Effect of Crystal Structure and Defect Diffusion on Damage Accumulation inSolids under Irradiation

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Effect of Crystal Structure and Defect Diffusion on Damage Accumulation in Solids under Irradiation

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Main Objective

Discuss the differences in damage accumulation in BCC, FCC and HCP metals

Outline

- Brief Introduction
- Earlier RD Model (FP3DM)
- Modern RD Model (PBM)
- Damage accumulation in cubic and HCP metals in the framework of PBM

Mechanical properties change during irradiation

- **Mechanical properties of material depends on microstructure**
- **Evolution of microstructure depends on temperature, mechanical treatment and radiation damage**
- **Radiation damage: production of atomic displacement, i.e. production of site / anti-site types of defects: vacancies, self-interstitial atoms (SIAs) and their clusters**
- **Consequence of defect production: accumulation of SIAs in the form of dislocation loops, tetrahedra, voids and development of dislocation network**

Radiation may also lead to

- ❖ **Production of variety of impurities , e.g. He, H, etc.**
- ❖ **Nucleation and growth of secondary phase precipitates**
- ❖ **Radiation-induced segregation etc.**

These phenomena are out of scope of this presentation

Main Focus: Accumulation of defects in the form of dislocation loops, voids and dislocation

Radiation effects on properties

- ☐ Swelling: volume increase
- ☐ Growth: inelastic deformation at constant volume without stress
- ☐ Creep: inelastic deformation at constant volume under stress
- ☐ Hardening, Embrittlement: yield stress increase, plasticity decrease

Maximum scale in damage accumulation:

- ☐ swelling ~1.0%/dpa: (~one FP survives out of 100)
- ☐ radiation growth ~0.1%/dpa: (~one FP survives out of 1000)

Swelling, creep, hardening and embrittlement are common phenomena for BCC, FCC and HCP crystals

There are remarkable differences in how phenomena proceed in materials with different crystal structures

Differences in damage accumulation in cubic and HCP crystal

- BCC: swelling rate is small and not sensitive to temperature; void lattice formation is frequent and perfect; raft formation
- FCC: swelling rate is high, sensitive to temperatures; void lattice is not frequent and not perfect; SFT formation

Void lattice is not observed after 1 MeV electron irradiation !

- HCP: Radiation growth is a unique phenomena among metallic materials; alignment of voids and vacancy loops along basal planes instead of void lattice

Alignment of voids is observed after 1 MeV electron irradiation !

Although these facts have been known over the last 40 years, they have not been consistently explained

Basic reasons for luck of understanding

Main assumptions of earlier RD model (PF3DM/SRT)

1. Primary damage, regardless of nature and energy of particles, consists of single vacancies and SIAs
2. Both types of defects diffuse three-dimensionally
3. Preferential absorption of single SIAs by edge dislocations, i.e. dislocation bias, is considered to be the main driving force for damage accumulation

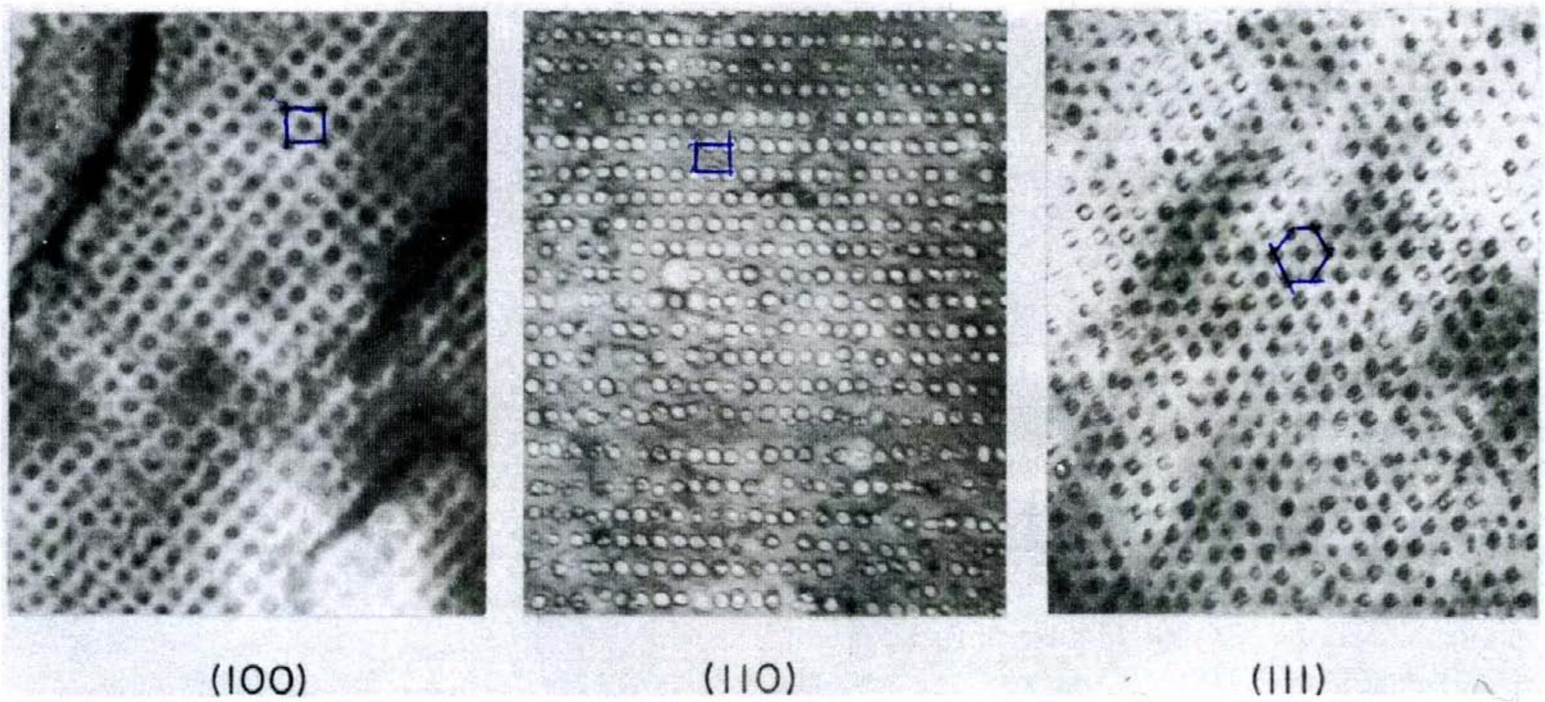
The model, which is still in use for all types of irradiation, is correct if all three assumptions are valid, for example, it may work for irradiation with 1 MeV electrons

Experiment, MD simulations and theory have revealed that:

All three assumptions are wrong
in the case of neutron and heavy ion irradiation

The main driving force for damage accumulation is the vacancy and SIA type of defects by dislocations irrespective of the difference in absorption

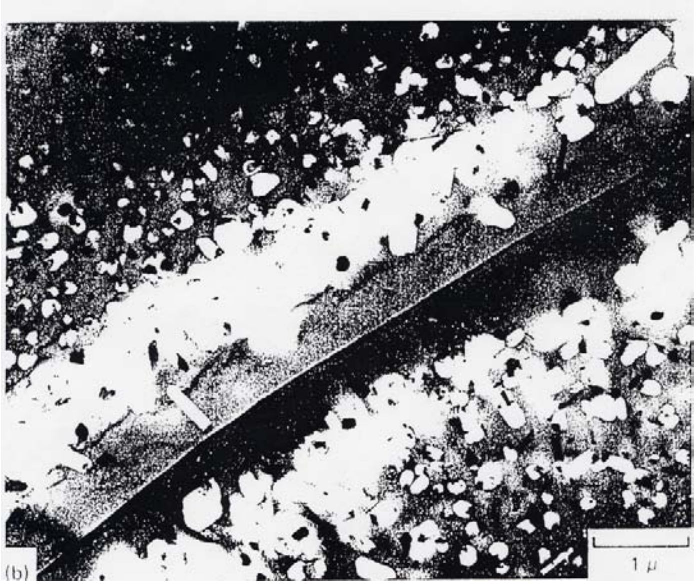
Void lattice in neutron-irradiated Niobium



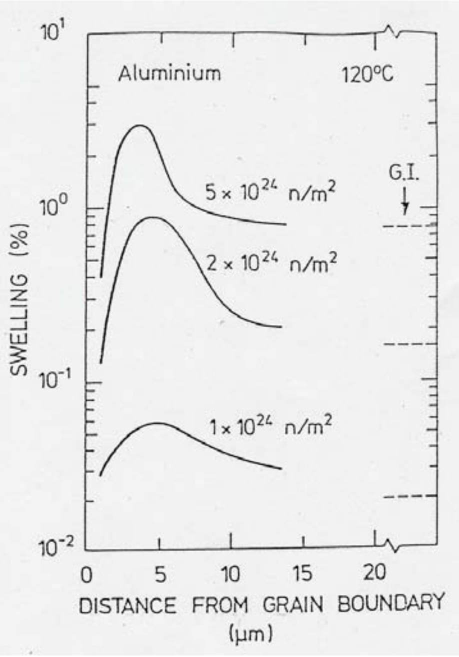
B. A. Loomis et al, J. Nucl. Mater. 68 (1977) 19

Void lattice has the same <u>symmetry</u> and <u>orientation</u> as the host lattice (BCC in BCC metals an so on)

Grain boundary effect in Al irradiated with neutrons



K. Farrell et al., Albany 1971, p.376

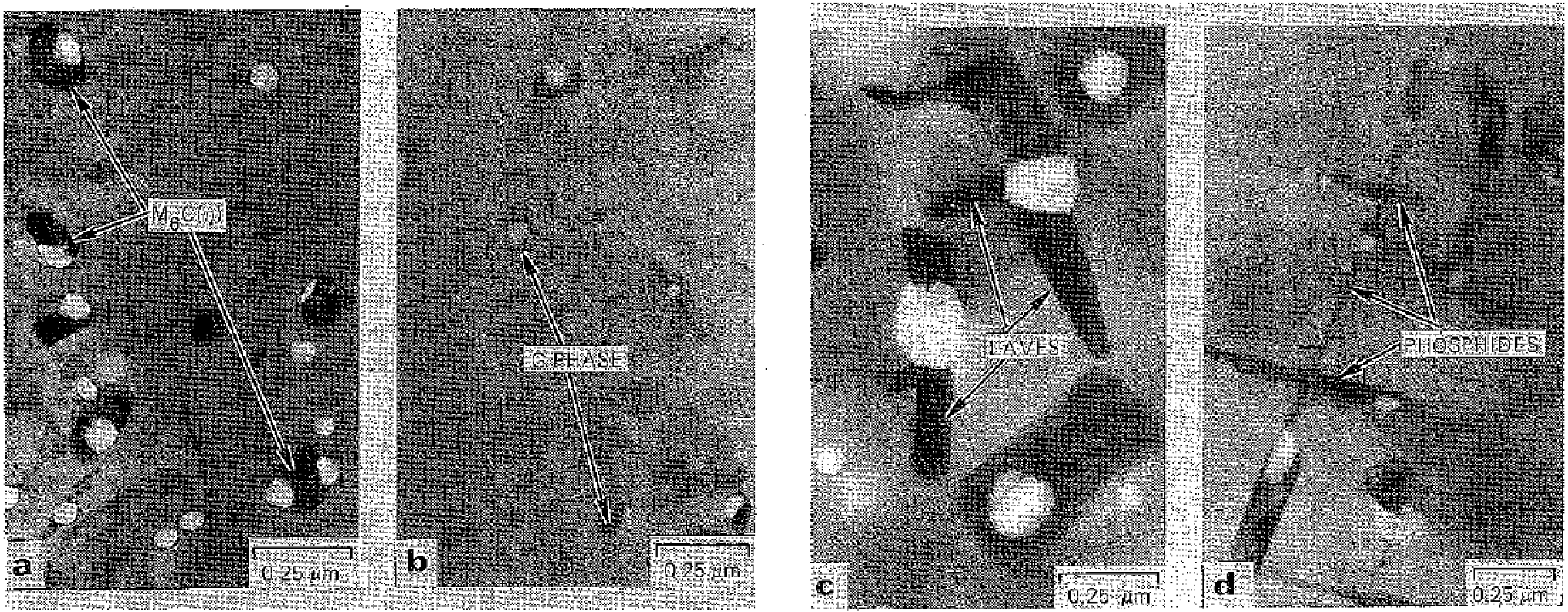


Foreman, Singh, Horsewell
Mater. Sci. Forum 15-18 (1987) 895

Length scale is orders of magnitude larger mean distance between voids

Spatial correlations between voids and second-phase particles

η-phase **G-phase** **Laves** **Phosphide**

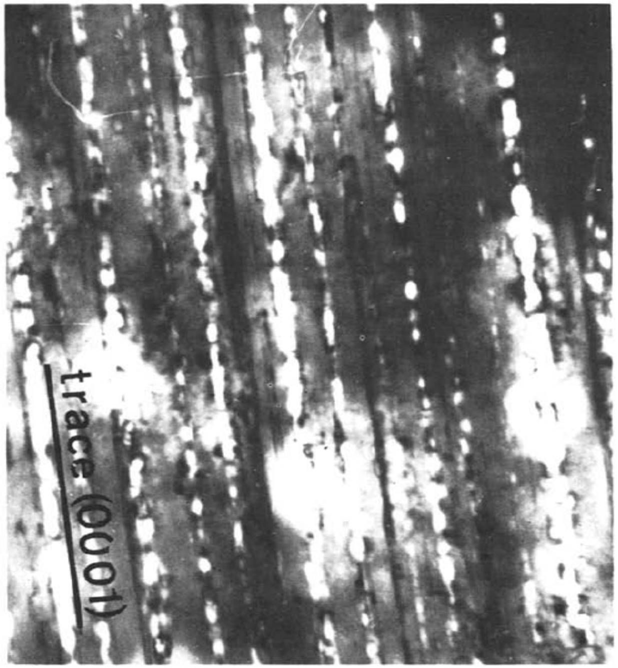


Pedraza & Maziasz (1987)

Space distribution of voids is not random

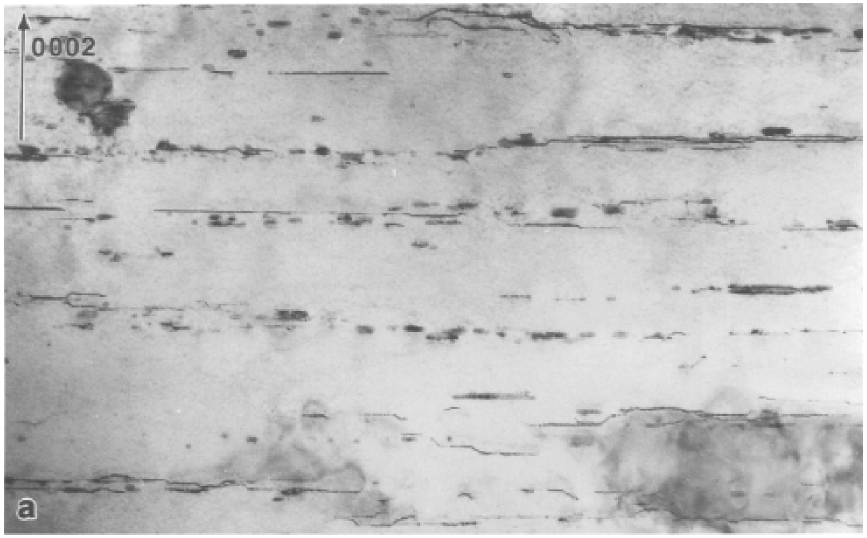
Alignment of Voids and Vacancy Loops Along Basal Planes in HCP Metals

Voids in Mg



Risbet & Levy, J. Nucl. Mater. 50 (1974) 116.

Vacancy loops in Zr



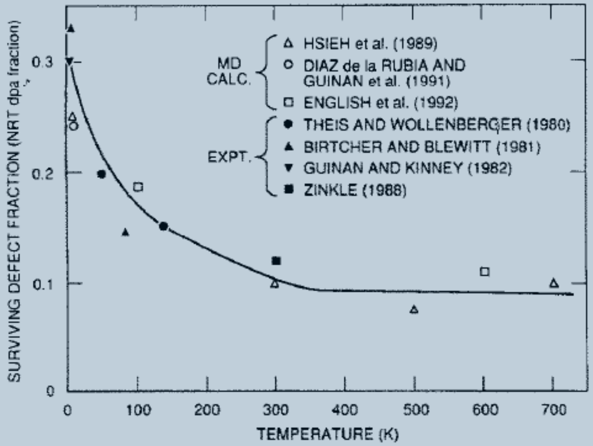
Griffiths et al. / Journal of Nuclear Materials 225 (1995) 245

Common point in all observations:

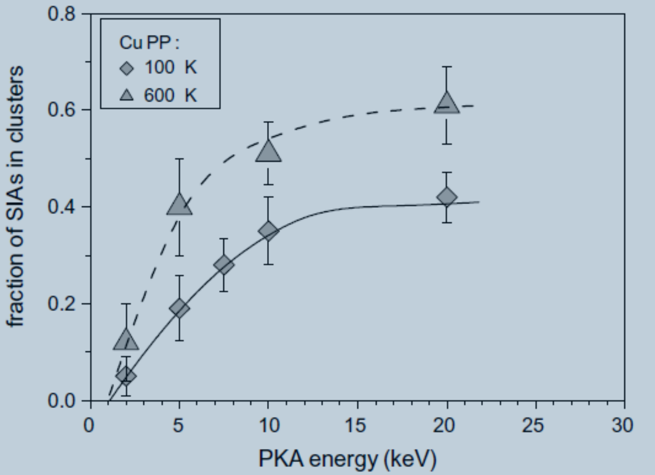
Non-random space distribution of vacancy type of defects

Primary damage under neutron irradaition

20keV cascade in iron (MD)



Temperature dependent fraction of defects surviving after cooling down phase of cascades in copper irradiated with neutrons.



Fraction of SIAs in clusters after cooling down phase of cascades as a function of PKA energy

- **Survival fraction is ~ 10 times less than that predicted by NRT standard**
- **Large fraction of defects survived if form of clusters**

The scale of effects is weakly dependent on the type of lattice and material composition since $E_{PKA} >> E_{cohesive}$

Production Bias Model

Key assumptions of the model:

1. **Defect production depends on particle energy and consists of PDs and PD clusters for the cases of neutron and ion irradiations (multi- displacement cascades)**
2. **Diffusion of interstitial types of defects, both single SIAs and SIA clusters, could be three-, two- and one-dimensional depending on the crystal structure where as it is three dimensional for vacancies**
3. **Absorption of defects by dislocations is determined by properties of migrating defects and dislocation structure**

The key to understanding the difference in damage accumulation:

The second and third properties are different in different crystal structures

Basic equations of PBM

PDs (3-D diffusion)

$$\frac{dC_{\mathrm{v}}}{dt} = G_{\mathrm{v}} - \mu_R D_{\mathrm{i}} C_{\mathrm{i}} D_{\mathrm{v}} - D_{\mathrm{v}} C_{\mathrm{v}} k_v^2 ,$$

$$\frac{dC_{\mathrm{i}}}{dt} = G_{\mathrm{i}} - -\mu_R D_{\mathrm{i}} C_{\mathrm{i}} D_{\mathrm{v}} - D_{\mathrm{i}} C_{\mathrm{i}} k_i^2 ,$$

2nd order reaction kinetics

SIA clusters (1-D diffusion)

$$\frac{dC_{\mathrm{cl}}^{\mathrm{m}}}{dt} = G_{\mathrm{cl}}^{\mathrm{m}} - D_{\mathrm{cl}} C_{\mathrm{cl}}^{\mathrm{m}} \left(k_{\mathrm{m}}^{cl}\right)^2 , \quad (\mathrm{m} = 1, 2 \ldots m_{\max}).$$

3rd order reaction kinetics

Sink strengths

$$k_{v,i}^2 = 4\pi R_v N_v + Z_{v,i}\rho \ ,$$

$$\left(k_{to}^{cl}\right)_t^2 = 2\pi R_v^2 N_v \left(\pi R_v^2 N_v + \pi\rho R_\rho / 2\right)^{1/2} \ .$$

.

R_v, N_v – mean radius and density of voids ,

$+Z_{v,i}, \rho$ – dislocation efficiency and density,

R_ρ – dislocation capture efficiency for SIA clusters.

Defect generation rates

$$G_{\mathrm{v}} = G_{NRT}(1 - \varepsilon_r) \ ,$$

$$G_{\mathrm{i}} = G_{NRT}(1 - \varepsilon_r)(1 - \varepsilon_i^g) \ .$$

$$G_{cl}^m = \frac{1}{< x_i^g >} G_{NRT}(1 - \varepsilon_r)\varepsilon_i^g \ .$$

ε_r - fraction of defects recombined in cascades

ε_i^g - fraction of SIAs in glissile clusters

$$G_{\mathrm{v}}, G_i \ \square \ G_{NRT} ,$$

$$G_{\mathrm{v}} \neq G_i \ .$$

Swelling in cubic crystals in the framework of PBM

$$\boxed{\begin{aligned}\frac{dS}{d\phi_{NRT}} = \left(1-\varepsilon_r\right)\left(1-\varepsilon_i^g\right) p_d \frac{4\pi RN Z_v\rho}{\left(4\pi RN + Z_i\rho\right)^2} + \\ \left(1-\varepsilon_r\right)\varepsilon_i^g\left[\frac{4\pi RN}{4\pi RN + Z_v\rho} - \frac{\pi R^2 N}{\pi R^2 N + \pi\rho R_\rho / 2}\right].\end{aligned}}$$

where p_d is the dislocation bias.

Since the first term is much smaller than that of the second one, the fraction of SIAs in SIA glissile clusters, i.e. the product

$$\boxed{\chi = \left(1-\varepsilon_r\right)\varepsilon_i^g ,}$$

is the critical parameter of PBM

Critical Properties and Damage Accumulation in FCC Metals

- **Dumbbell configuration of SIAs is the most stable, diffuse 3-D**
- **SIA clusters have a form of crowdion bundle, diffuse 1-D along close park (110) directions**
- **Edge dislocations are extended due to low stacking fault energy (SFE)**
- **Absorption efficiency of dislocations for PDs is less that that of perfect dislocations**

Reaction kinetics is a mix of 3-D and 1-D thus

$$\boxed{\frac{dS}{d\phi} \approx \chi \left[\frac{4\pi RN}{4\pi RN + Z_v\rho} - \frac{\pi R^2 N}{\pi R^2 N + \pi\rho R_\rho / 2} \right].}$$

$$\Downarrow$$

$$\boxed{\left(\frac{dS}{d\phi}\right)_{\max} \approx \frac{\chi}{2}.}$$

PKA Energy Effect in Copper Irradiated with Different Particles

Irradiations:

- **2.5 MeV electrons (PKA=0.05 keV)**
- **3 MeV protons (1 keV)**
- **fission neutrons (60 keV)**

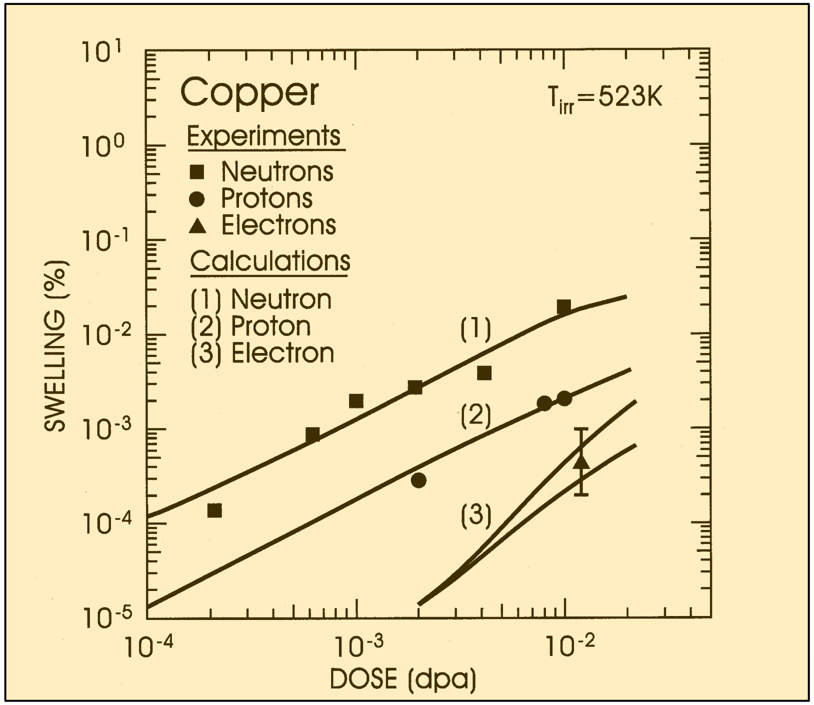
Irradiation conditions:

$G_{NRT} = 10^{-8}$ dpa/sec

T=520 K

in all three cases !

The only experiment with such low generation rates at electron and proton irradiations



Singh et al. *Phil. Mag.* A 80 (2000) 2626.
Golubov et al. . *Phil. Mag.* A 81 (2001) 2533.

50 times higher swelling in case of neutrons compared to electrons at defect production rate 10 times smaller !

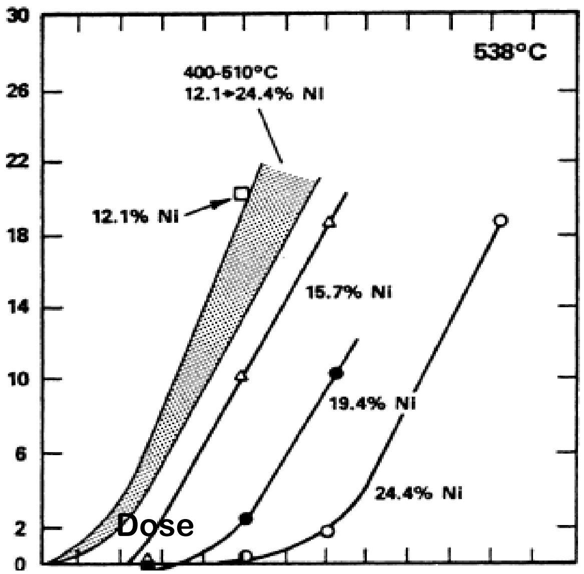
Cascade parameters adjusted for neutron irradiation:

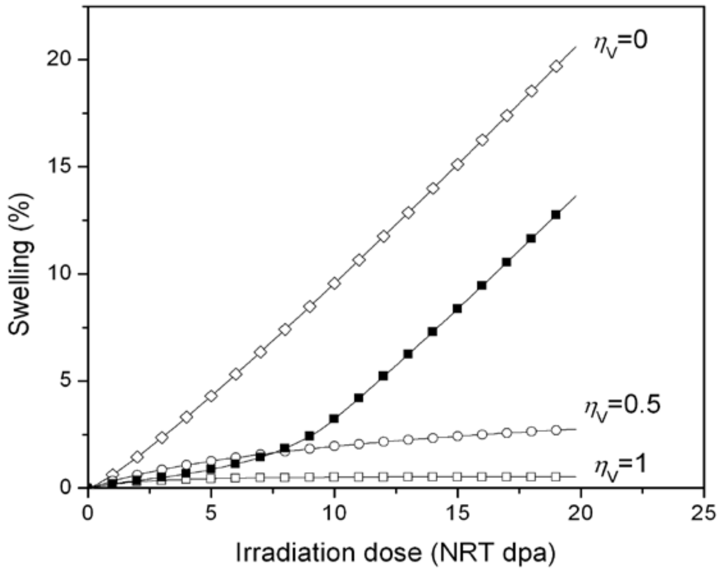
$$(1-\varepsilon_r) = 0.1,$$

$$\varepsilon_i^g = 0.2.$$

$$\longrightarrow \quad \chi = 0.02$$

Steady State Swelling in Neutron Irradiated Stainless Steel





Barashev, Golubov,
Phil. Mag. 89 (2009) 2833.

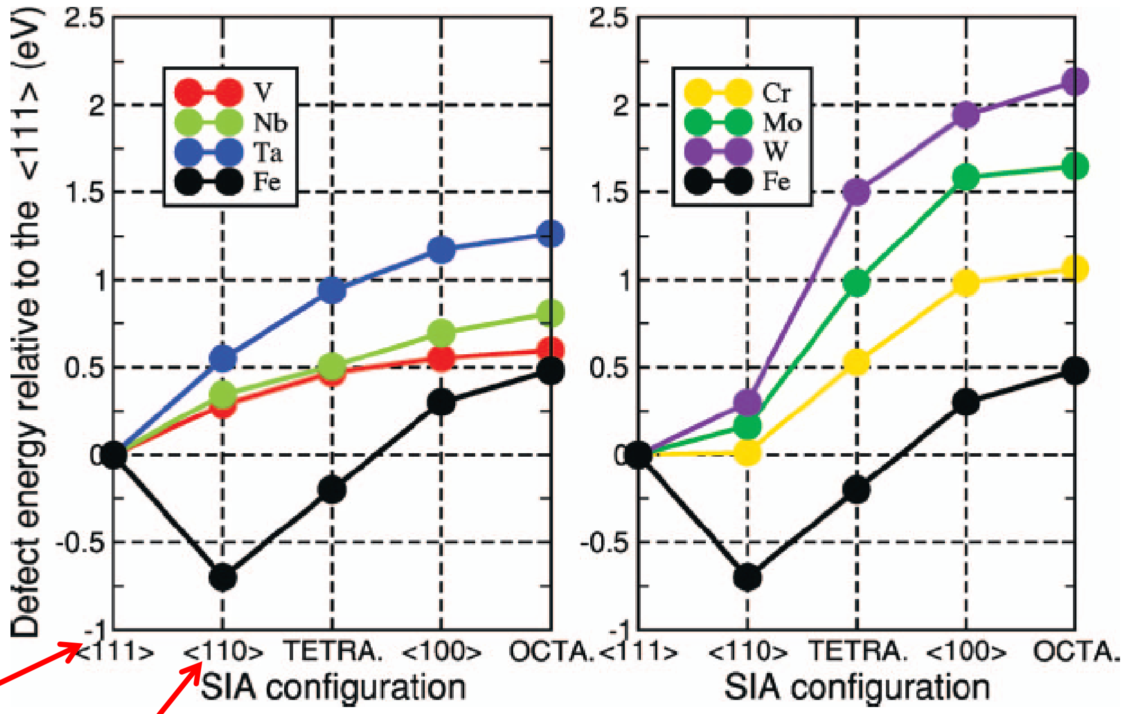
Maximum swelling rate:

$$\frac{dS}{d\phi^{\mathrm{NRT}}} \approx \frac{\chi}{2} \approx 1\% / \mathrm{dpa}$$

Swelling rate of 1%/dpa found in variety of stainless steel is explained for the first time

Damage Accumulation in BCC Crystals

Ab-initio results for SIA stability in BCC metals



crowdion

dumbbell

Nguyen-Manh, Horsfield, Dudarev, Phys. Rev. B 73, 020101R (2006).

Crowdion SIA configuration is the most stable in all metals accept Fe

Critical properties and damage accumulation in BCC metals

- **Crowdion configuration of SIAs is the most stable, diffuse 1-D**
- **SIA clusters have a form of crowdion bundle, diffuse 1-D along close park (111) directions**
- **Edge dislocations are perfect due to high stacking fault energy**

Reaction kinetics is pure 1-D

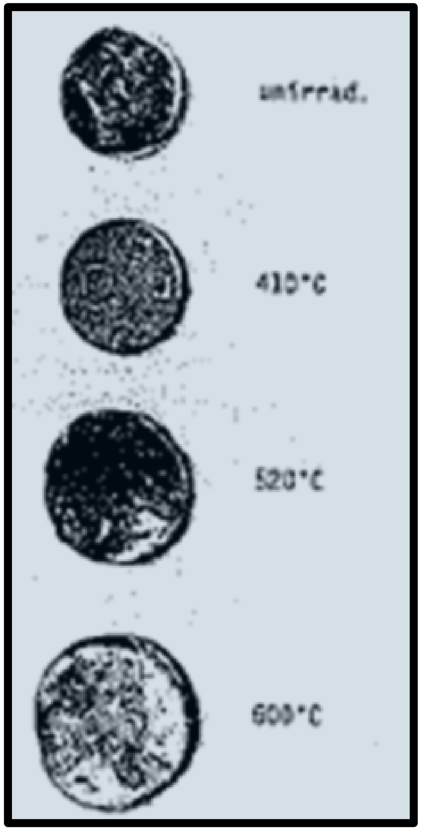
$$\boxed{\frac{dS}{d\phi} = (1-\varepsilon_r)\left[\frac{4\pi RN}{4\pi RN + Z_v\rho} - \frac{\pi R^2 N}{\pi R^2 N + \pi\rho R_d/2}\right].}$$

$$\varepsilon_i^g = 1 \quad \chi = (1-\varepsilon_r) = 0.1$$

$$\boxed{\left(\frac{dS}{d\phi}\right)_{\max} \approx \frac{1}{2}(1-\varepsilon_r) \approx 5\%/dpa.}$$

Potential for damage accumulation is remarkably larger than that of FCC, corresponding to void nucleation and ordering

Swelling in Neutron Irradiated V-5Fe

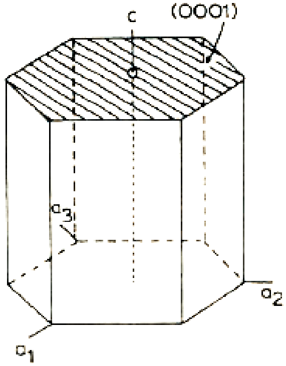


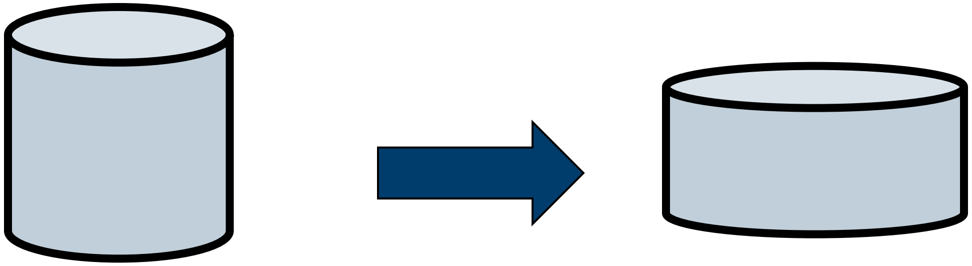
Swelling ~100% at 30 dpa thus swelling rate ~2%/dpa

Maximal swelling rate ever found !

Higher potential for damage accumulation than thait in FCC, correspondingly for void nucleation and ordering

Radiation Growth in Zirconium, I





- ☐ **Expansion in prismatic directions (*a-directions*)**
- ☐ **Contraction along perpendicular to basal ones (*c-direction*)**
- ☐ **Volume conservation**

Extreme case: radiation growth in Zr-7%Pt binary alloy

unirradiated

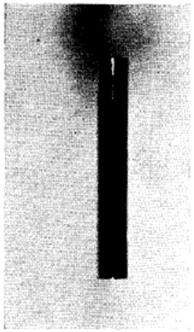


Fig. 1. Typical preirradiation appearance of Zr-Pu alloy specimens. 2 ×.

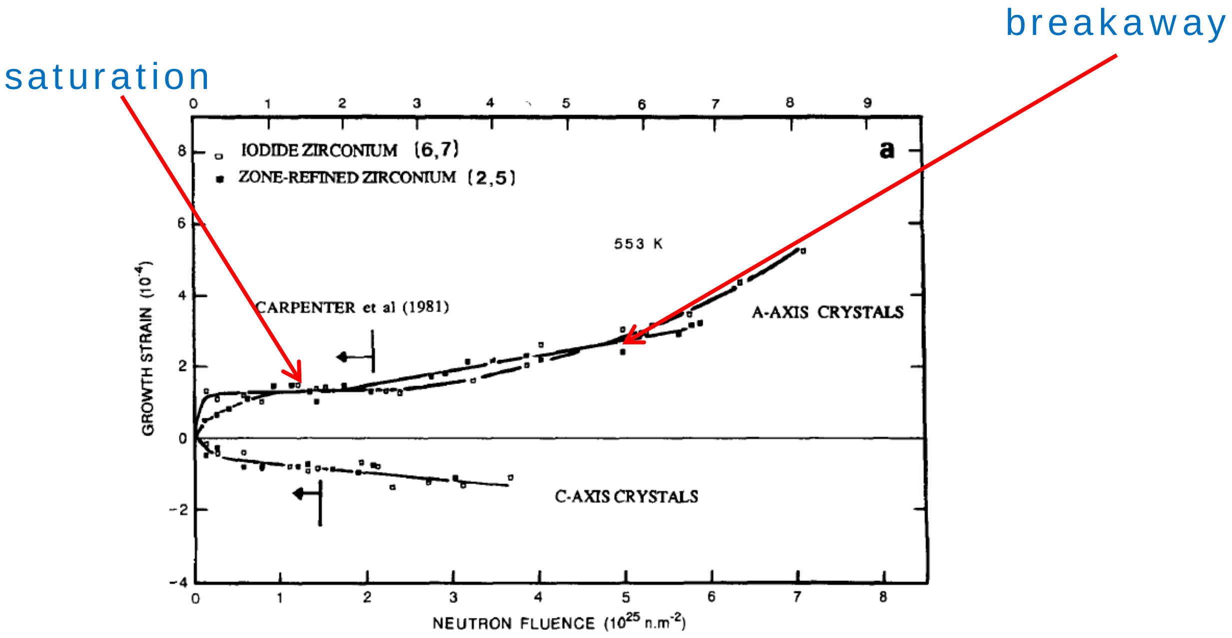
Fig. 2. Typical postirradiation appearance of Zr-5 wt % Pu fuel specimens. 2 ×.

Fig. 3. Postirradiation appearance of Zr-7 wt % Pu fuel specimens. 2 ×.

irradiated

(strain ~100%)

Radiation Growth in Zirconium, II



breakaway

saturation

Observations, which have never been explained:

- High strain rate at small doses followed by strain saturation
- Breakaway growth (why and to where it goes?)
- Coexistence of about the same sizes vacancy and SIA type a-loops
- Negative a-strain

No estimation has been done for the maximal strain rate

Radiation Growth in Zirconium, III

The first radiation growth model was developed at 1962

S.N. Buckley, in: Proc. Int. Conf. on Properties of Reactor Materials and the Effects of Radiation Damage, Ed. D.J. Littler (Butterworths, London, (1962) p. 413.

Recent reviews:

> *'..the basic physical parameters that would be needed to construct reliable mechanistic models to predict the deformation of even a pure Zr single crystal are not known ... We therefore still rely a phenomenological approach.'*

Holt, Journal of Nuclear Materials 372 (2008) 182.

> *' ... understanding of the basic creep mechanisms in anisotropic materials like zirconium alloys is still not strong enough to be truly predictive.' '... Today, most models are empirical in nature, ... '*

Adamson at al., *Advance Nuclear Technology International* (2009).

The phenomenon has not been understood for the last ~ 50 years !

Critical properties and damage accumulation in Zirconium

- ☐ **Several stable configurations of SIAs, diffuse predominant 2-D along basal planes**
- ☐ **C/A ratio is less than ideal**
- ☐ **Edge dislocations of basal and prismatic types are largely different due to difference in their Burgers vectors**
- ☐ **SIA clusters have a form of crowdion bundle, diffuse 1-D along close park directions <u>along basal planes</u>**

Reaction kinetics is mix 2-D/3-D with highly anisotropic 1-D SIA cluster diffusion

Radiation Growth in Framework of PBM

Strain rates in Deckart coordinate system where x is parralel to a_1 prizmatic direction and z is parralel to *c* direction:

$$\frac{d\varepsilon_x}{d\phi} = \chi\left(\frac{1}{2} - \frac{\rho_x}{\rho}\right), \qquad \chi = (1-\varepsilon_{\mathrm{r}})\varepsilon_{\mathrm{i}}^{\mathrm{g}}$$

$$\frac{d\varepsilon_y}{d\phi} = \chi\left(\frac{1}{2} - \frac{\rho_y}{\rho}\right),$$

$$\frac{d\varepsilon_{\mathrm{z}}}{d\phi} = -\chi\frac{\rho_{\mathrm{z}}}{\rho},$$

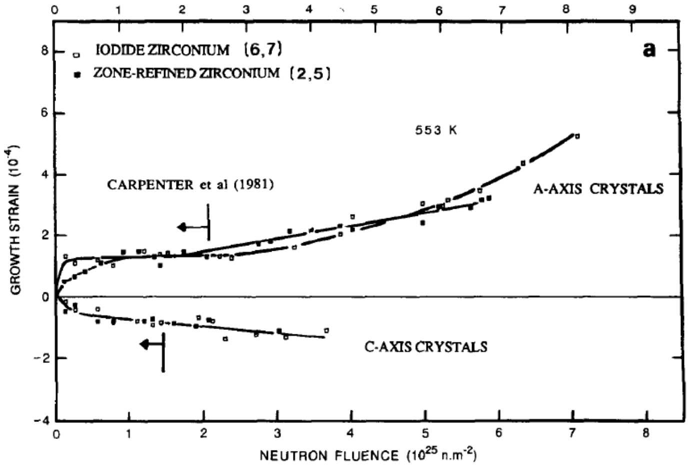
$$\frac{d\varepsilon_x}{d\phi} + \frac{d\varepsilon_{\mathrm{y}}}{d\phi} + \frac{d\varepsilon_{\mathrm{z}}}{d\phi} = 0\,.$$

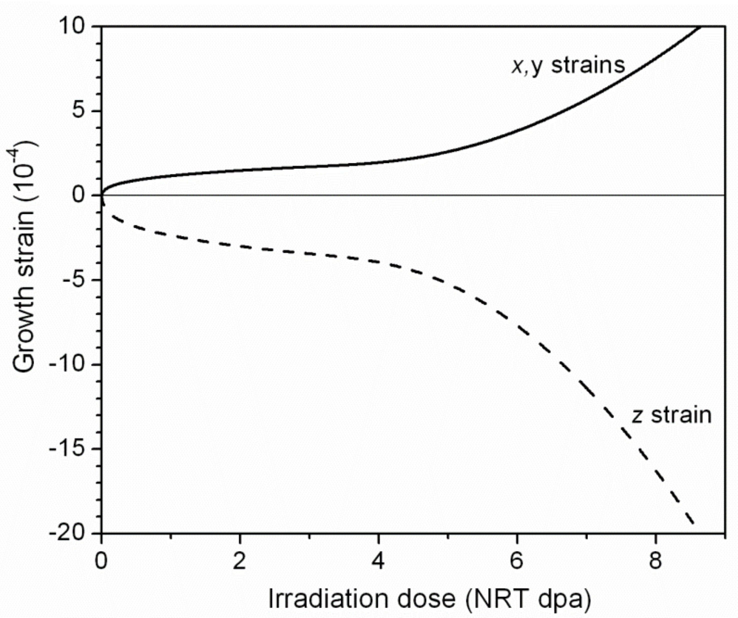
where ρ_x, ρ_y, ρ_z are density of dislocation with Burgers vectors parallel to x, y, and z directions and ρ is total dislocation density

Equations above provide explanations of all striking observations, including estimates of the maximum strain rate

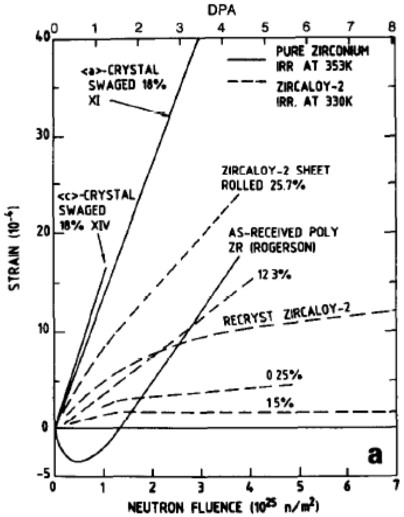
Dose Dependence of RG Strains Predicted by the Model

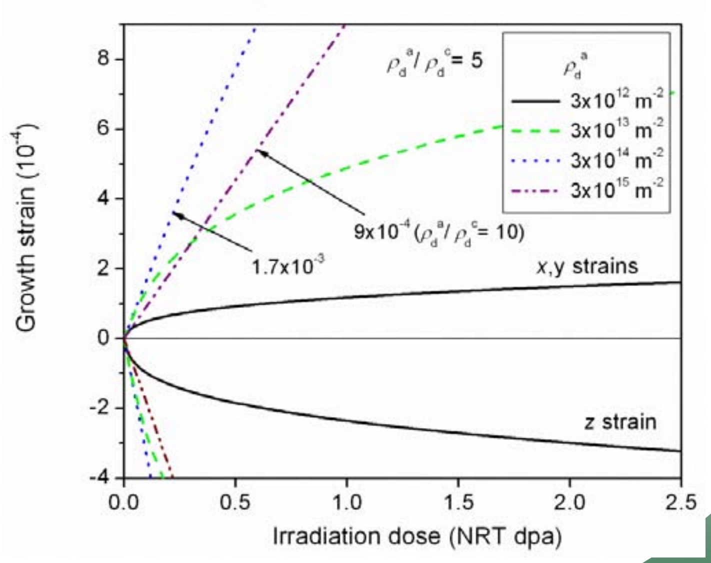
RG in annealed Zr





Impact of cold work





Summary

- **Explanation of radiation growth by PBM originally developed for cubic crystals provides strong validation of the model**

- **For the first time, radiation damage in metals with all three crystal structures are explained in the framework of a single model**

End of presentation,

Thanks for your attention