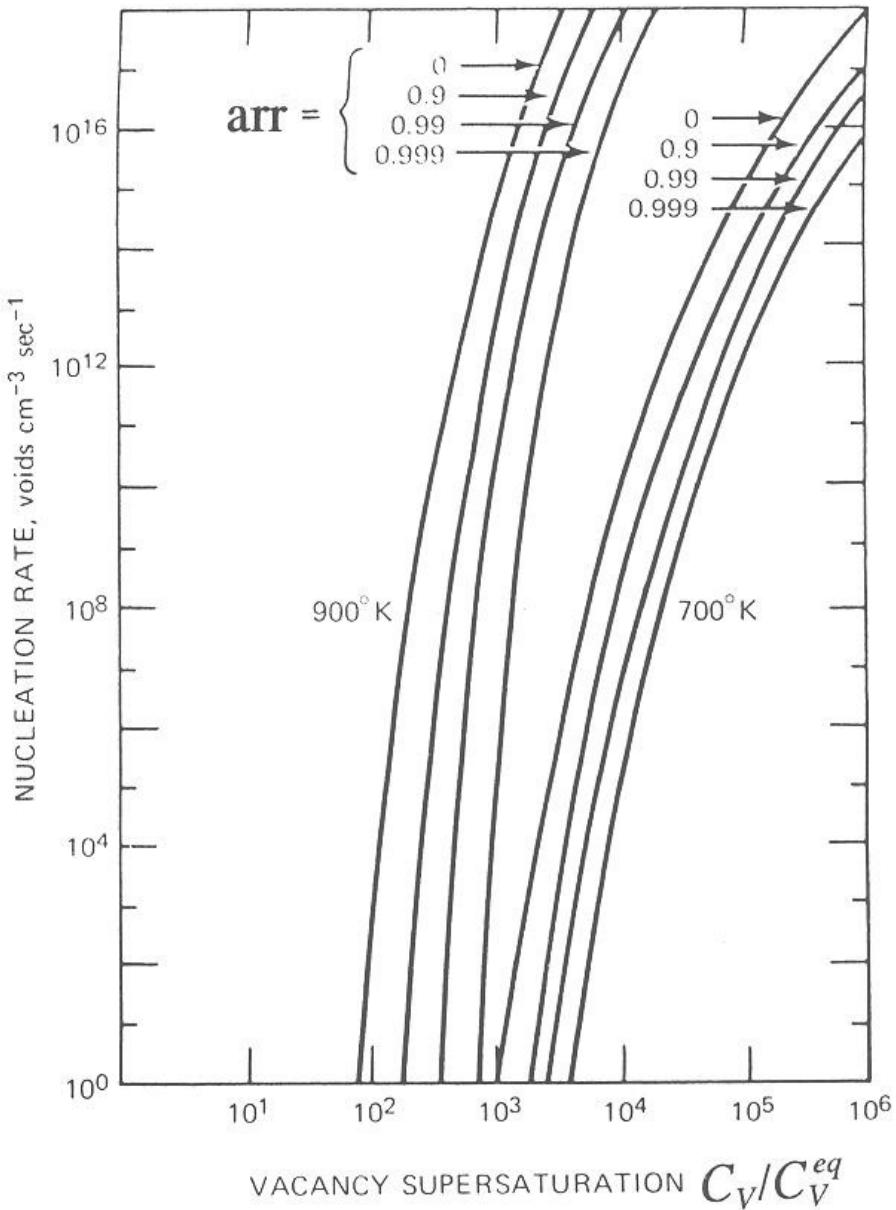
Typical results:

$$T = 700 \text{ K}; C_v/C_v^{eq} = 10^4$$

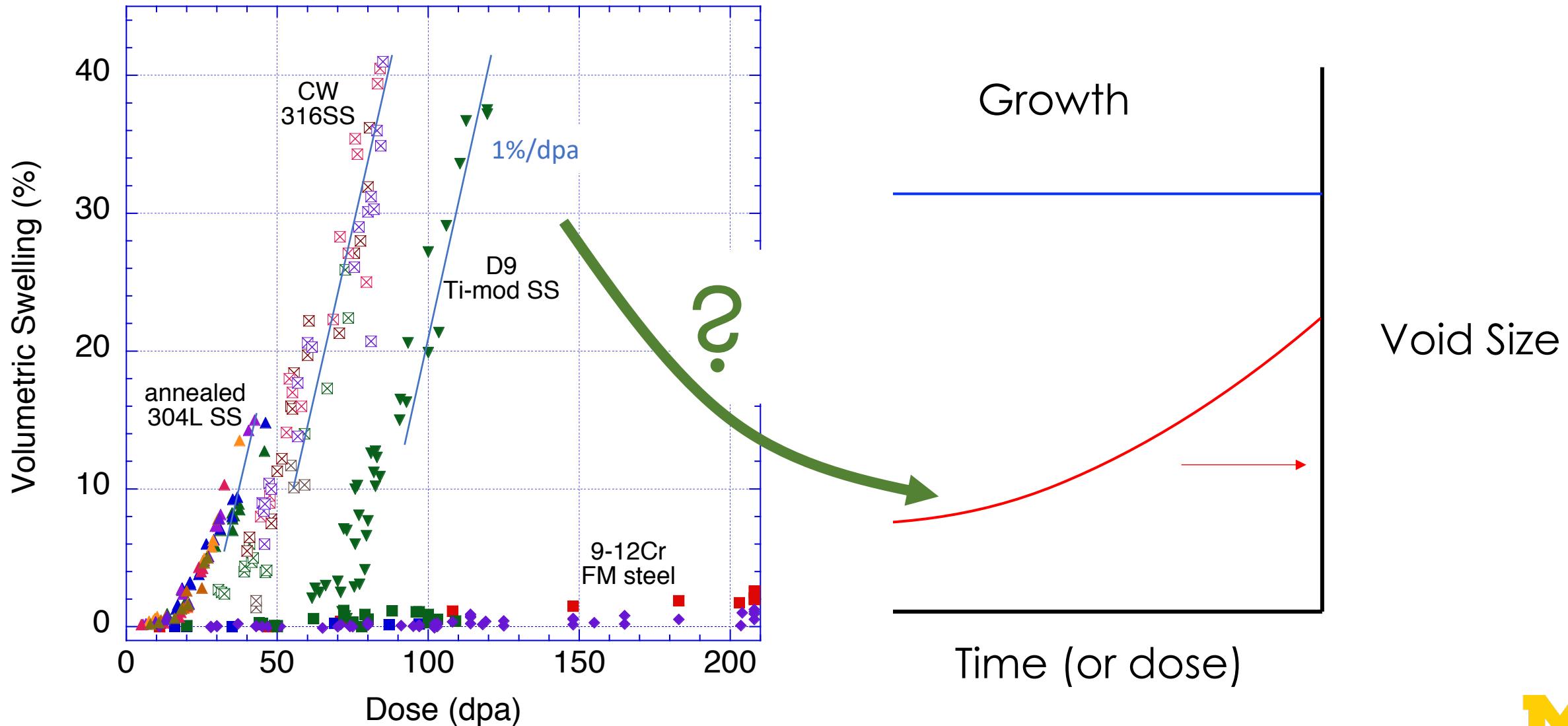
$$\text{arr} = (D_i C_i)/(D_v C_v) = 0.99$$

$$J \sim 10^8 \text{ voids nucleated}/\text{cm}^3/\text{s}$$

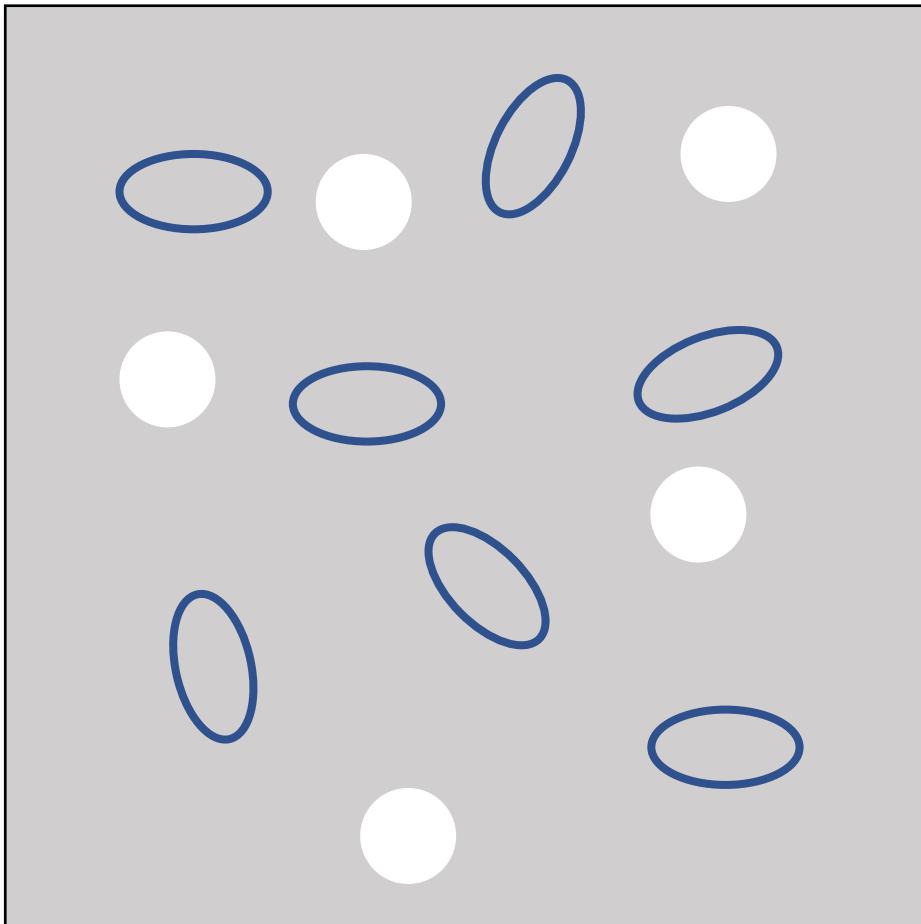
- After 1 year,  $3 \times 10^{15}$  voids/cm<sup>3</sup>
- The voids are small, about the critical size



# Nucleation vs. Growth



# 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:

$$J_{net}^V = J_v^V - J_i^V = 4\pi R \Omega D_v (C_v - C_v^V) - 4\pi R \Omega D_i (C_i - C_i^V)$$

$$J_{net}^V = dV/dt = 4\pi R \Omega (D_v C_v - D_i C_i)$$

$$dR/dt = \dot{R} = \frac{\Omega}{R} (D_v C_v - D_i C_i)$$

$$C_v^V = C_v^0 \exp\left(\frac{2\gamma\Omega}{Rk_b T}\right)$$



# 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

$$\frac{dR}{dt} = \dot{R} = \frac{\Omega}{R} (D_v C_v - D_i C_i)$$

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

Next slide

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



# Void Growth

Remember:

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

and

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

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

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:

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

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

Where:

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



# Void Growth

- With everything defined,

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

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

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

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

- We can now rewrite the growth law as:

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



# 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:

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

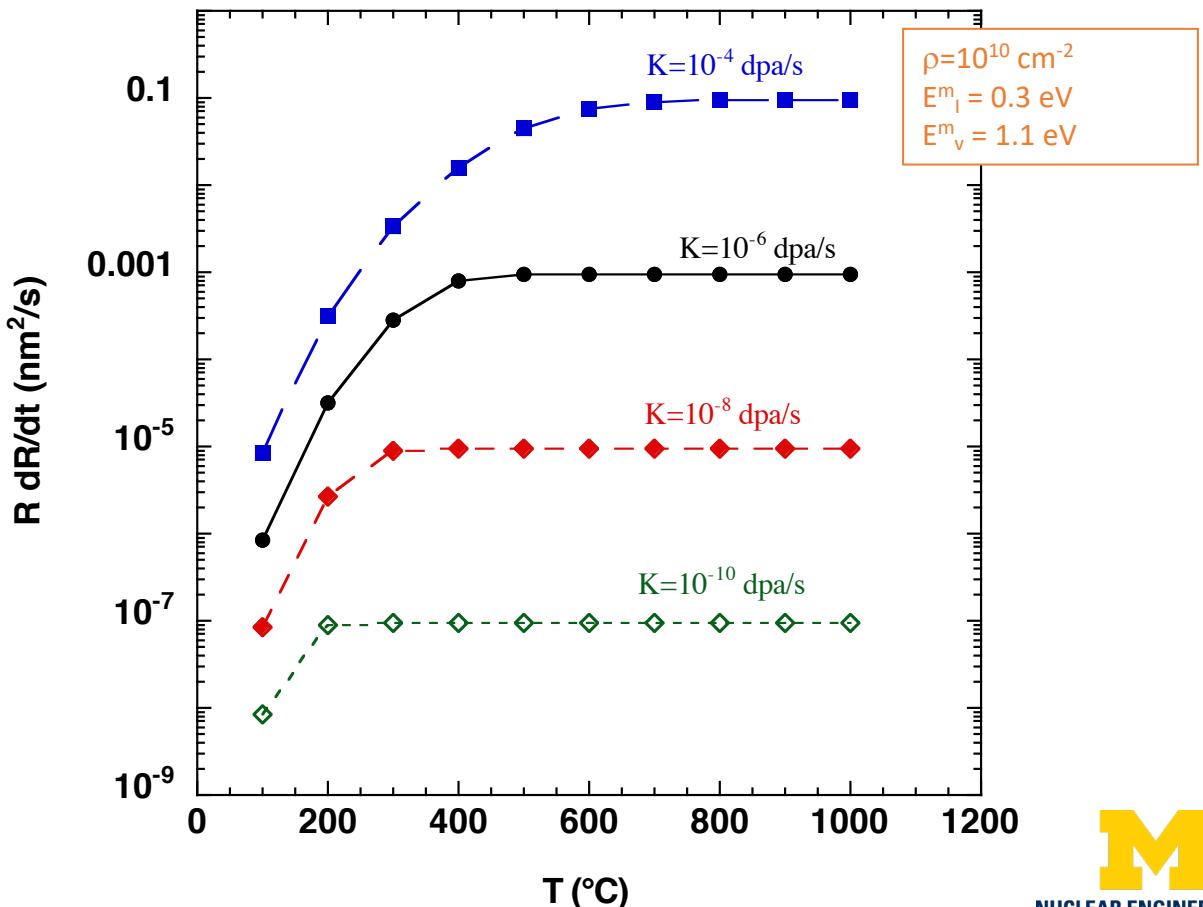
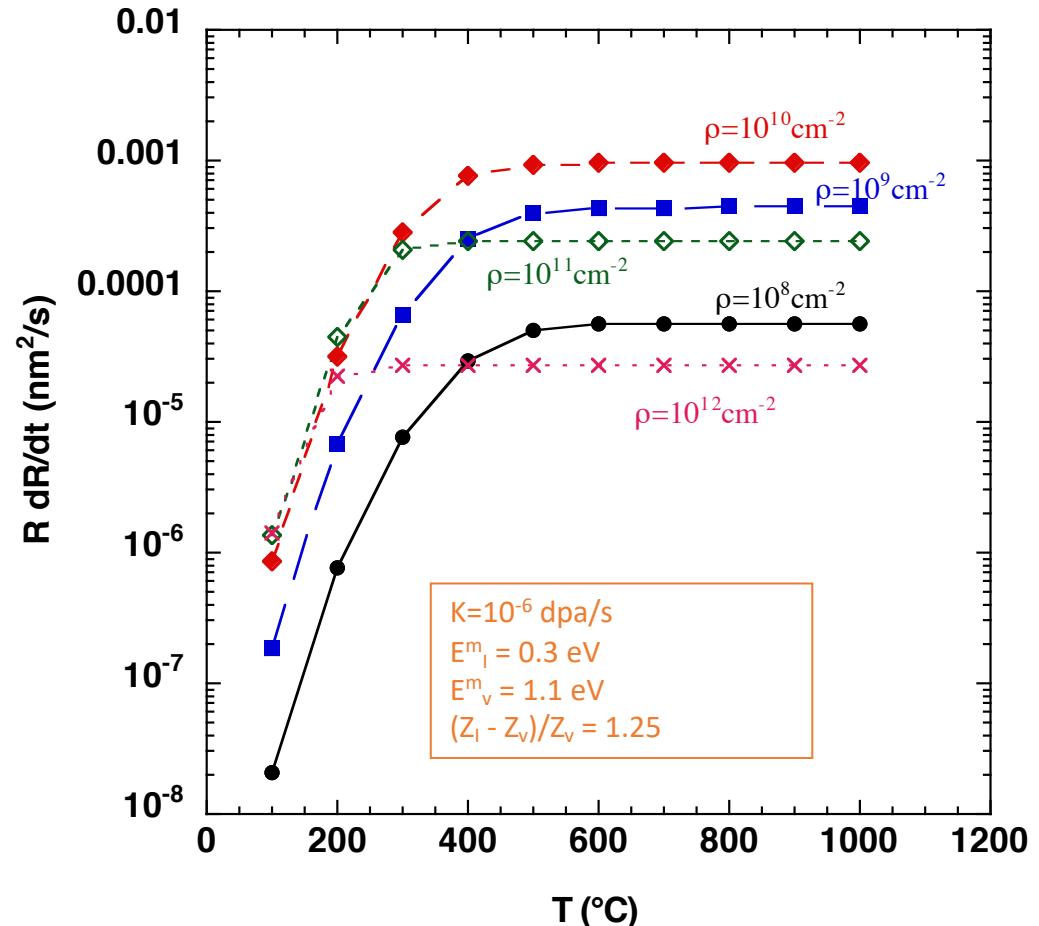
Since  $\eta$  decreases with increasing temperature and  $F$  decreases with increasing  $\eta$ :

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

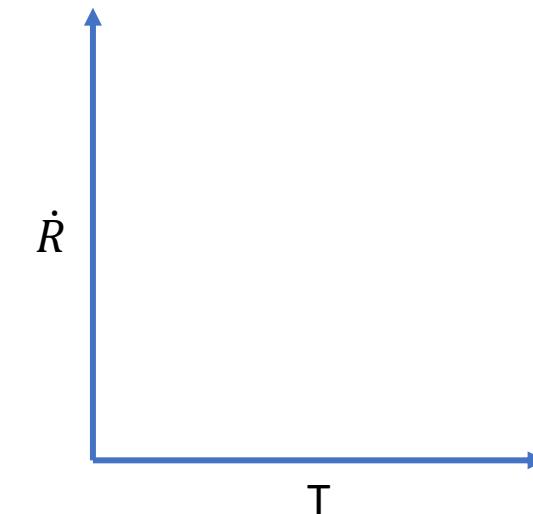
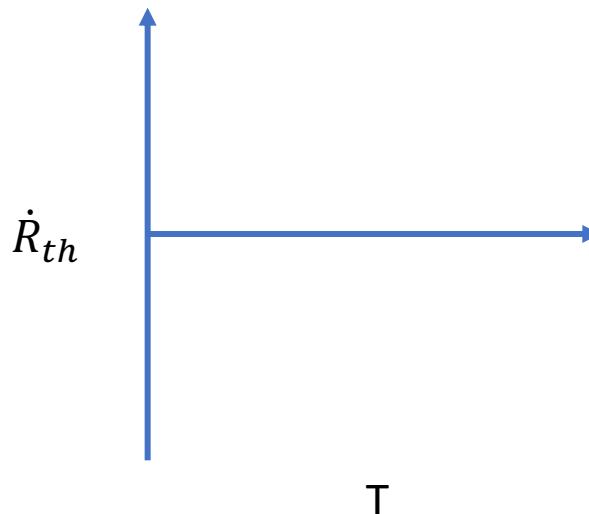
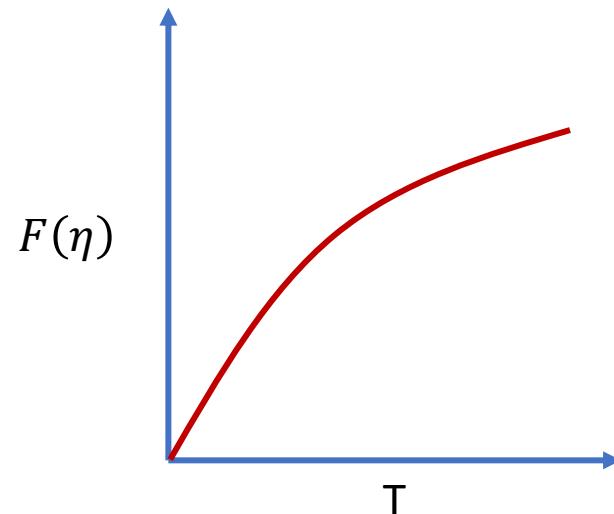


# Void Growth

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



# 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:

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



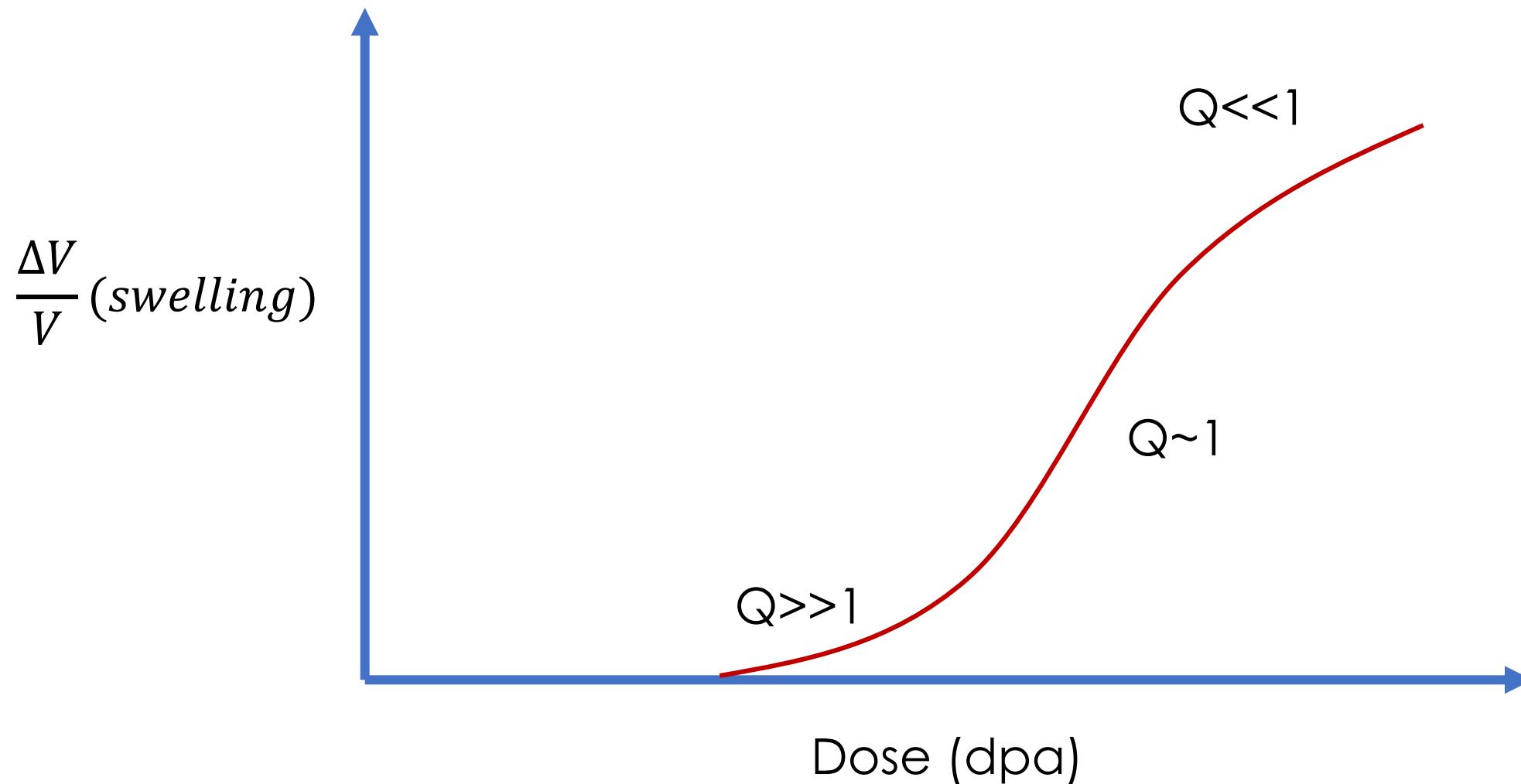
# Effect of Structure on Void Swelling

- Ferritic steels swell at rates ~0.2%/dpa
- Structure alone is not sufficient to explain the difference between  $\alpha$ -Fe (BCC) and  $\gamma$ -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 3<sup>rd</sup> term, put simply:

$$\left\{ \frac{z_v \rho_d}{(4\pi R \rho_v + z_v \rho_d)(4\pi R \rho_v + z_i \rho_d)} \right\} \frac{Q}{(1+Q)^2} \quad \text{Where: } Q = \frac{\rho_d z_d}{z_v 4\pi R \rho_d}$$

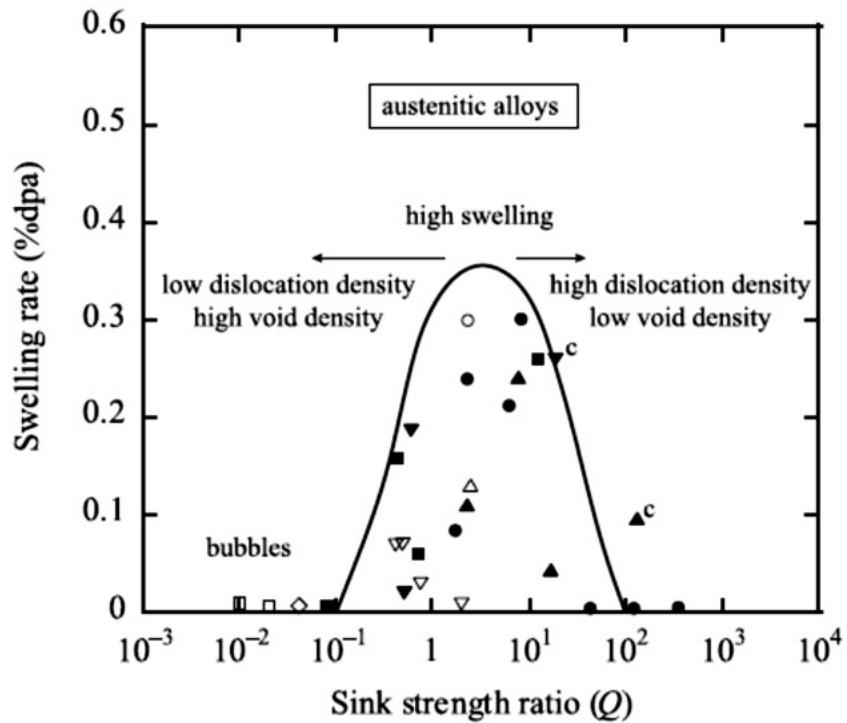


# The Q-factor for structure dependence on dose



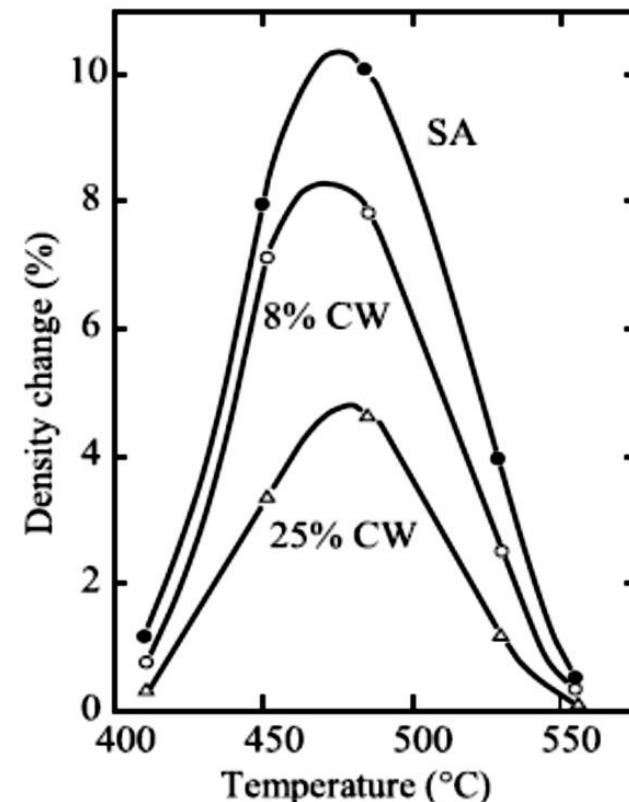
# Effect of Structure on Void Swelling

● Johnston et al.  
▲ Appleby et al.  
■ Packan and Farrell  
▼ Maziasz  
▽ Sprague et al.  
○ Westmoreland et al.  
□ Tanaka et al.  
△ Smidt et al.  
◊ Lee and Mansur  
c coalescence



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

- Growth rate is maximum when  $Q \sim 1$
- Growth decreases for  $Q \neq 1$
- Observed experimentally
- CW reduces swelling because  $Q \gg 1$

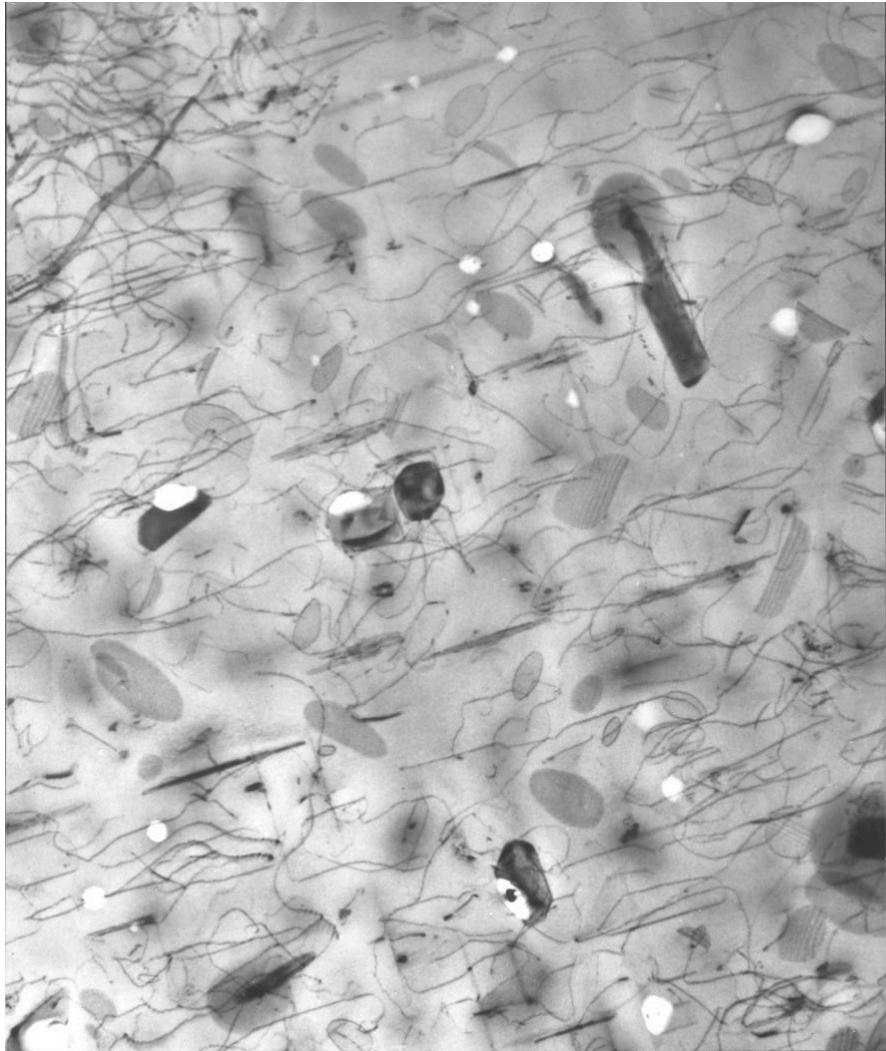


*Dependence of swelling on cold-work for various temperatures for 316 stainless steel irradiated in the RAPSODIE reactor to doses of 20–71dpa (Dupouy JM, Lehmann J, Boutard JL (1978) In: Proceedings of the Conference on Reactor Materials Science, vol. 5, Alushta, USSR. Moscow, USSR Government, pp 280–296)*

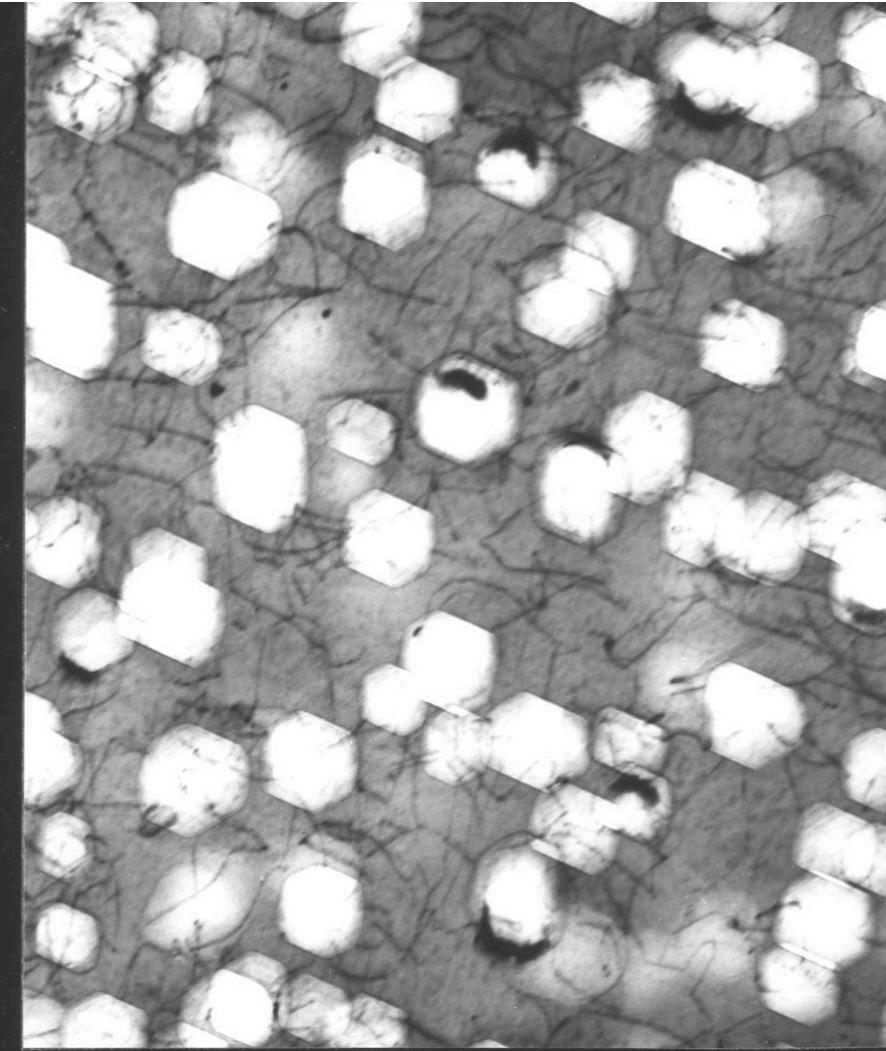


# Image of voids + effect of sink strength

Commercial 316 SS (high sink strength)

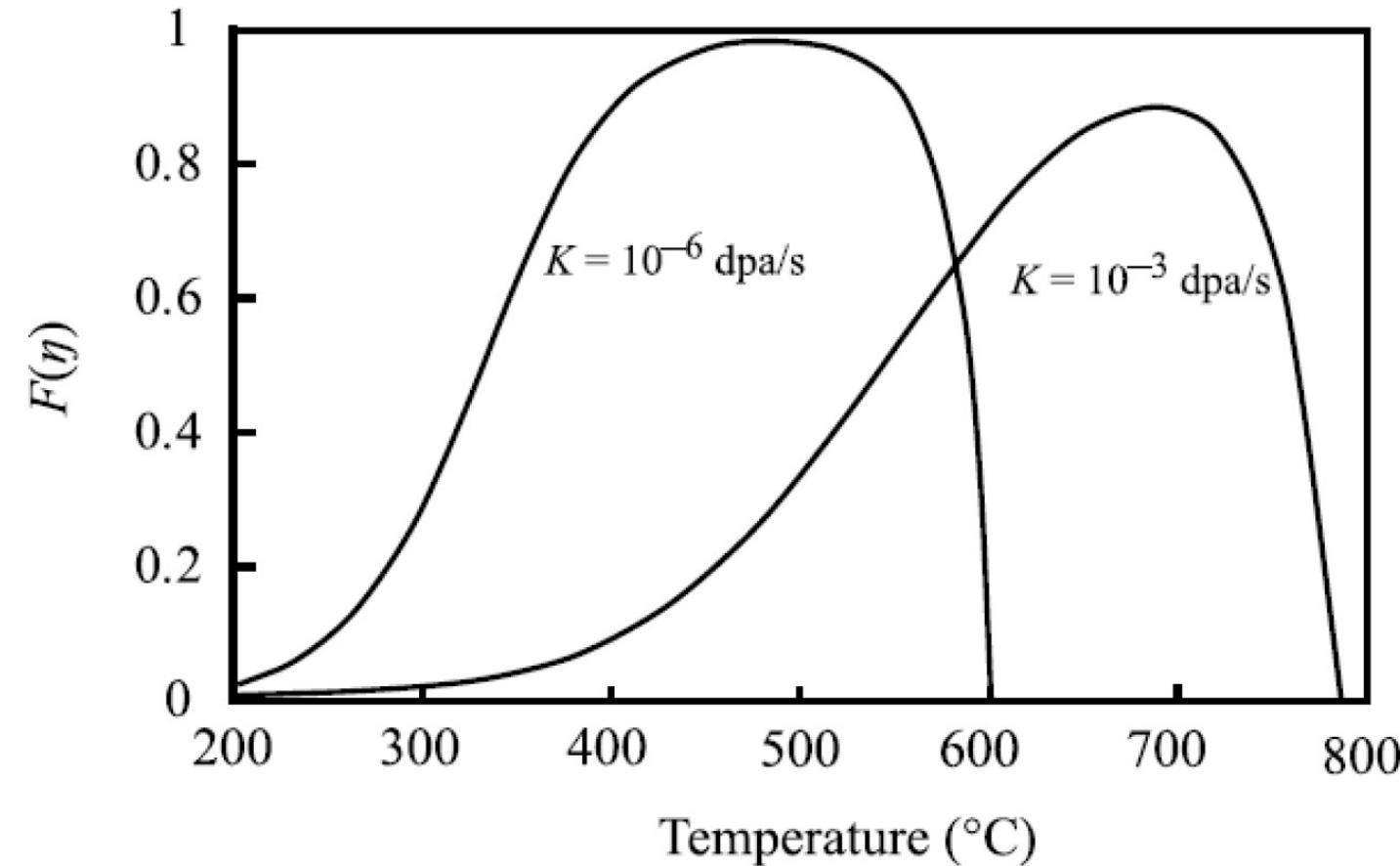


High-purity (low sink strength)



100 nm

# Effect of Dose Rate



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

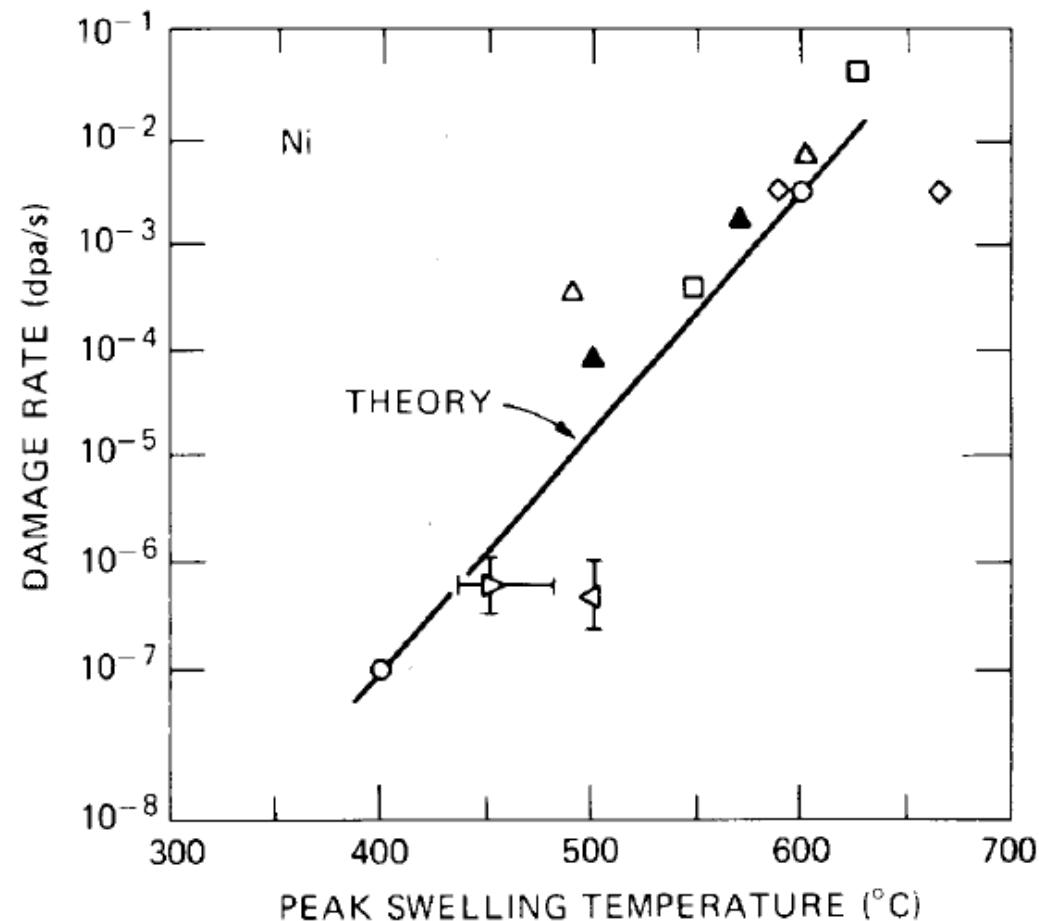
$$F(\eta) = 2\left(\sqrt{1 + \eta} - 1\right)/\eta$$

And:

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



# 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:

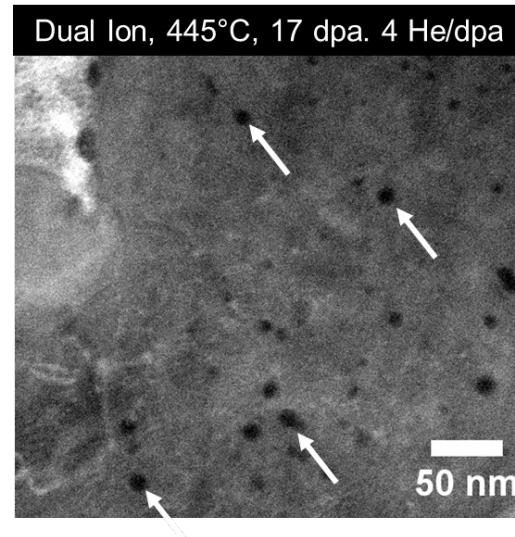
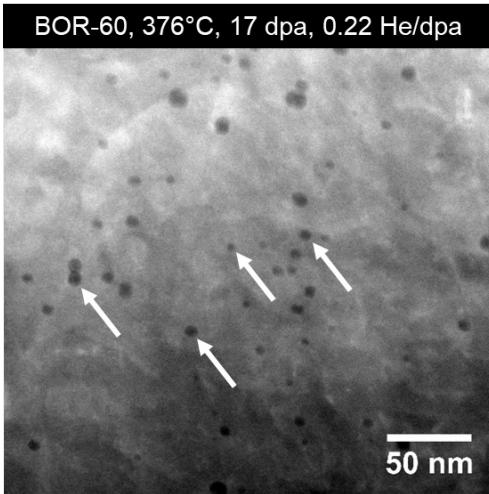
$$F(\eta) = 2\left(\sqrt{1 + \eta} - 1\right)/\eta$$

And:

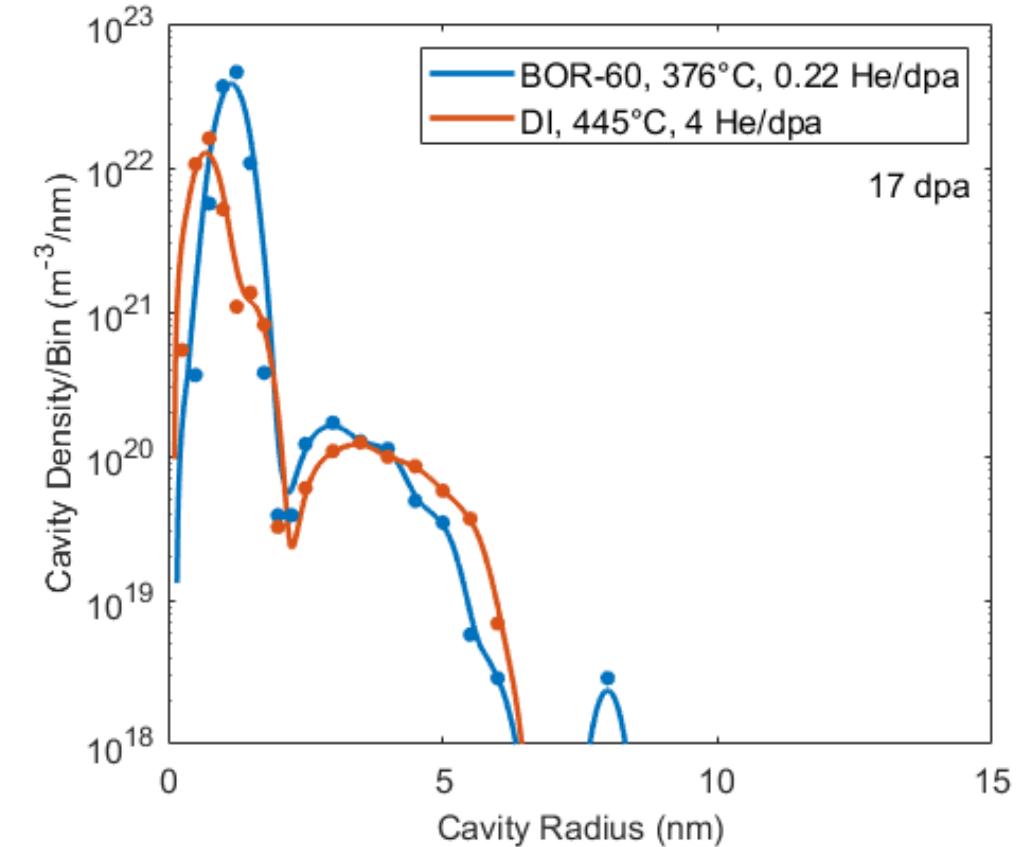
$$\eta = \frac{4K_0 K_{iv}}{D_i D_v (4\pi R \rho_v + z_v p_d)^2}$$

# Effect of Dose Rate – Real World Example

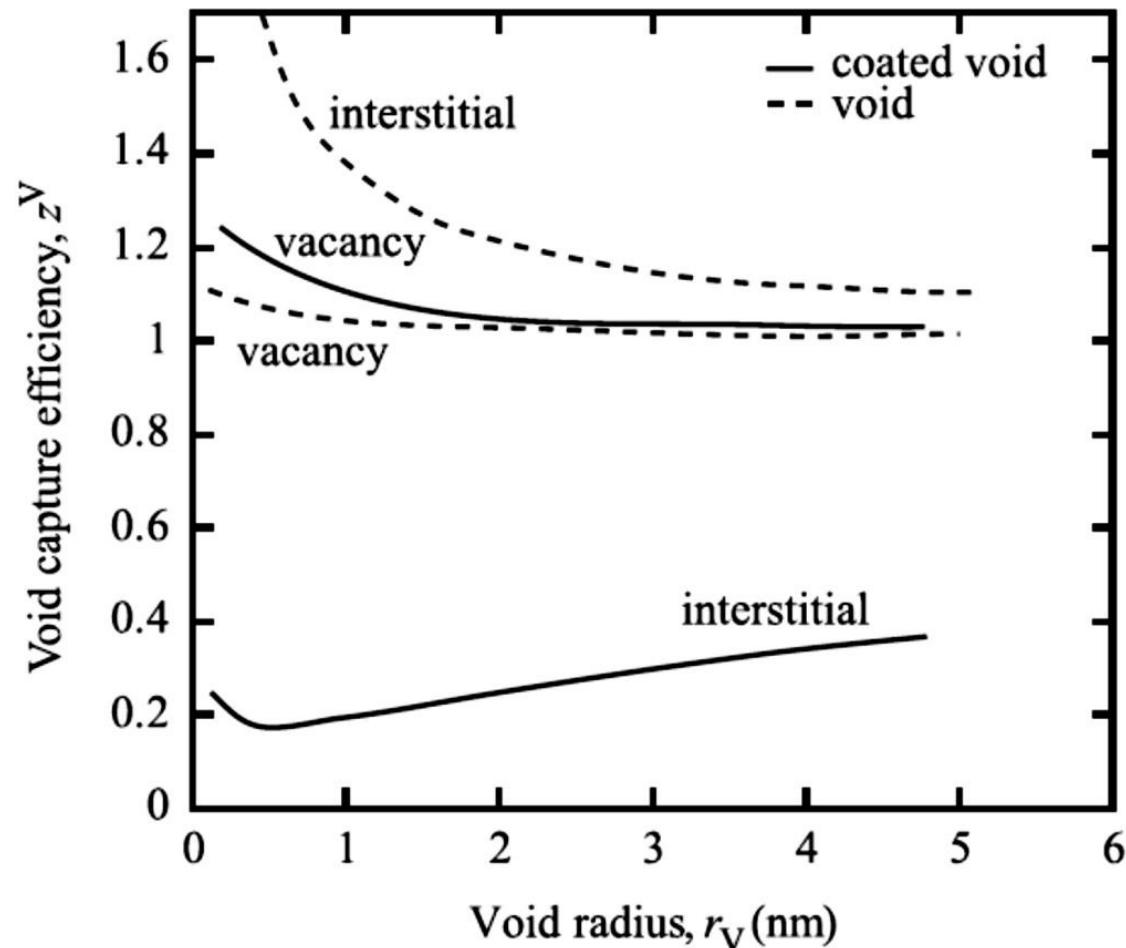
## STEM HAADF



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



# Effect of void surface segregation on defect bias



- 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

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

# 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
- 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:

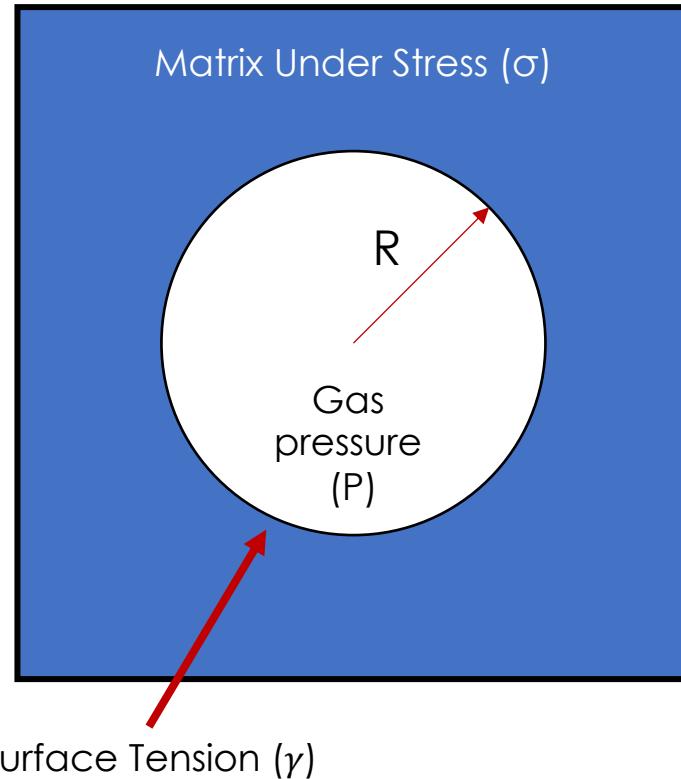
Void: open volume in a solid not pressurized by inert gas

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

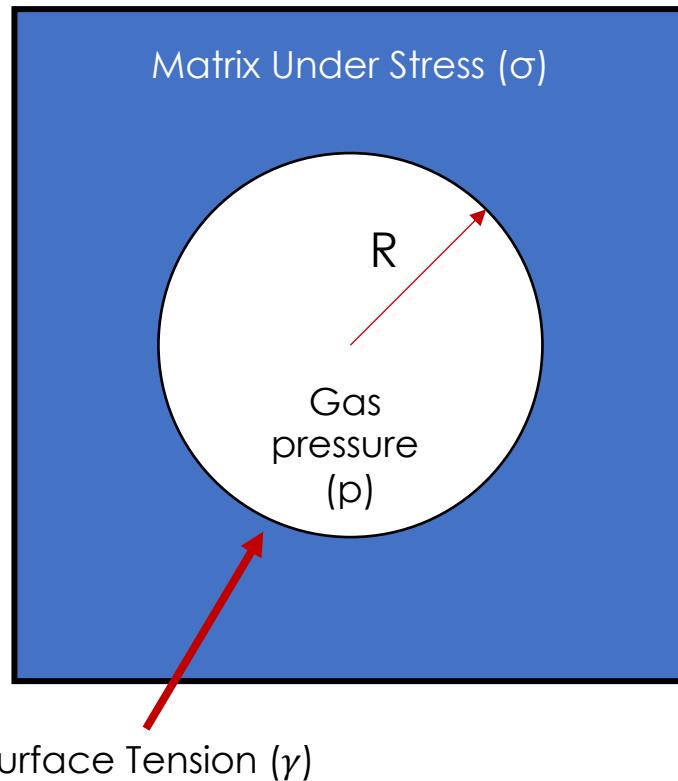
Cavity: Generalization for open volume in a solid – can be a bubble or void

# Effect of Inert Gas: Bubbles & Voids

- 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:



# Effect of Inert Gas: Bubbles & Voids

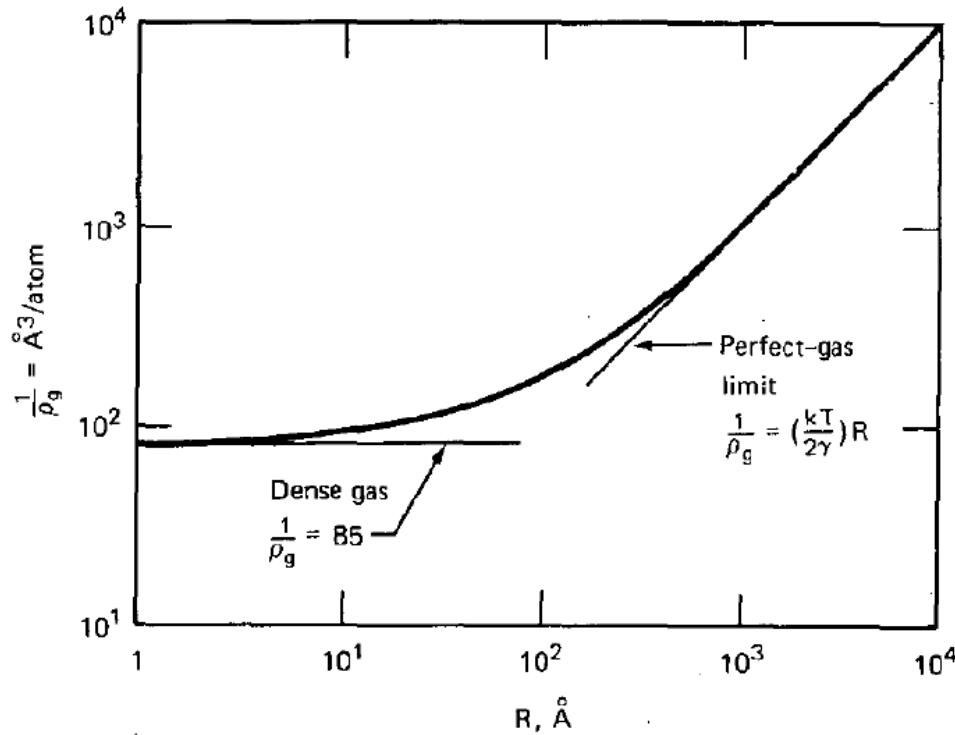


Fig. 13.3 Density of xenon gas in a spherical bubble imbedded in a stress-free solid of surface tension of 1000 dynes/cm.

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**:

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

And for non-equilibrium bubbles:

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

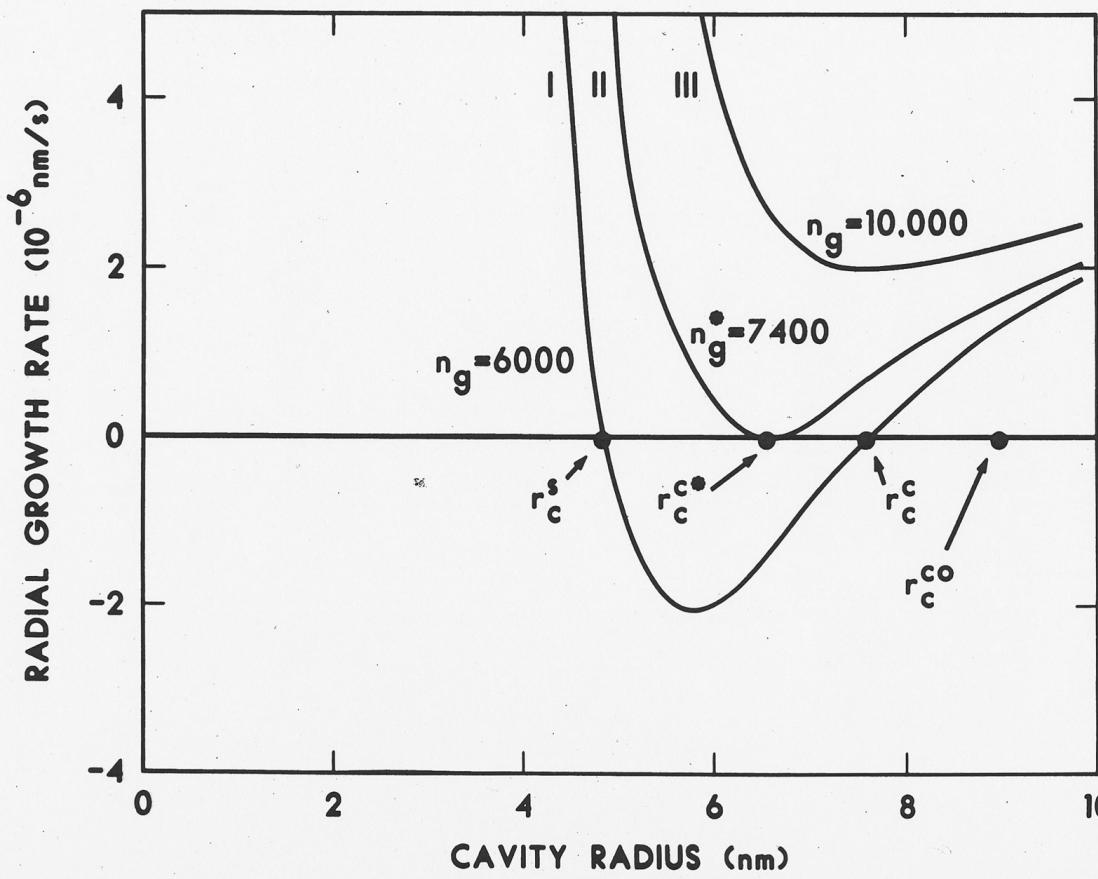


Now that we have an expression for  $n_x$  and  $P$ , we can add these into the terms for the growth rate law including thermal emission, we then get:

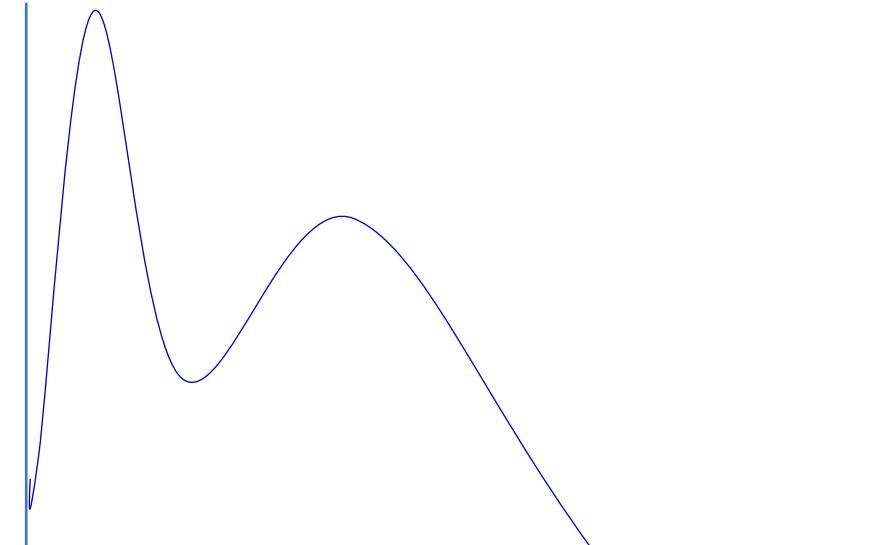
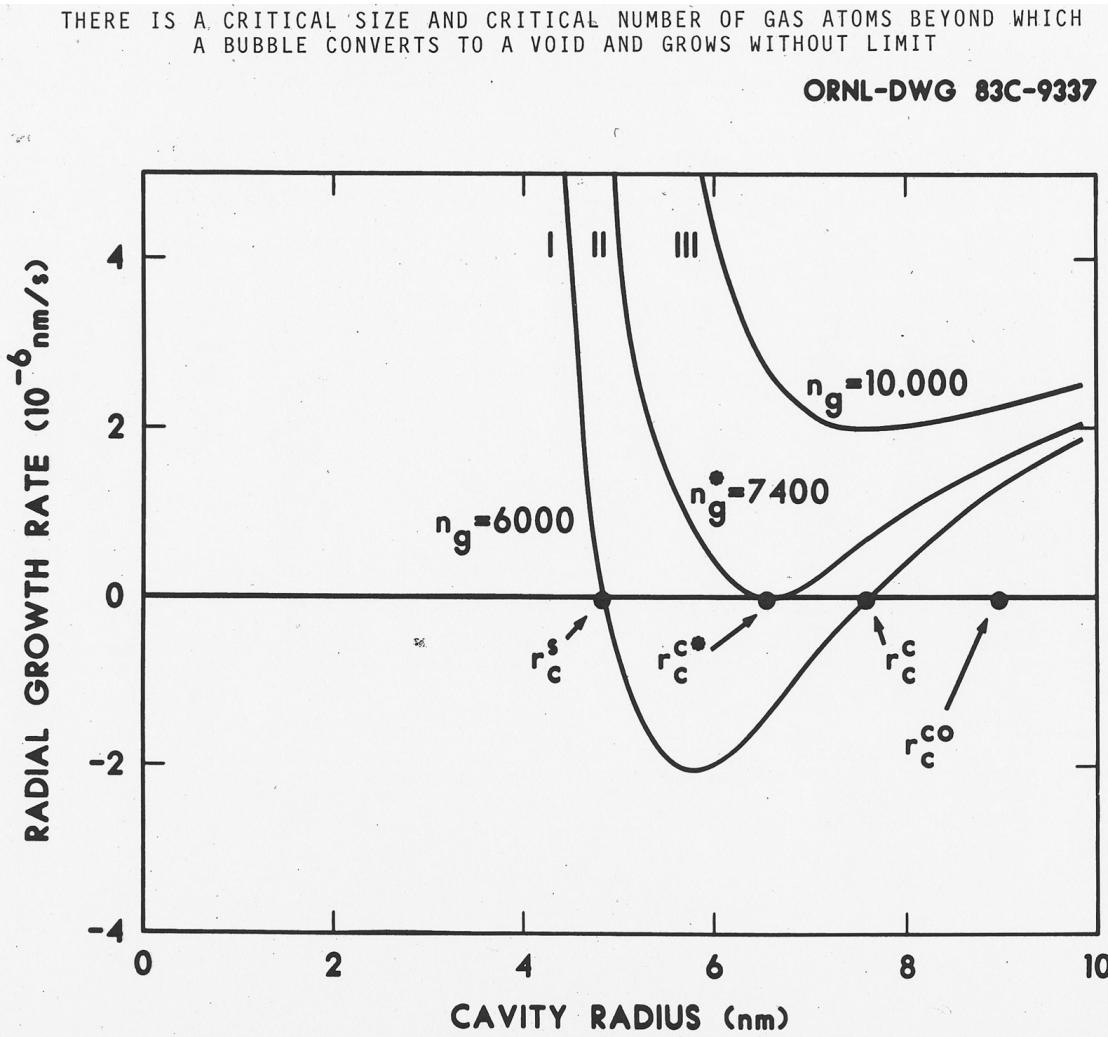
$$R\dot{R} = K_o \Omega \left( \frac{z_i - z_v}{z_v} \right) \frac{z_v \rho_d}{(4\pi R \rho_v + z_v \rho_d)^2} F(\eta) - \frac{D_v C_v^0 \Omega^2 z_v \rho_d}{kT(4\pi R N + z_v \rho_d)} \left( \frac{2\gamma}{R} - \frac{n_x kT}{4/3 \pi R^3 - n_x B} \right)$$

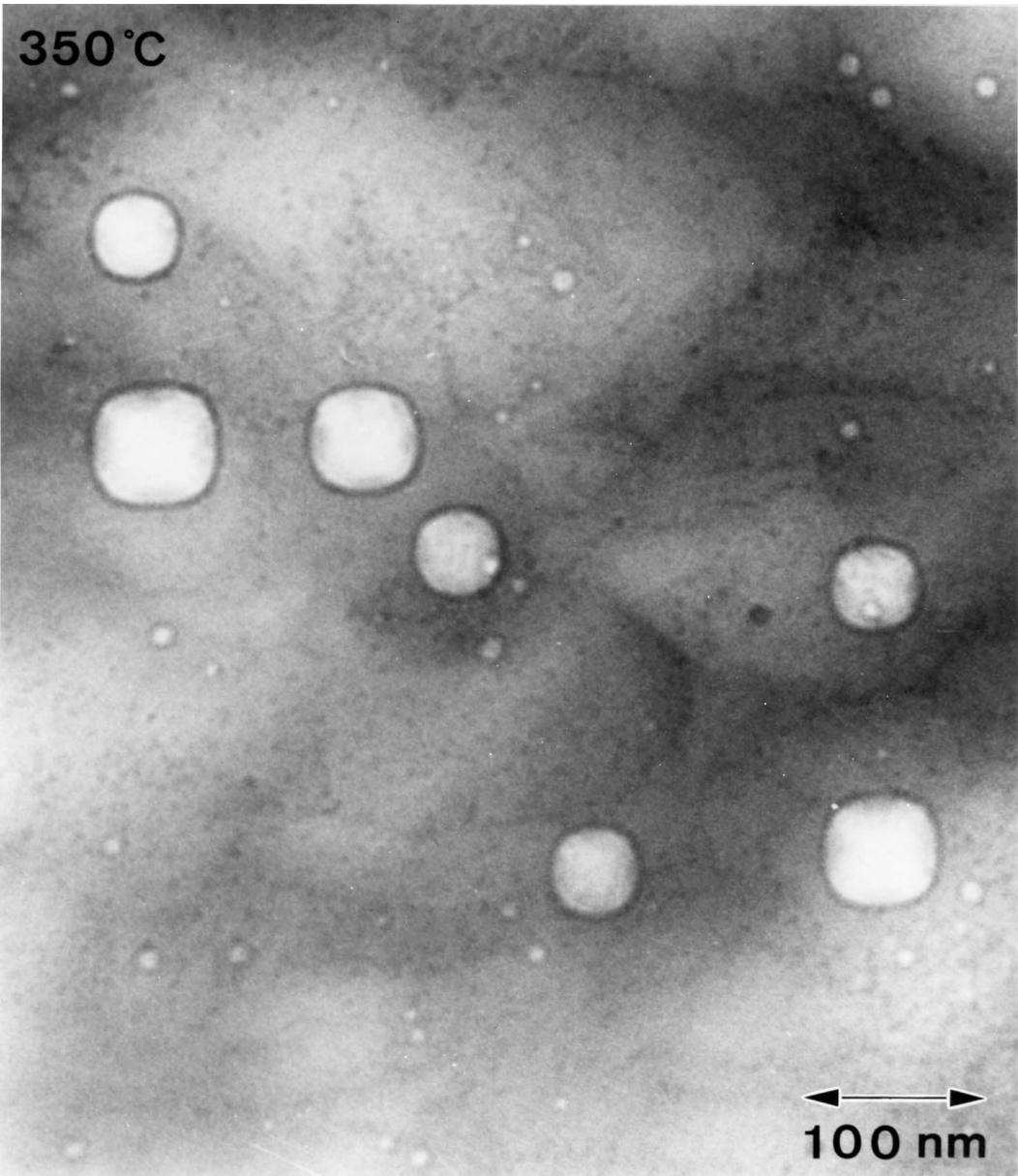
THERE IS A CRITICAL SIZE AND CRITICAL NUMBER OF GAS ATOMS BEYOND WHICH  
A BUBBLE CONVERTS TO A VOID AND GROWS WITHOUT LIMIT

ORNL-DWG 83C-9337



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





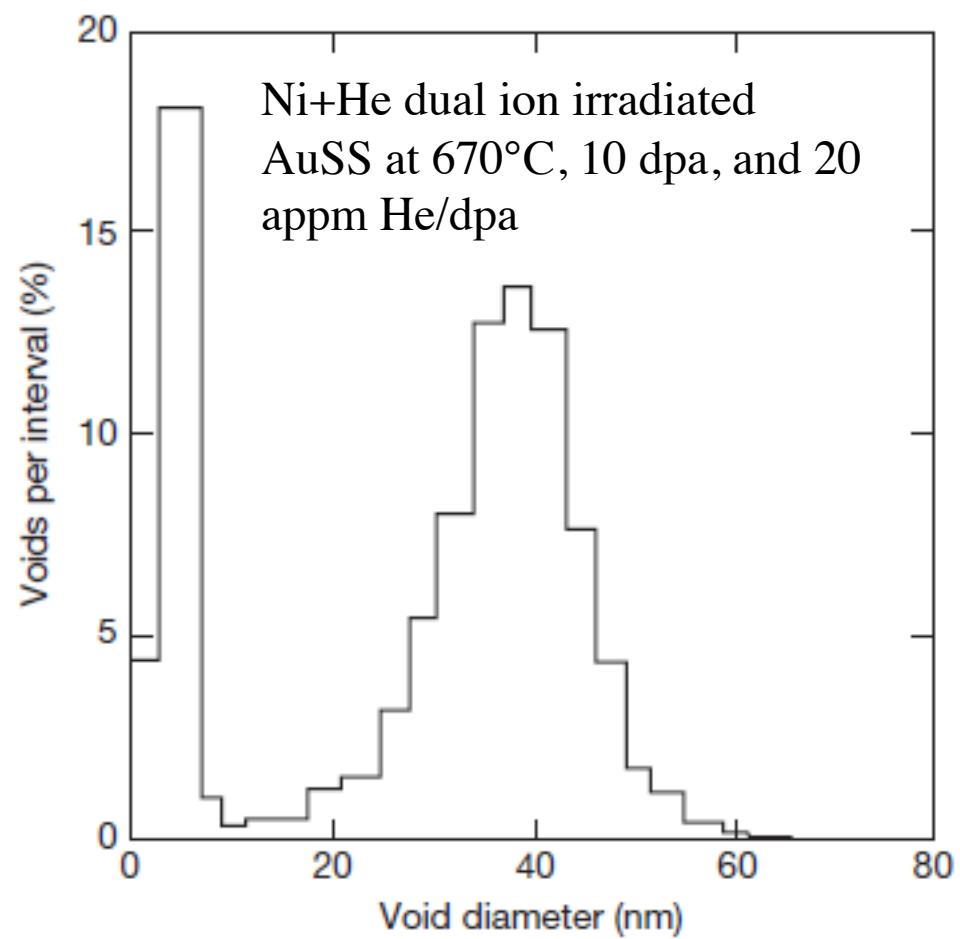
Void and He  
bubble formation in  
Cu-100 ppm B  
following fission  
neutron irradiation  
to 1.2 dpa at 350°C

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

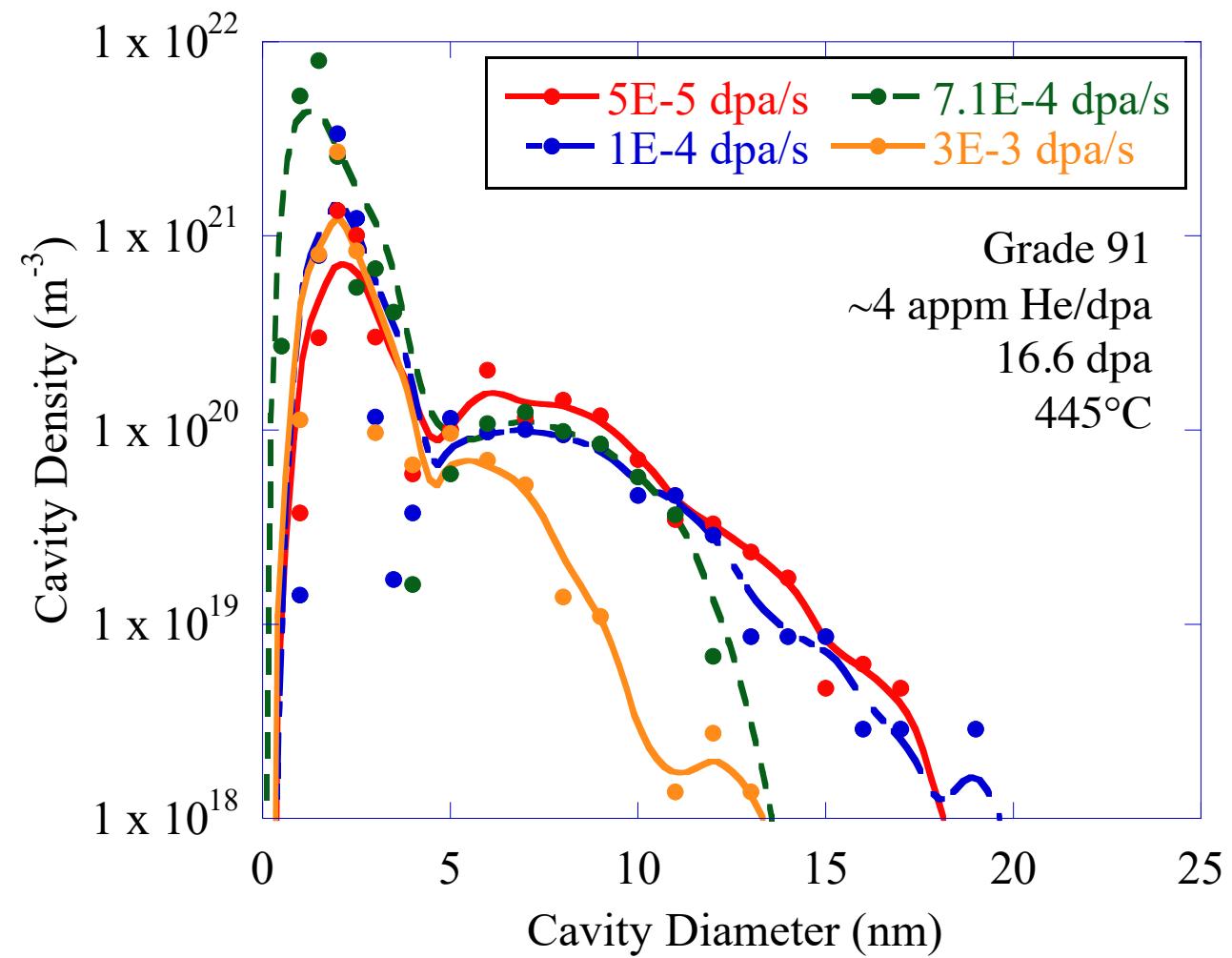


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# Experimental examples



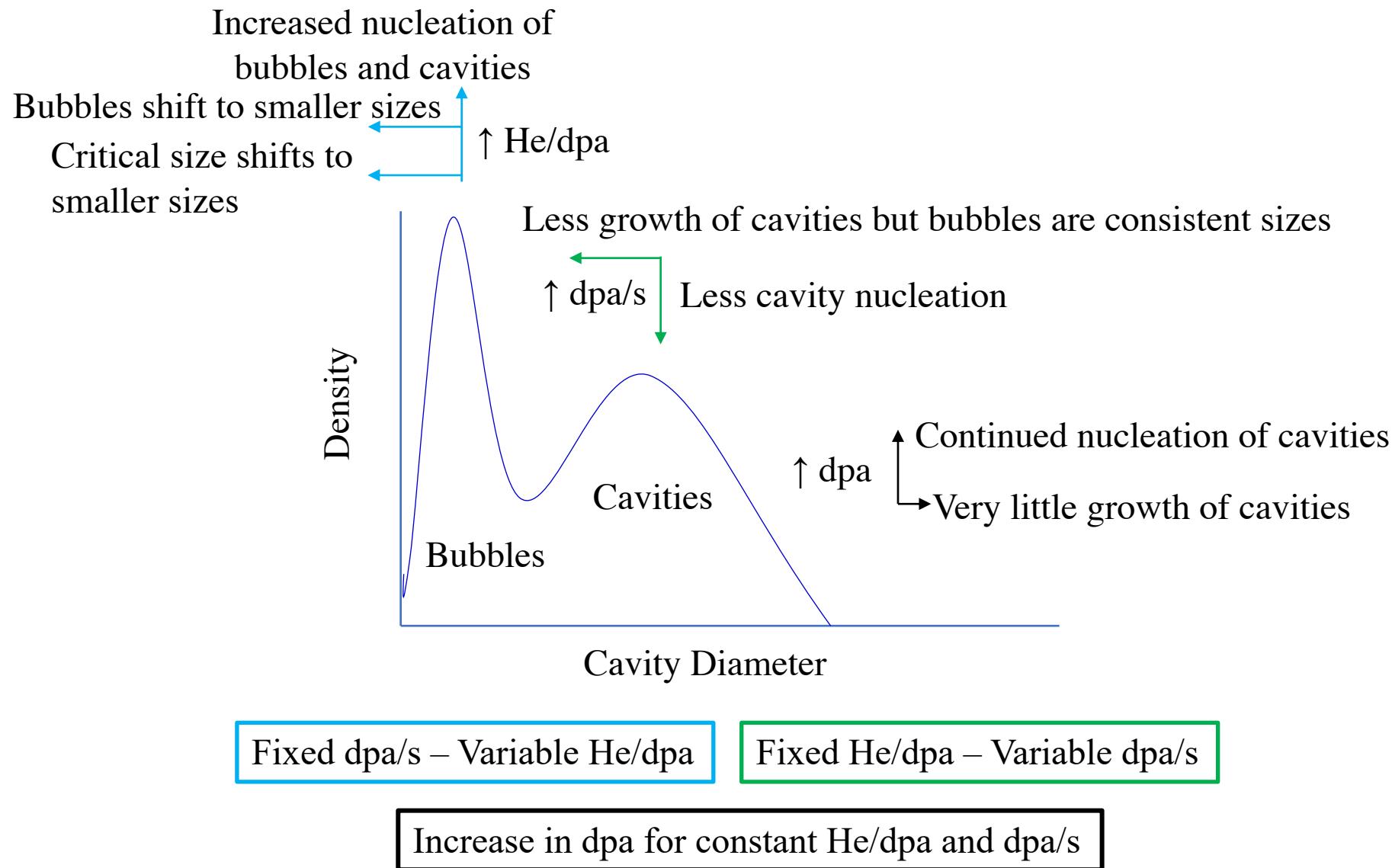
Mansur, Coghlan JNM 119 (1983)



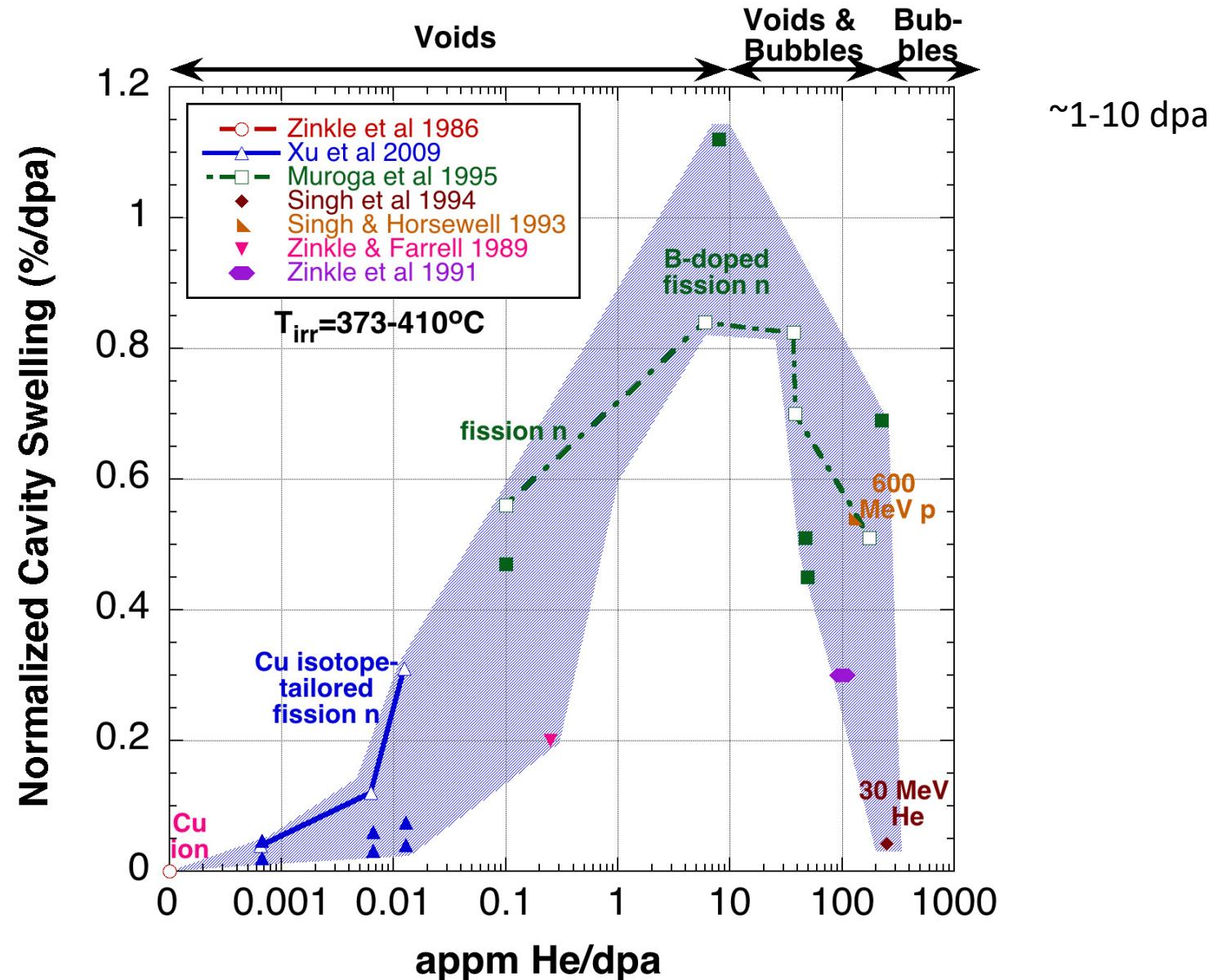
Taller (2019)



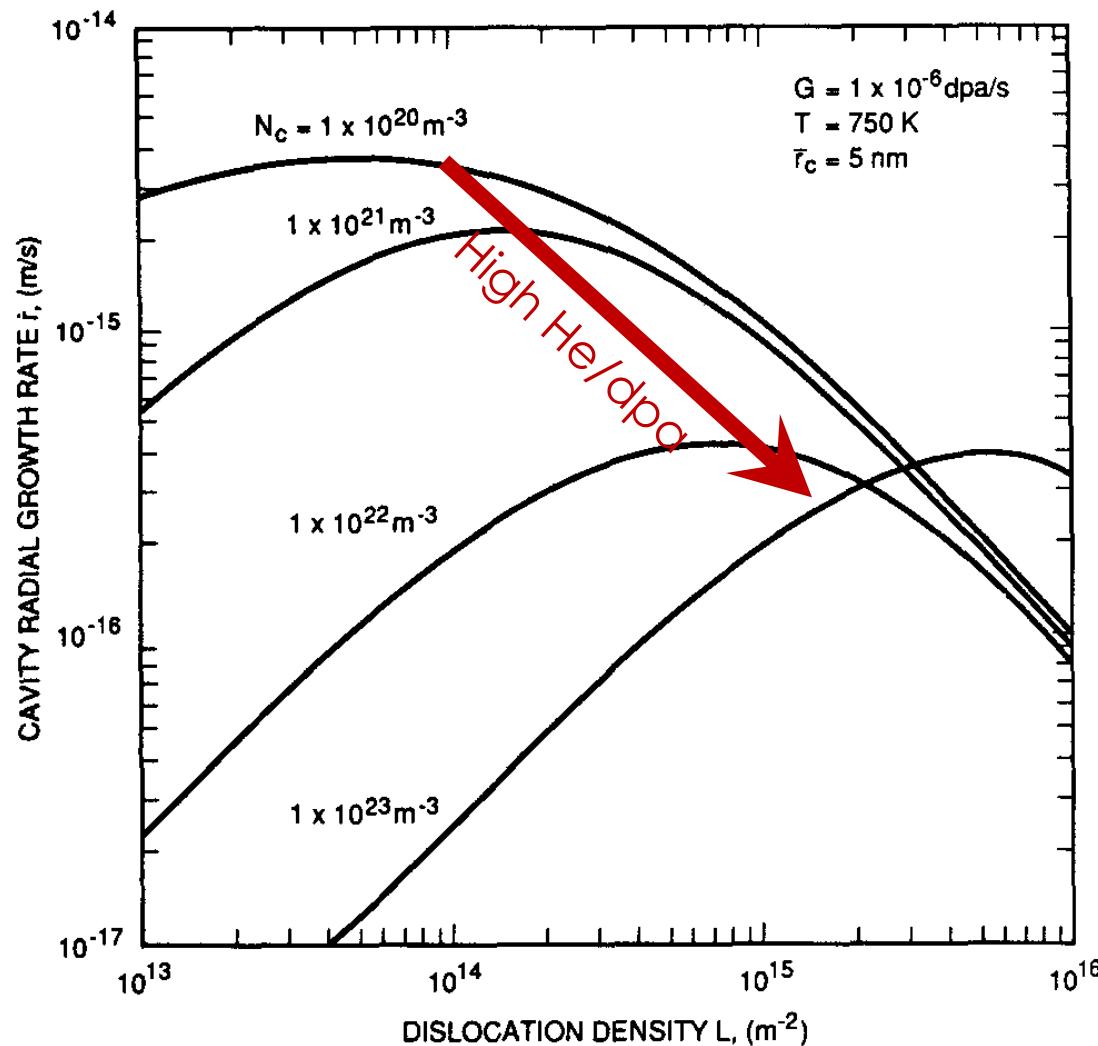
# Experimental examples



# Cavity swelling vs. He/dpa ratio in irradiated copper

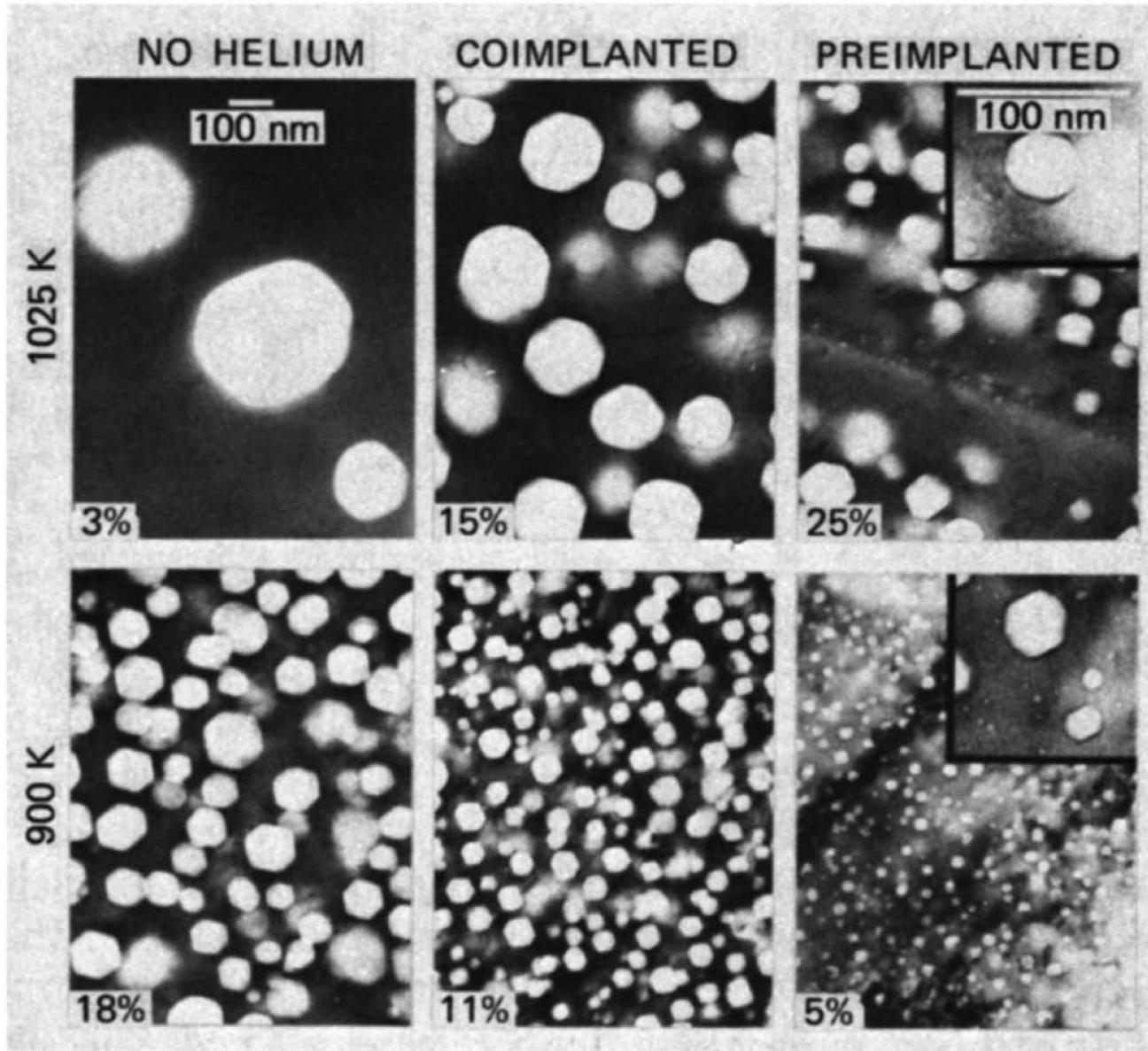


# Calculated void growth rate is typically reduced for high cavity and dislocation sink strengths



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

# 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

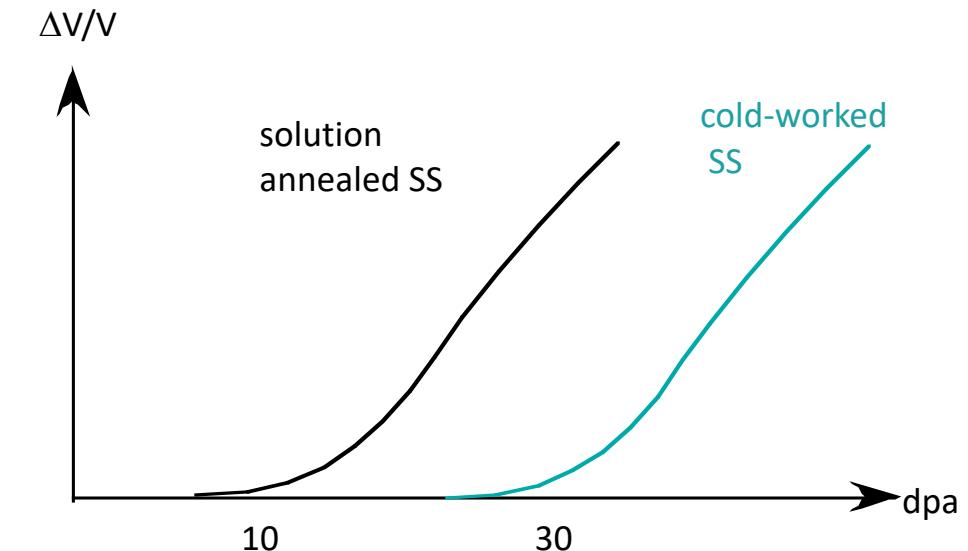
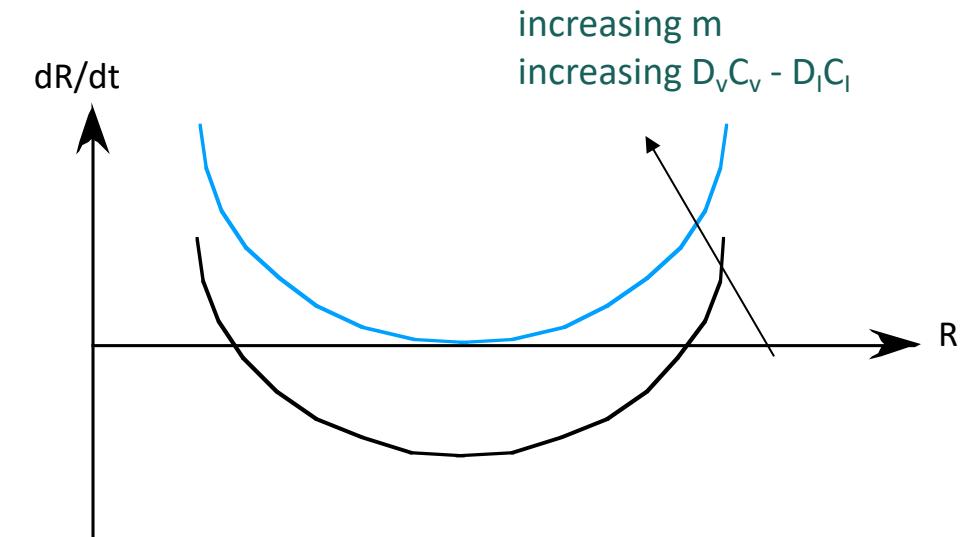
Packan & Farrell, NT-Fusion, 1983

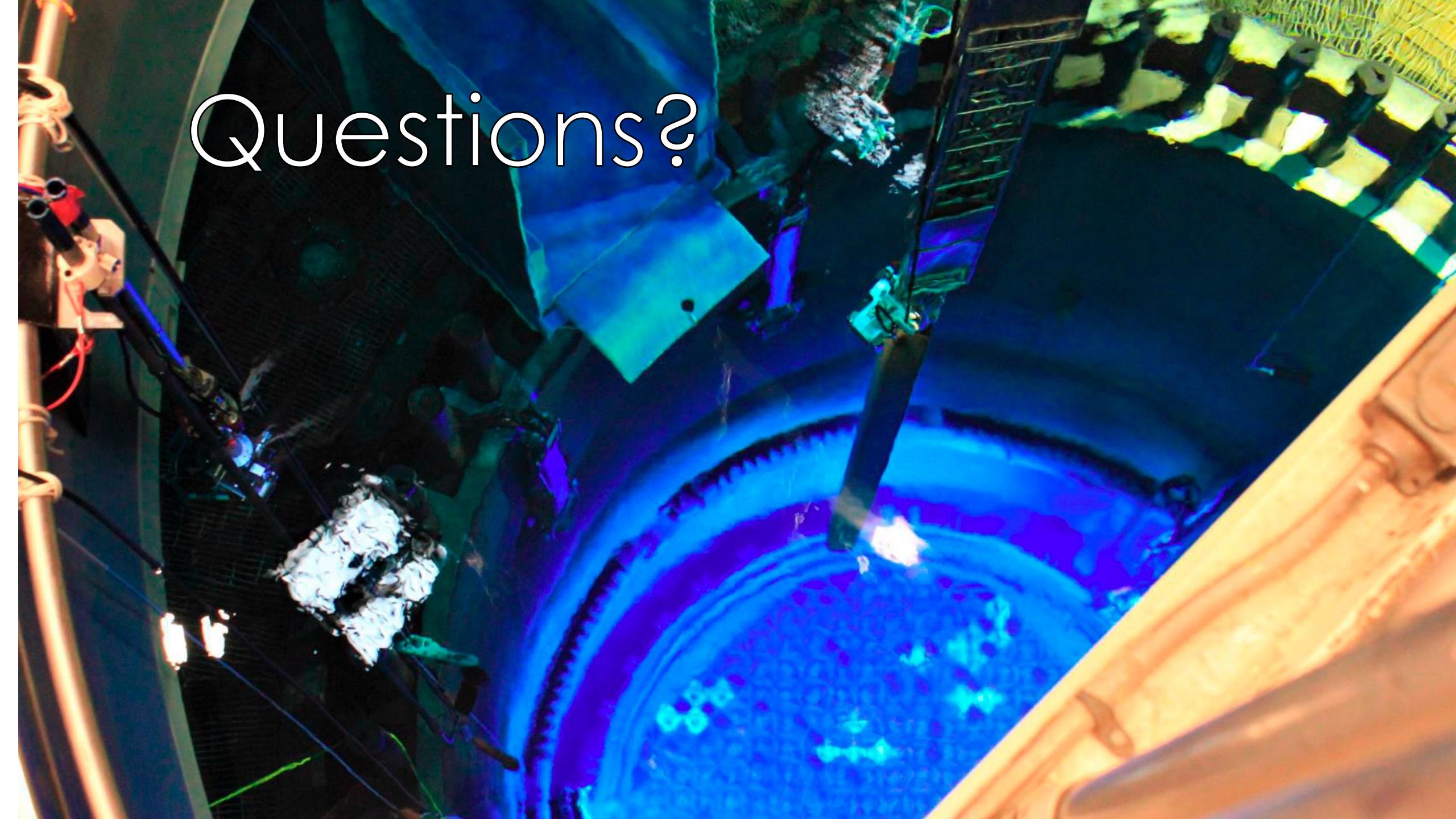
# Remedies for void swelling?



# Remedies for void swelling?

- Decrease  $D_v C_v - D_l C_l$  arriving at cavity;
- Eliminate He gas production  
(expensive or impractical)
- Reduce  $C_v, C_l$ :
  - 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





Questions?