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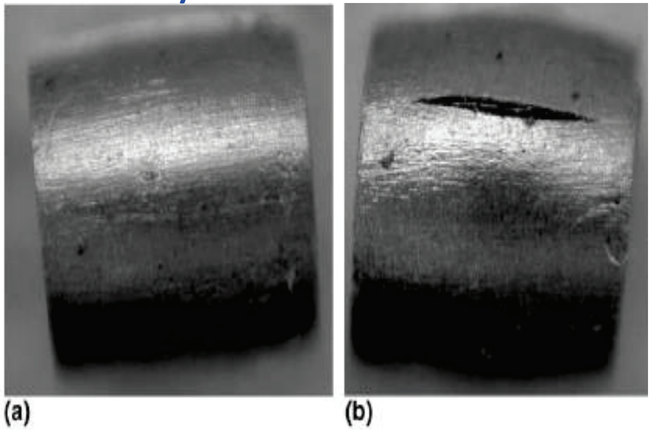
Michael Wohlmuther: Paul Scherrer Institut

Radiation Damage of Components in the Environment of High-Power Proton Accelerators

Cyclotrons 2016, Zürich, Switzerland, 12.9.2016

Radiation damage on materials: Examples

Change of mechanical properties:
Ductility → Embrittlement

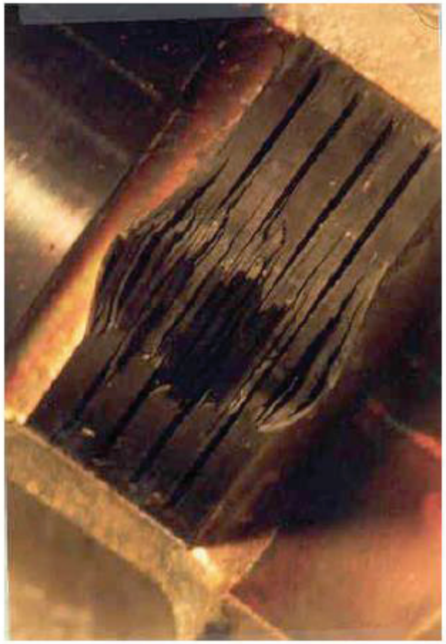


before after irradiation
with 800 MeV protons (23 DPA), LANL

Tungsten after compression test

Maloy et al,
J. Nuc. Mater. 343 (2005), 219

Swelling + Deformation



500 MeV
Protons
at TRIUMF,
150 μA

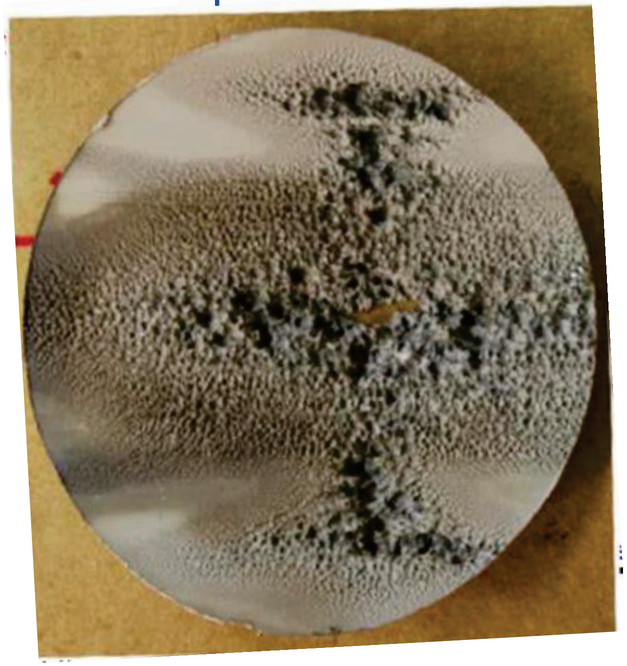
Water-cooled/Edge-cooled
pyrolytic graphite target

E.W. Blackmore *et al.*,
in *Proc. PAC 2005*, 1919

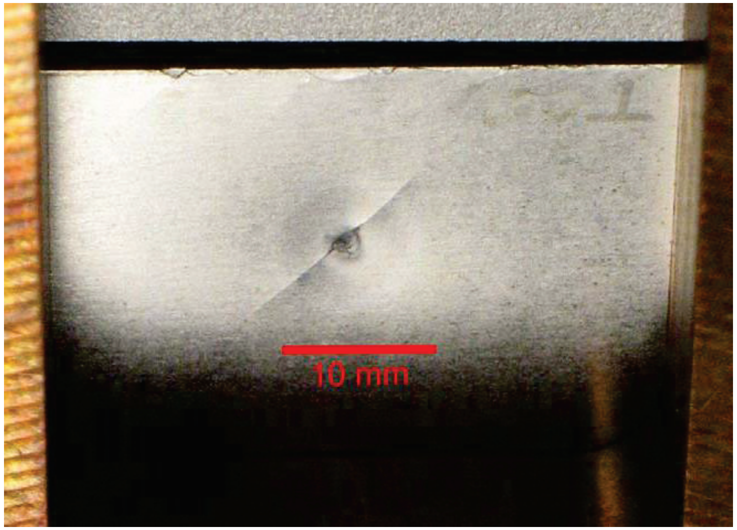
important for high-power beams
on targets, collimators, beam dumps

Radiation damage on materials: Examples

SNS inner target vessel:
Central part



Rare Isotope production target
at NSCL, MSU (FRIB)

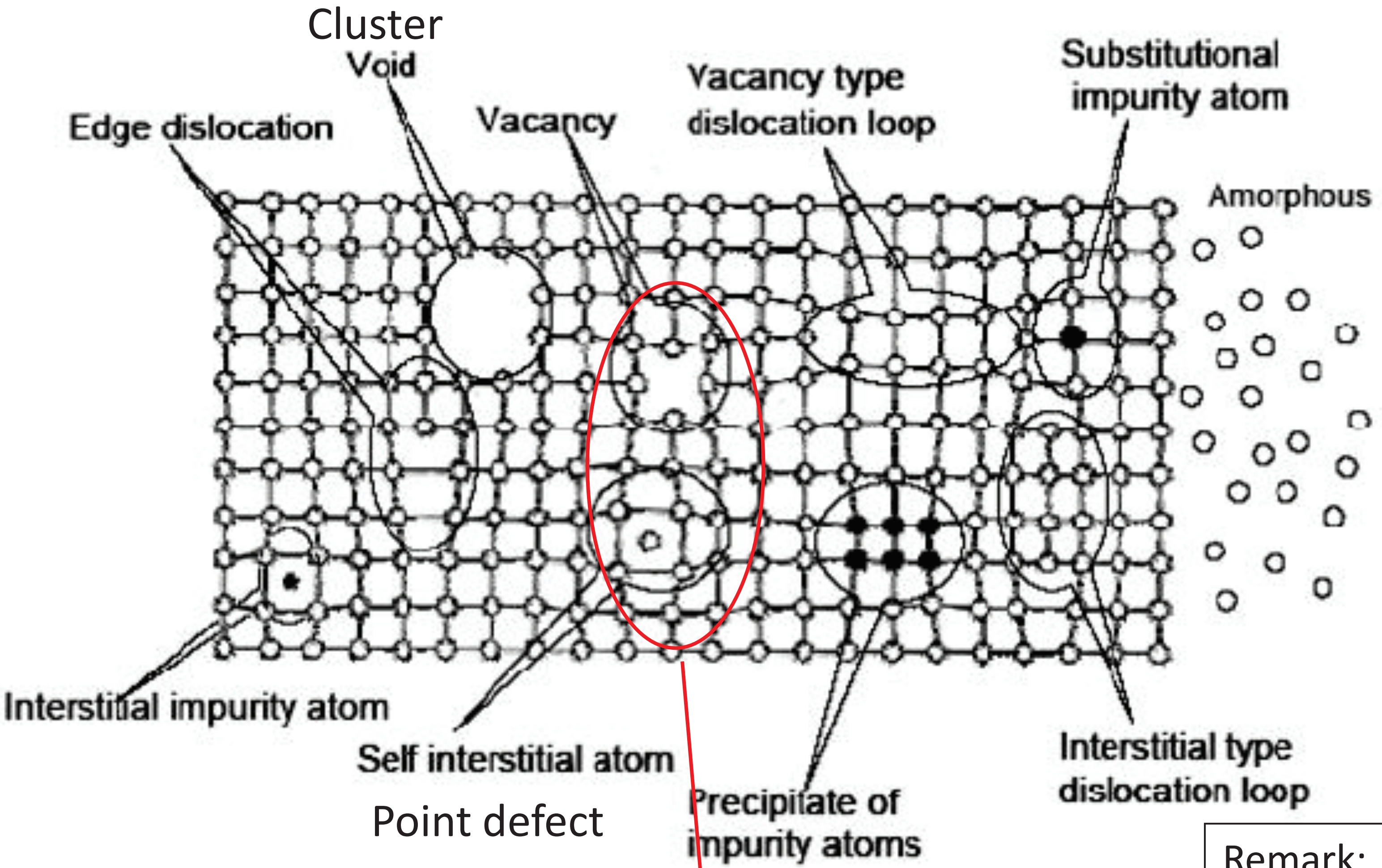


Tungsten: 580 mg/cm2 (0.03 cm)
Beam: $^{76}\text{Ge}^{30+}$ at 130 MeV/u, (3.2 DPA)

Stainless steel in contact with mercury:
Structure does not follow beam profile
but regions of high tension
→ cavitation, pitting
Reason: mainly thermal shock due to the beam
B. Riemer et al, JNM 450 (2014) 183–191

Reason:
swelling & embrittlement
enhanced thermal stress → crack
R. Ronnigen, HB2010

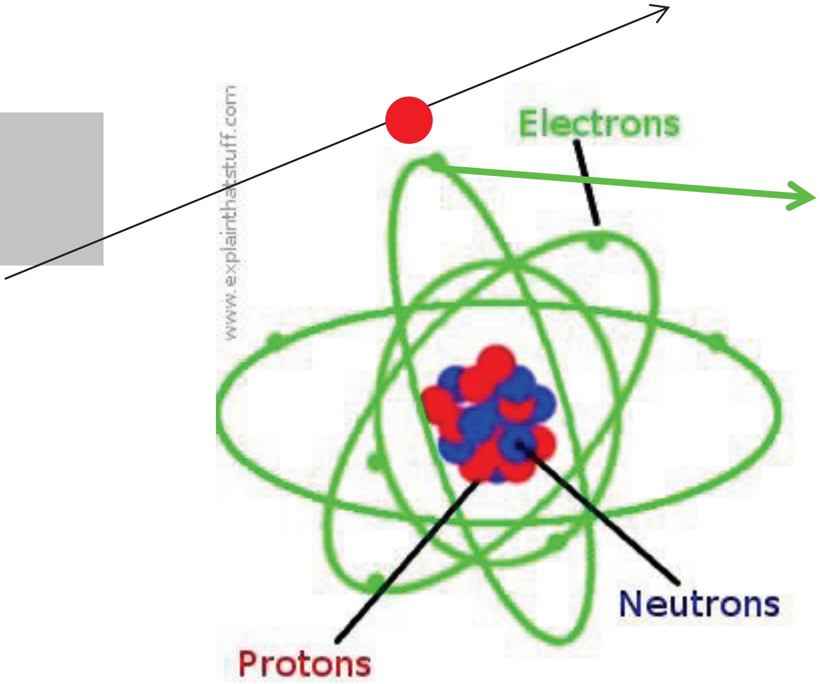
Microscopic picture of radiation damage in structural material



vacancy + self-interstitial atom = Frenkel pair.

Remark:
Liquids do not suffer radiation damage

1. Passage of charged particles through material



1) Ionization & excitation:

→ Dissipation of heat (cooling!)

→ No damage except for

- electronics:
  charge build-up, threshold shift
  failure of transistors etc.
- insulators:
  can become conductive
- organic materials: destroying of electronic bonds
  plastics, grease gets brown and brittle

Physical quantity: ionizing dose (= absorbed dose in material) in Gy

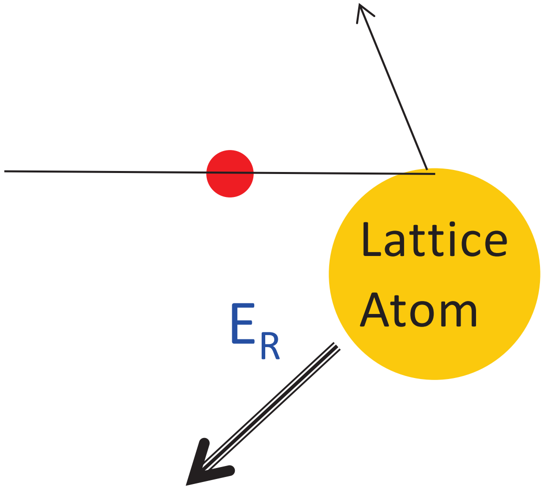
→ cumulative effect over time

→ calculation is well known

2. Passage of particles through material

2) Elastic scattering:

Transferring recoil energy E_R to a lattice atom



To displace an atom: bonds need to be broken

→ E_R > displacement energy E_D

range: 10 - 60 eV Cu: 30 eV, Fe, Ni, Co: 40 eV

~ twice the sublimation energy

→ production of vacancies, interstitials

1. recoil atom = primary knocked-out atom (PKA)

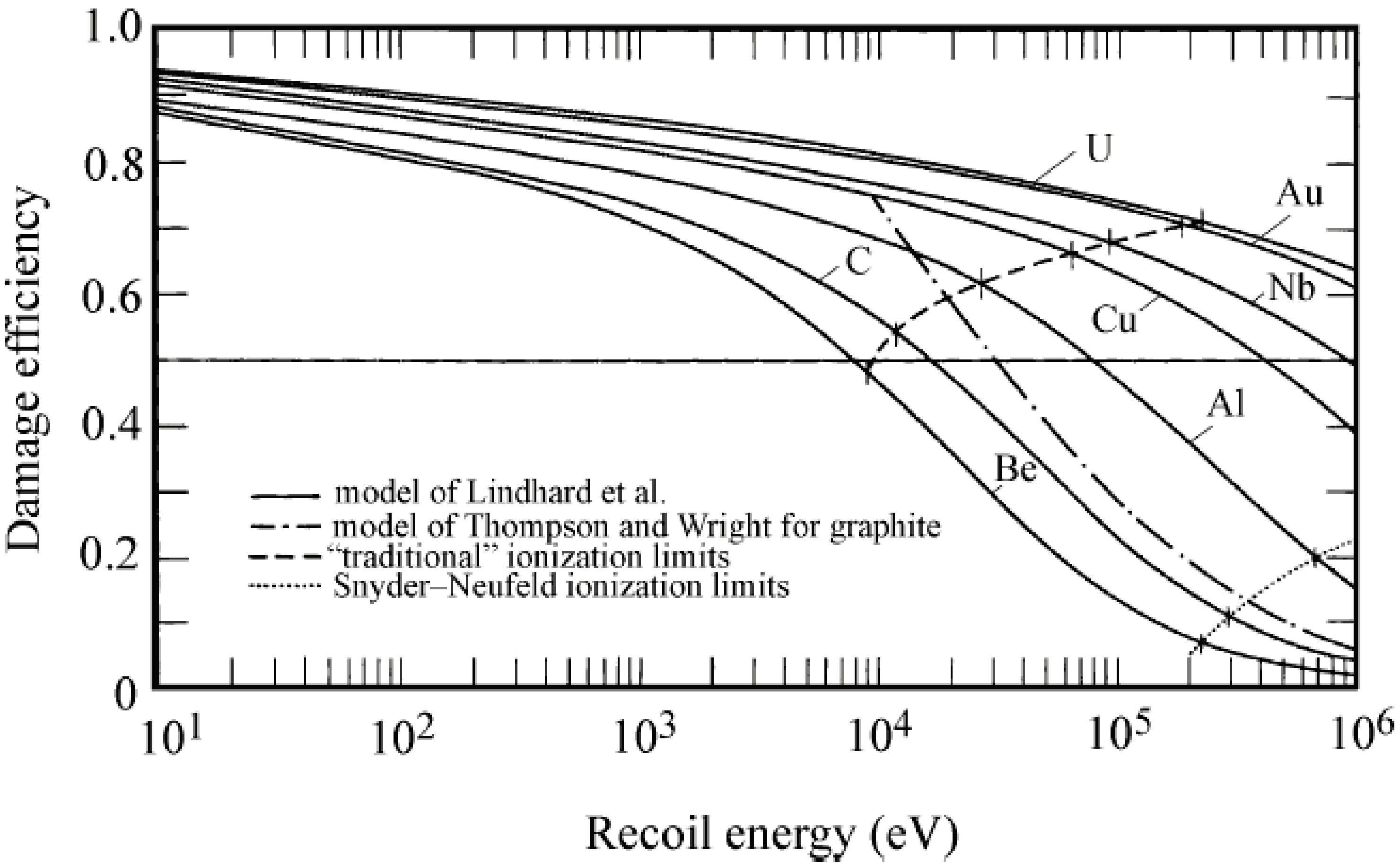
Recoiling nucleus is ionized and loses energy E_R due to

- ionization/excitation E_e
- nuclear reactions: $T_{dam} = E_R - E_e = \xi(E_R)\, E_R$

damage energy

partition function, damage efficiency

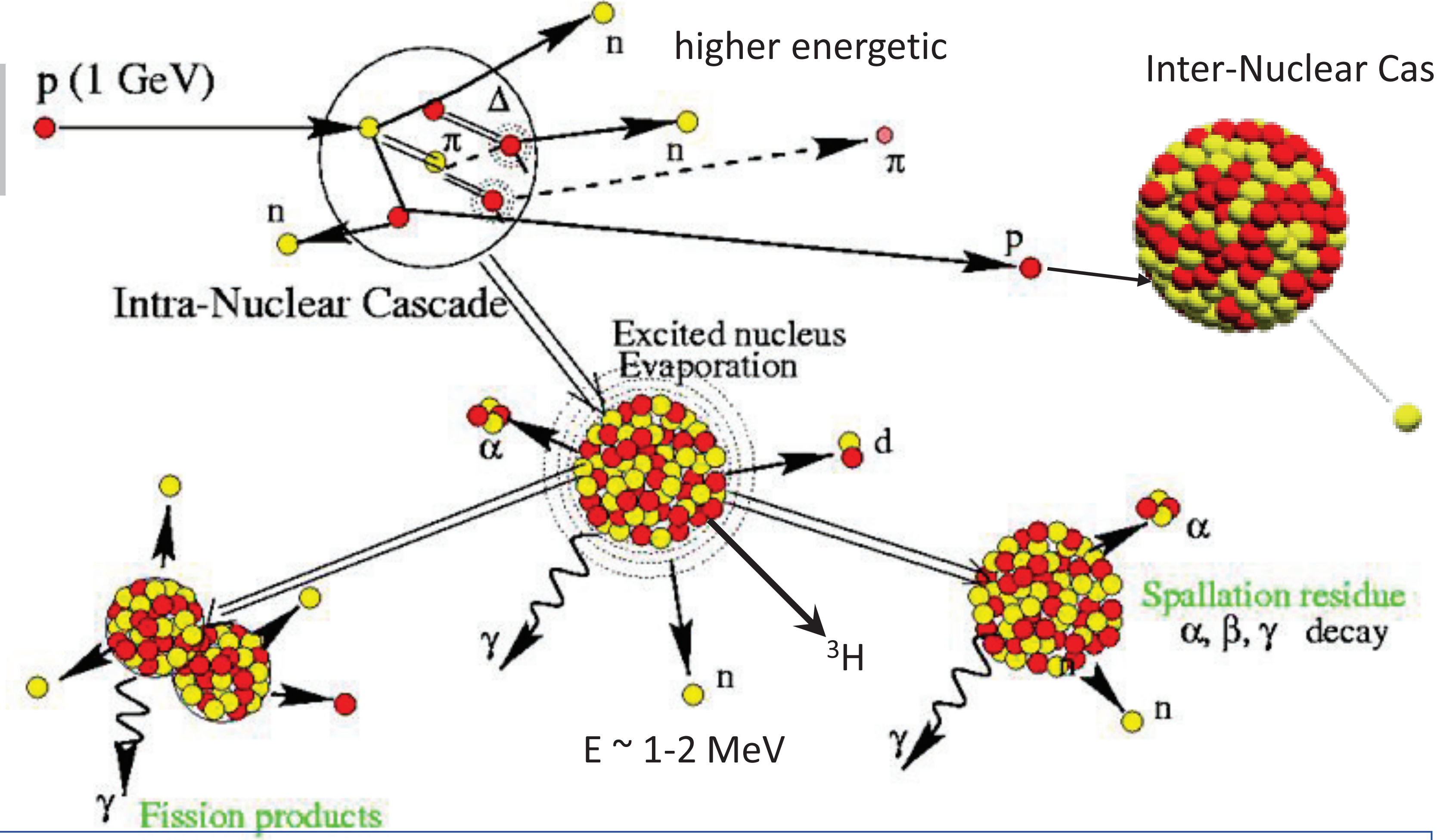
ξ(T): partition function, damage efficiency



- Lighter ions loose much more energy by ionization than heavy ions
- at E_R > 10-100 keV more energy is lost by ionization

→ less damage

Inelastic nuclear reaction



Hadronic cascade

higher energetic

Inter-Nuclear Cascade

^{3}H

E ~ 1-2 MeV

Complicated coupled processes due to many particles involved
→Monte Carlo particle transport simulation
like FLUKA, MARS, MCNP(X), PHITS

Number of produced defects

$T_{dam} = E_R - E_e = \xi(E_R)\ E_R$

ν: Number of produced defects

a) $T_{dam} < E_D$: $\nu = 0$

b) $E_D < T_{dam} < 2\ E_D$: $\nu = 1$

1 atom is displaced to an interstitial site

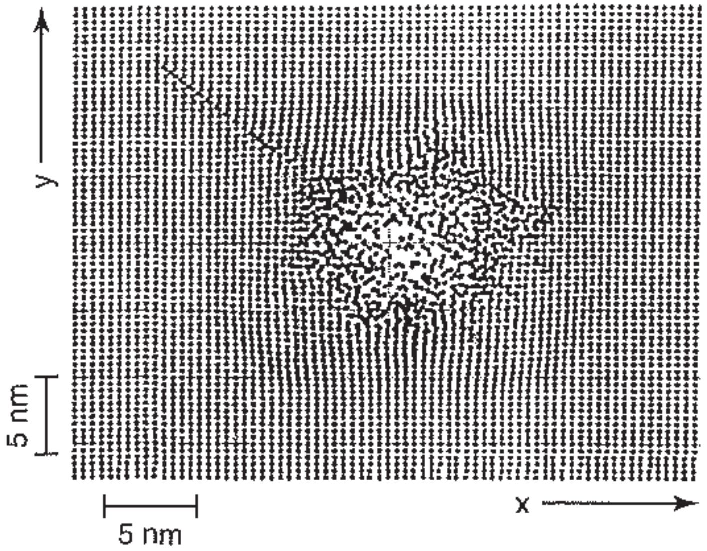
→ a vacant lattice site is created

c) $T_{dam} > 2E_D$: $\nu(E_R) = \dfrac{\kappa T_{dam}(E_R)}{2E_D}$ $\kappa = 0.8$

cascade of collisions within a small range

→ displacement spike

modified Kinchin-Pease m.
= NRT model:
Norgett, Robinson, Torrens
Nucl.Eng.Des. 33 (1975) 50



Later, most of the defects heal out

∅ ~ 10 nm after 1ps

for E_R = 10 keV on PKA

Simulation (Molecular dynamics)

H. Ullmeier, MRS Bulletin 22, p. 14, 1997

Displacement cross section (dcs)

$$\sigma_{dis}(E) = \int_{E_D}^{E\max} \frac{d\sigma_{dam}(E,E_R)}{dE_R}\, \nu(E_R)\ \mathrm{d}E_R$$

particle energy

damage cross section:

$$= \frac{w(E,E_R)}{xN_V}$$

damage function (no. of displaced atoms):

$$= \frac{\kappa T_{dam}}{2E_D}$$

modified Kinchin-Pease m.
= NRT model:
Norgett, Robinson, Torrens
Nucl.Eng.Des. 33 (1975) 50

$w(E_R)$: recoil spectrum
needs nuclear reaction models

x: thickness of the sample (thin)

N_V: atomic density (atoms/cm^3)

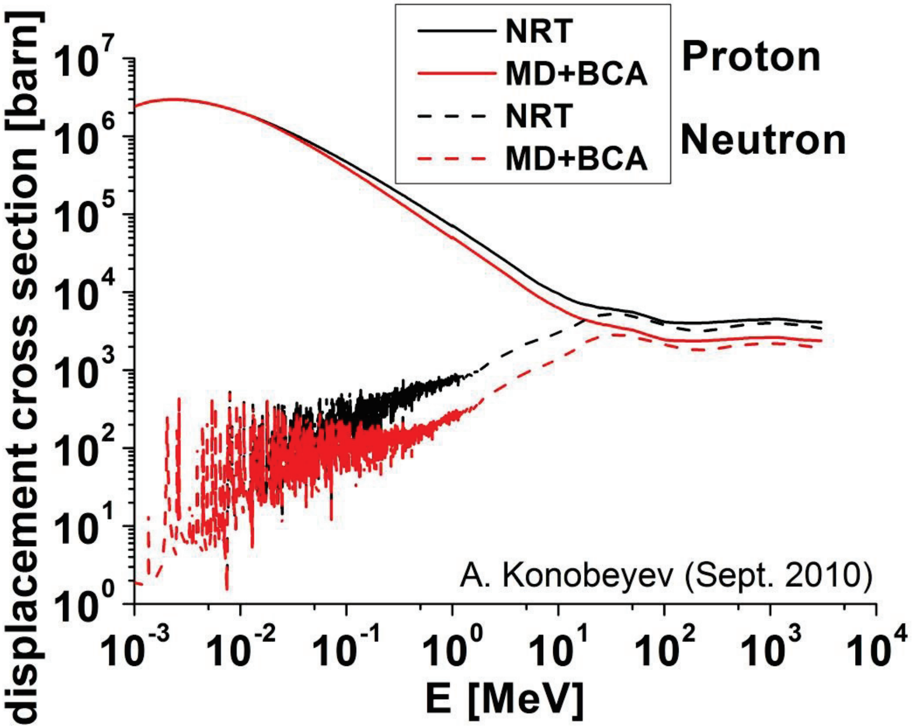
$T_{dam}(E_R)$: damage energy

displacement efficiency $\kappa = 0.8$

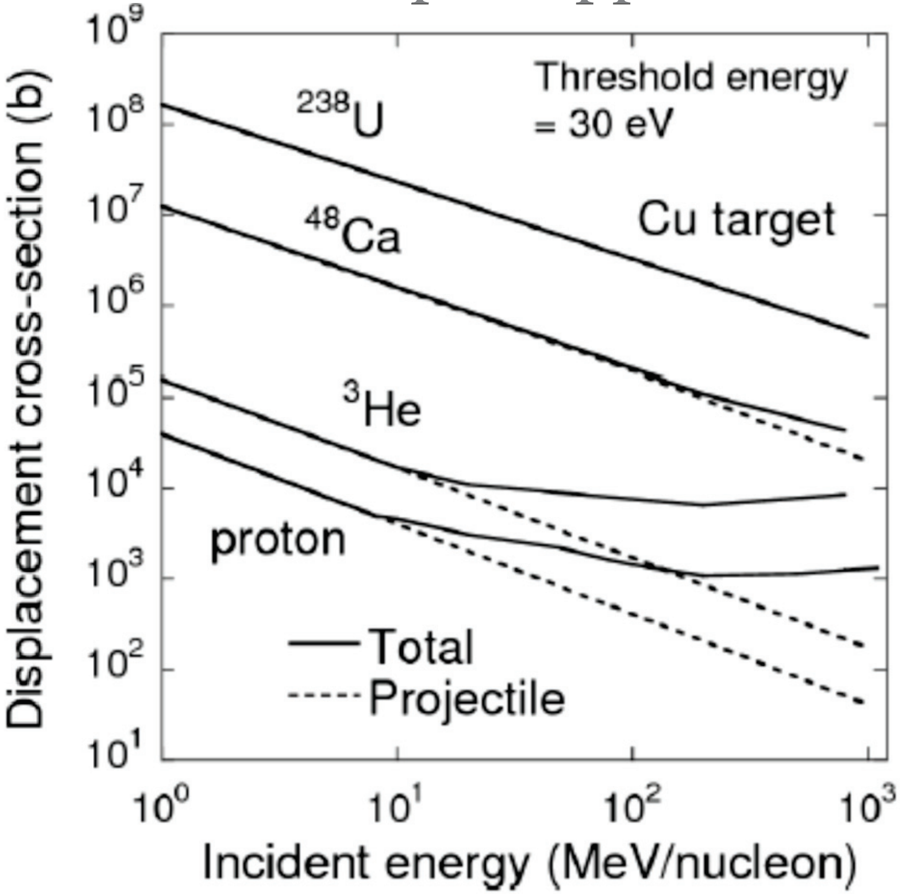
some remarks on uncertainties:

- E_D e.g. in Cu set to 30 eV but varies 18 – 43 eV
- $\kappa = 0.8$: correction derived from BCA simulation of Robinson, Torrens 1972 for common materials like Cu, Fe, Au, W

Displacement cross sections: Example copper



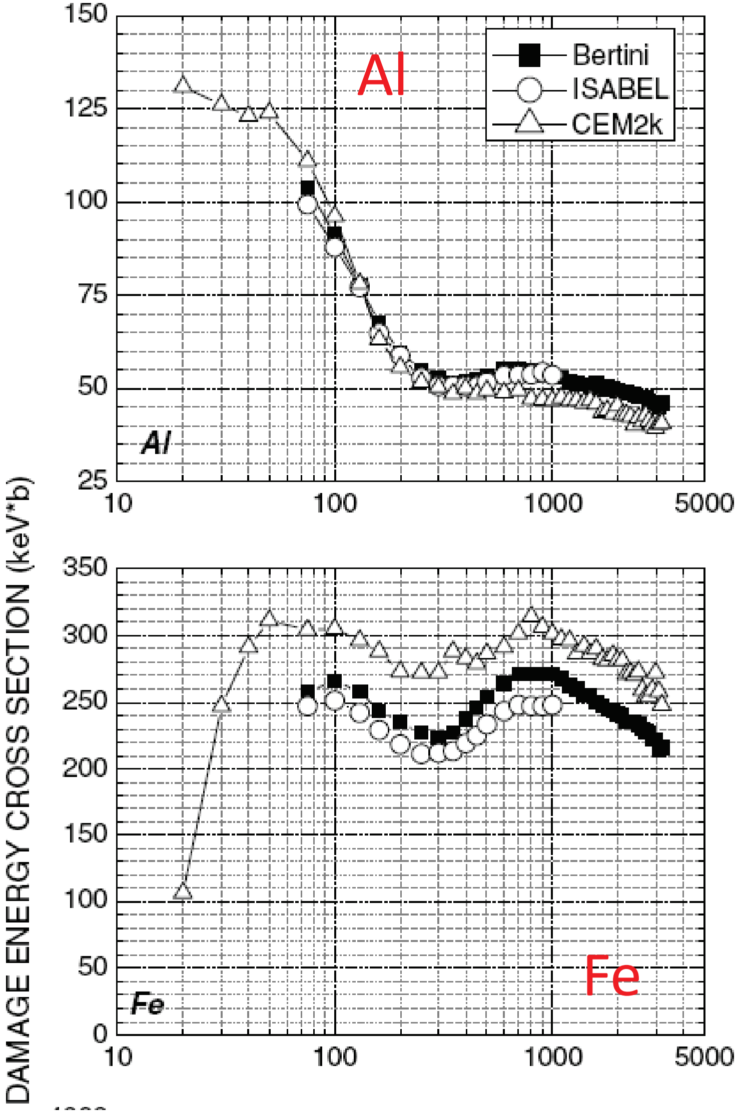
A. Konobeyev (Sept. 2010)



- Charged particles have large c.s. at low energy due to Coulomb interaction
- much more damage for ions compared to proton
- for heavier ions damage comes from primary particle
- due to shorter range of ions: → damage due to ions is very localized

For protons & neutrons: > 10 MeV similar c.s. due to nuclear interaction
main effect due to secondaries

Example: Damage cross section for Al, Fe,W

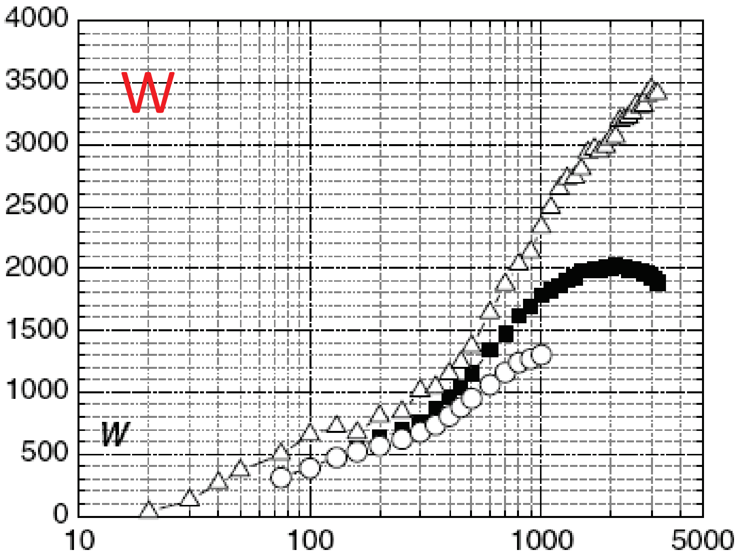


projectile energy [MeV]

integrated over recoil energy:

$$= \int \sigma_{dam}(E,T)dT$$

energy dependence and amplitude varies largely for different material



projectile energy [MeV]

Lu et al., J.Nuc.Mat.356(2006),280

Displacements per atom

Displacements Per Atom (DPA):

- how often an atom is displaced during the irradiation period

$$DPA = \int \sigma_{disp}(E) \; \frac{d\phi(E)}{dE} \; dE$$

$\phi(E)$: fluence (particles/cm^2)

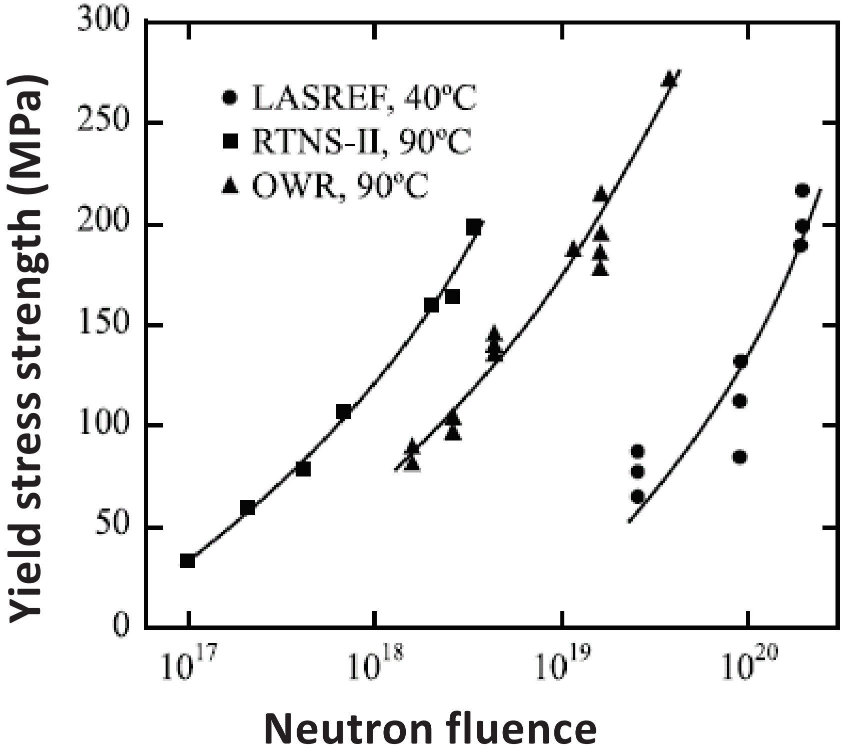
σ_{disp}: displacement cross section

Some remarks:

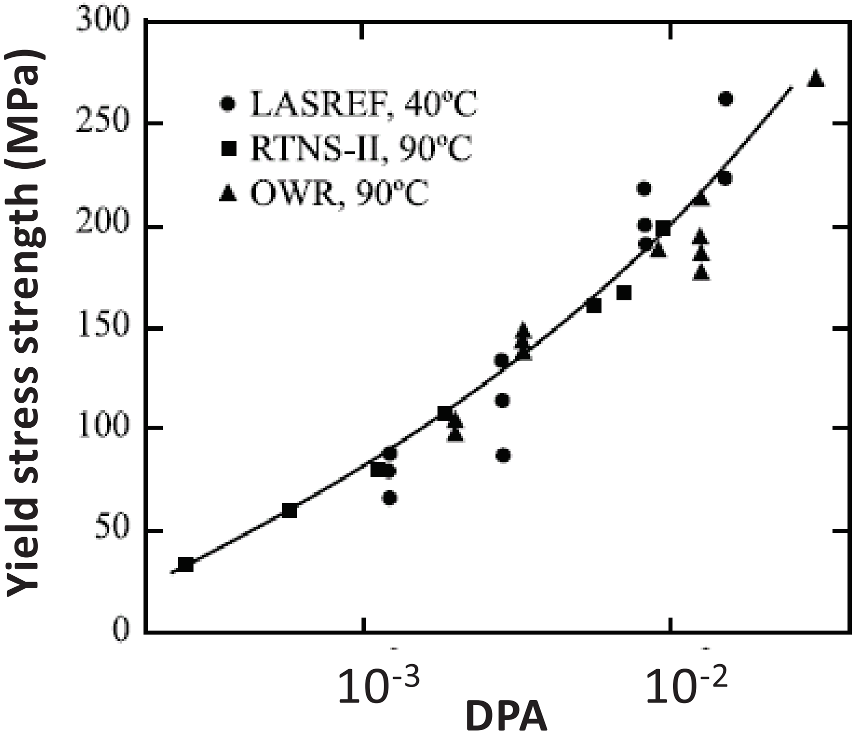
- DPA is purely derived from the energy transferred to the lattice using interaction cross sections
- No information about the number of (stable) defects
- No information about the damage of material properties
- DPA cannot be directly measured as most of the defects heal out after ~ 10 ps
- It is used as a measure of radiation damage, just for quantification.

DPA scaling

Example: 316L stainless steel



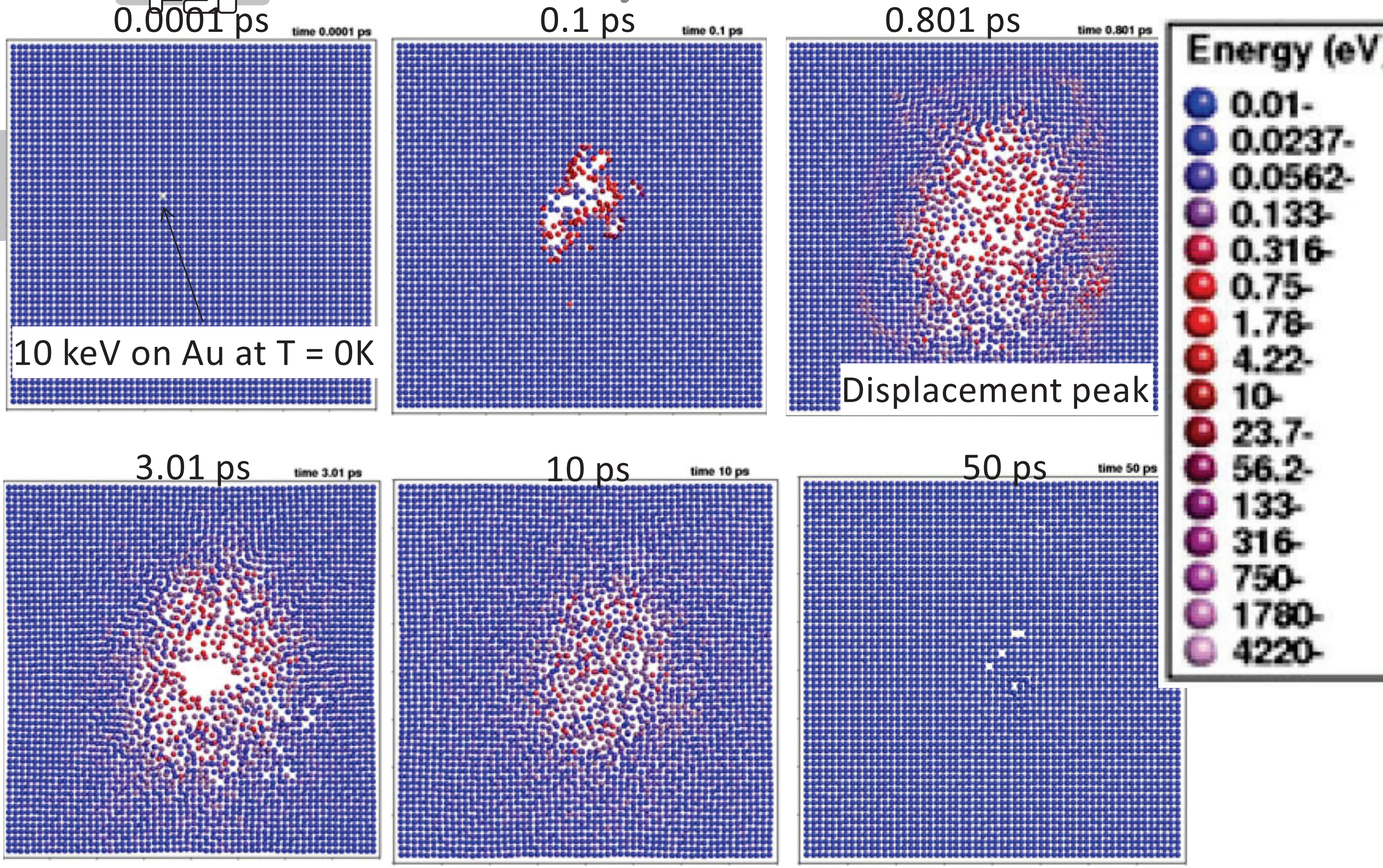
Greenwood, J. Nuc. Mater. 216 (1994), 29



LASREF: neutrons from 800 MeV p on W, broad spectrum, peak at ~ 1 MeV
RTNS-II: 14 MeV n-source
OWR: thermal n-spectra

DPA scaling works for same kind of particles under similar conditions (temperature), when other effects like helium production does not play a major role (here: very low DPA).

Molecular dynamics simulation

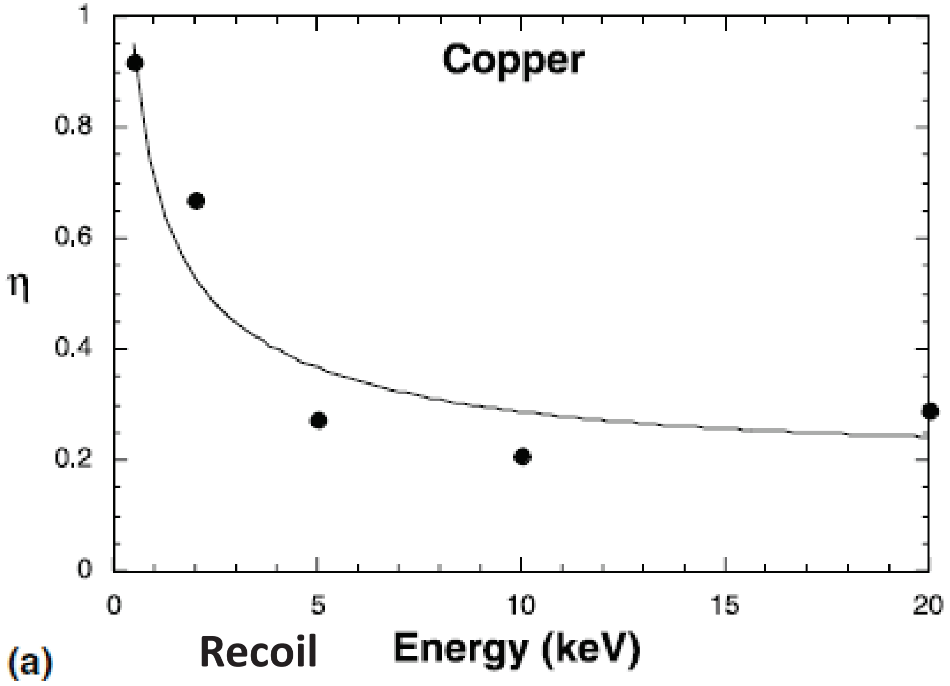


Nuclear Science, NEA/NSC/DOC(2015)9

Healing of defects

2 Effects:

1) athermal: for large E_R, < 50 ps
  - independent of temperature but dependent on E_R
  - large energy stored in displacement spike (~10000 K),
  - atoms are displaced several times until most of them reach a stable position during «cool down»



Cartula et al.,
J. Nucl. Mat. 296 (2001) 90

MD calculations

Defect production efficiency η

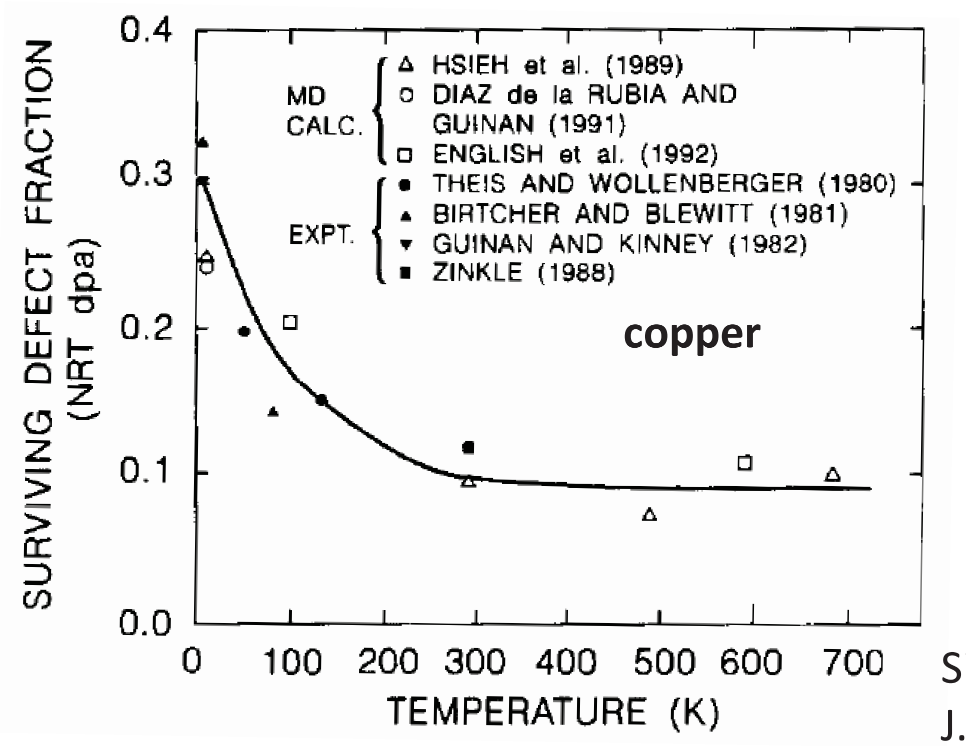
$$\eta = \frac{\text{stable defects}}{\text{displacements}}$$

Healing of defects

2) thermal: annealing, long-time scale

- Defects (atoms) get mobile at elevated temperatures (> 10 K)

→ Leading to clustering, precipitation, segregation at grain boundaries but also recombination, i.e. healing of defects



Correction to the damage function:

$$\nu(E_R) = \eta(E_R, T)\,\frac{\kappa T_{dam}}{2E_D}$$

Defect production efficiency η

important when comparing damage produced by low- and high-energy particles and at different temperatures.

Singh, Zinkle et al.,
J. Nucl. Mat. 206 (1993) 212

Measuring of defects

DPA cannot be measured since healing starts within picoseconds.

1) Increase of Electric resistivity:

Frenkel pairs N_{FP} =
vacancy + self-interstitial atom

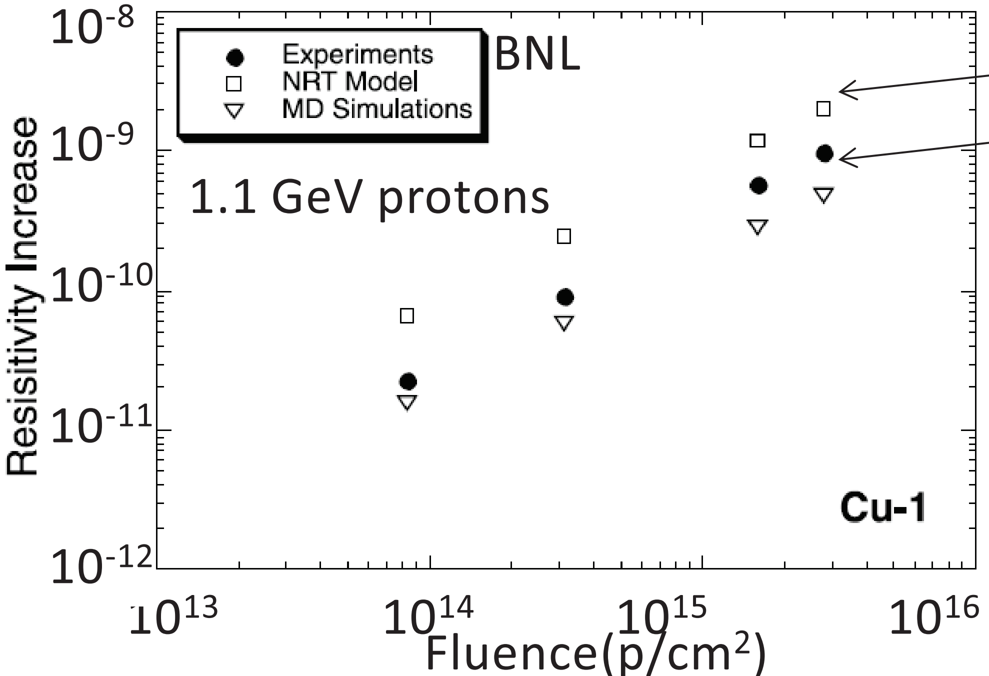
Resistivity per Frenkel pair ρ_{FP} known

Measuring change in resistivity with irradiation dose:

$$\Delta\rho = N_{FP}\rho_{FP} = \eta(E_R, T)DPA\rho_{FP}$$

No other defects like clusters, loops should be present → exp. at small DPA

However, in pure metals ρ of other defects is similar to ρ_{FP}



NRT

Experiment

Defect production efficiency η

$$\eta = \frac{\rho(\exp)}{\rho(NRT)}$$

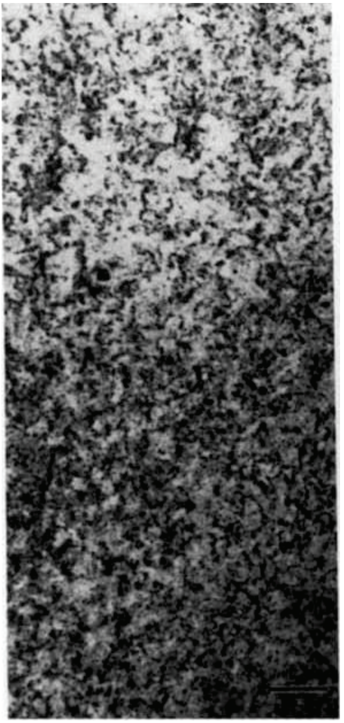
Cartula et al.,
J. Nucl. Mat. 296 (2001) 90

Visualizing of defects

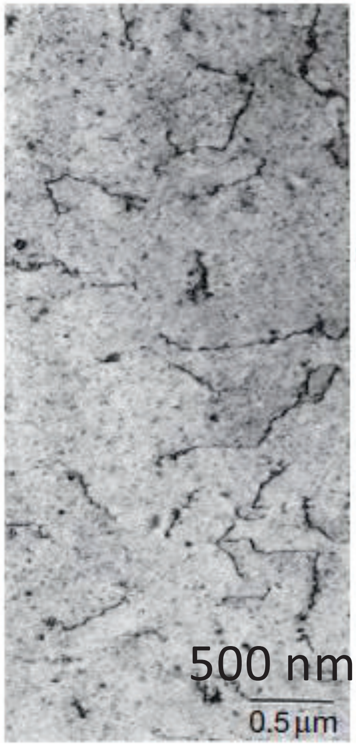
2) Transmission electron microscope (TEM):

thin samples required, careful preparation required

750 MeV protons on Cu



0.4 DPA, 60°C



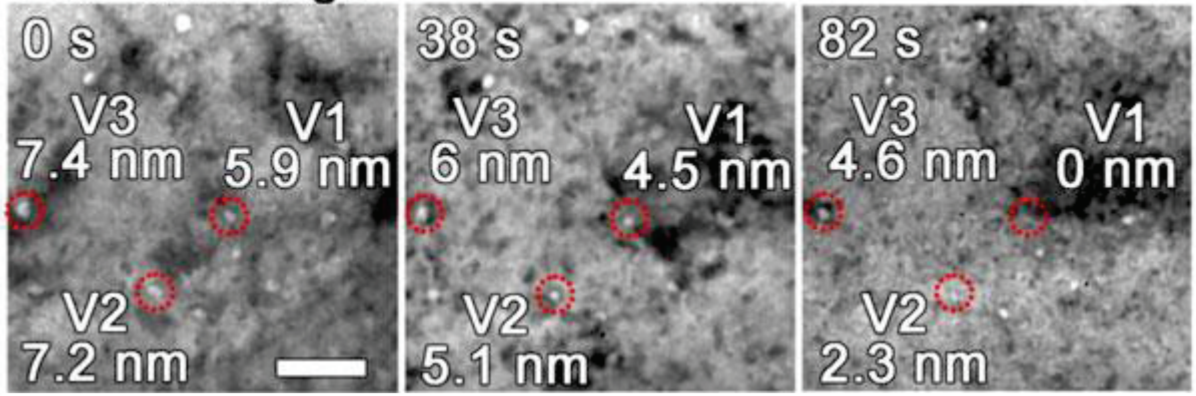
500 nm

2 DPA, 200°C

In situ high-resolution TEM:

void shrinkage of nanovoids

Kr ions on Cu



50 nm

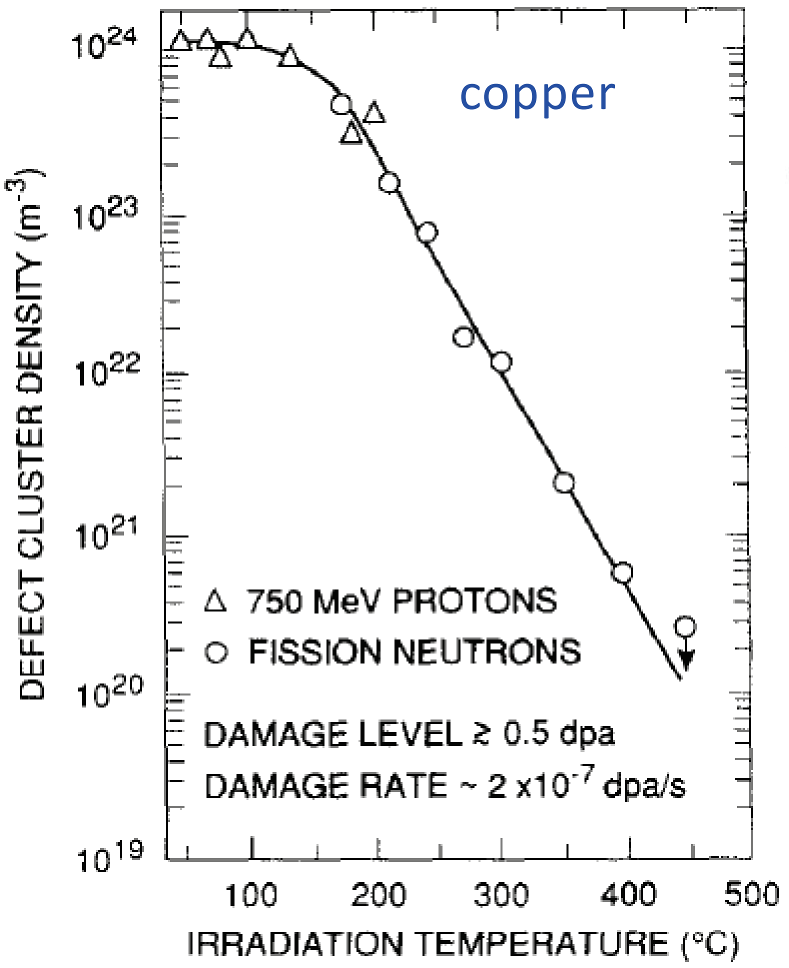
Chen et al, Nature Comm., 2015.

Defects on the photo are counted.

Only defects above resolution are visible.

Zinkle et al, J. Nuc. Mater. 212 (1994) 132

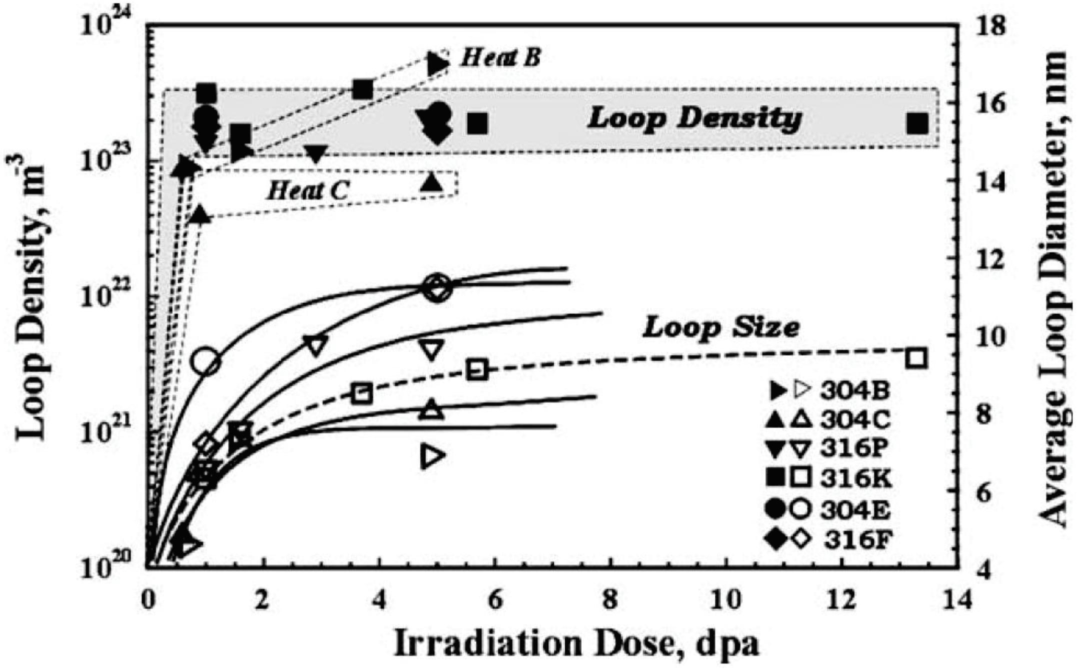
Decreasing and saturation of defect densities



copper

Zinkle et al, J. Nuc. Mater. 212 (1994) 132

stainless steel



Edwards et al., J. Nuc. Mater. 317 (2003) 13

Cluster density decreases,
however, single defects increases

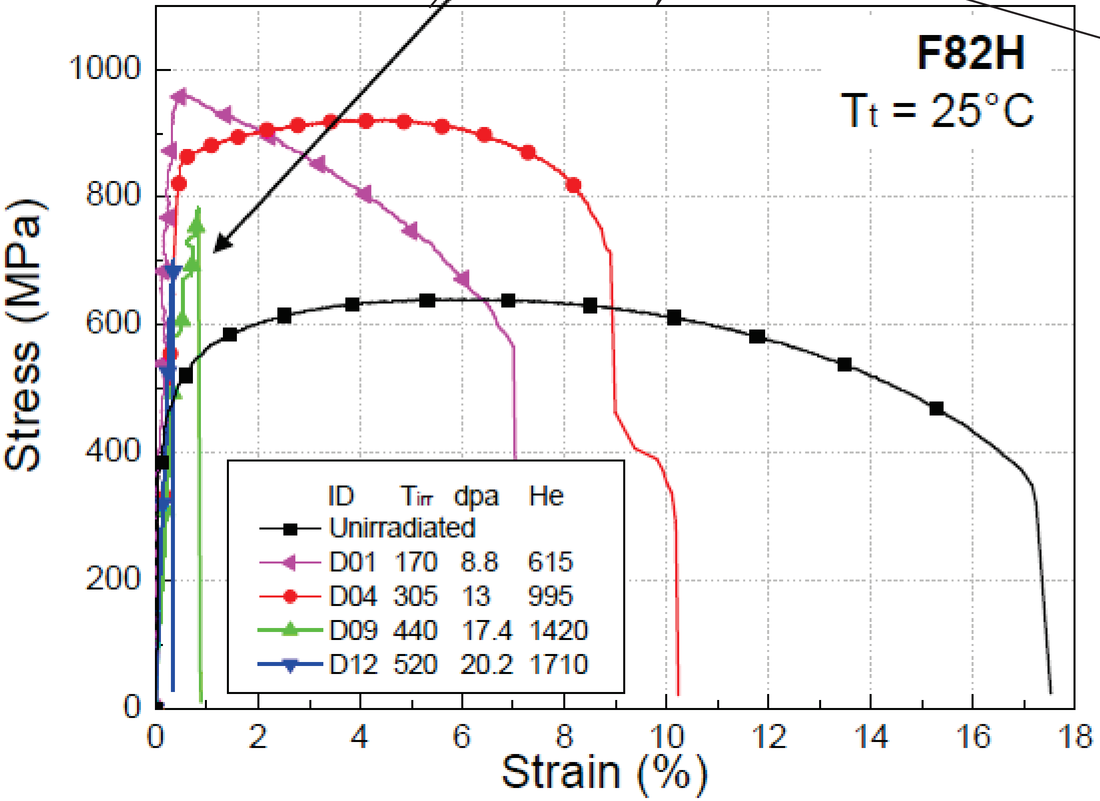
Tension Test: ferritic/martensitic (FM) steel

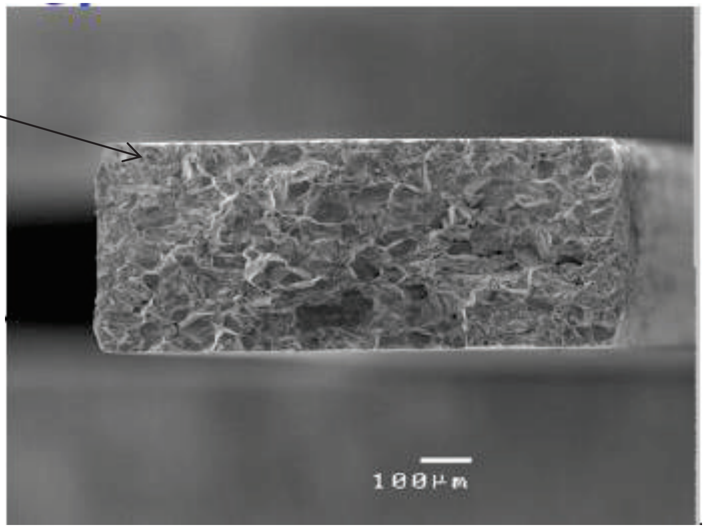
- Higher strength for > 500 °C
- Superior resistance to irradiation inducing creep and swelling
- Candidate materials for high irradiation areas/ Gen. IV reactors

Test in SINQ-neutron spallation source at PSI

Very brittle

> 440 °C, > 17 DPA





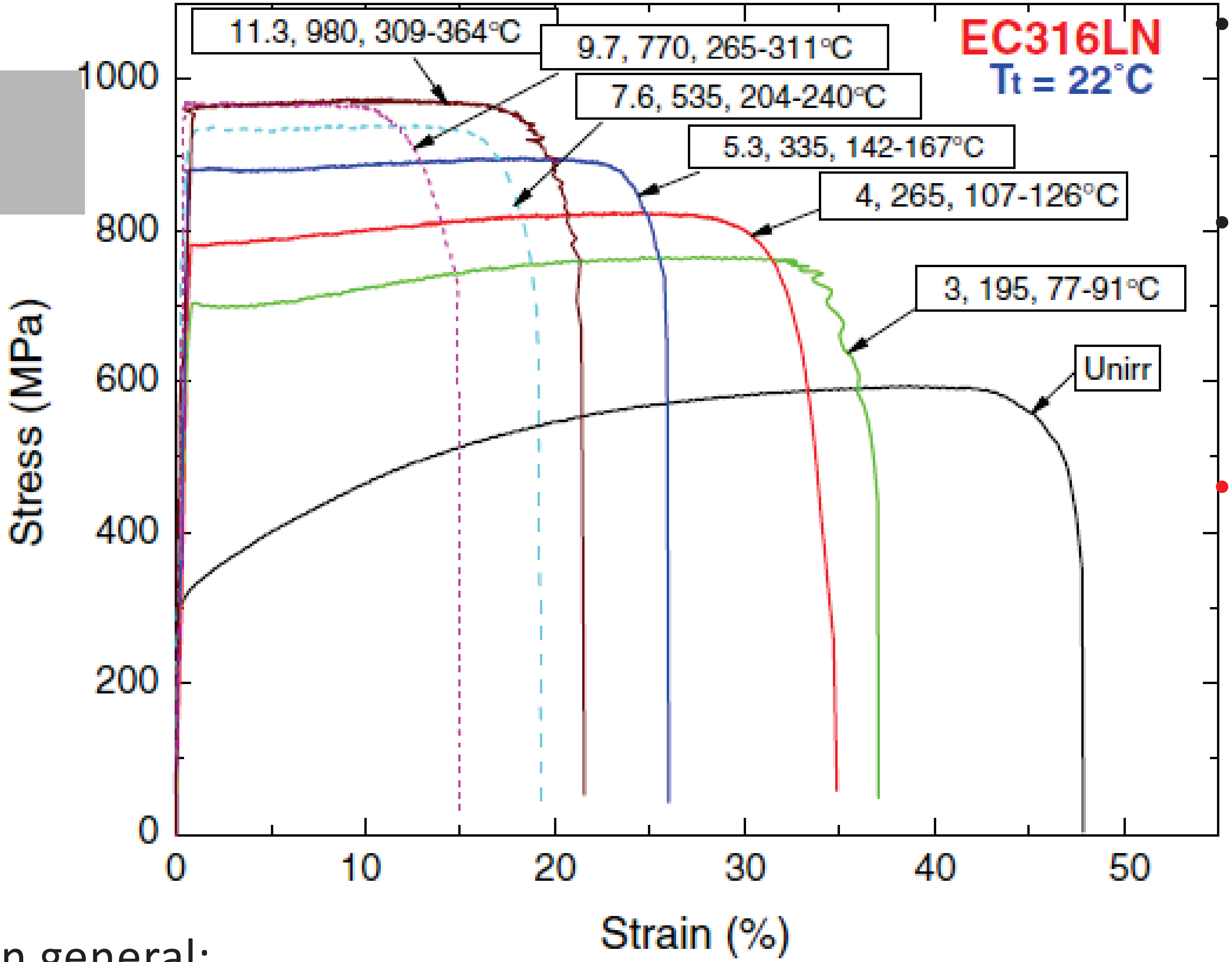
Break without necking

→ Very brittle

Henry et al, J. Nuc. Mater. 377 (2008) 80

Tension Tests: austenitic steel

DPA. He appm, T



- Still ductile fracture (up to 11 DPA, 350 °C)
- more resistant against embrittlement at lower temperature
- at high temperature embrittlement induced by very low He content (> 1 appm

Y. Dai et al, J. Nuc. Mater. 377 (2008) 109

in general:

Irradiation leads to an increase of tensile strength but to a loss of ductility, i.e. the material gets brittle

H/He production in reactors and accelerators

Irradiation facility	Fission reactors (incl. Gen-4)	Fusion reactor first wall	Spallation targets
Energy	1-3MeV	14MeV	1-1000 MeV P^+: 0.6 - 3GeV
dpa rate (in Fe)	< 1.0E-6 /s	< 1.0E-6 /s	< 0.7E-6 /s
dpa range (in Fe)	< 200	< 50 to 200	< 50
He per dpa (in Fe)	< 1	~ 10	< 80
H per dpa (in Fe)	< 1	~ 40	< 400
Temperature (° C)	270 - 950	300 – 800	50 - 600

note: rough values!

Courtesy of Y. Dai

→ large database from measurements in reactors has to be used with care to predict radiation damage at accelerators

Effects due to H/He production

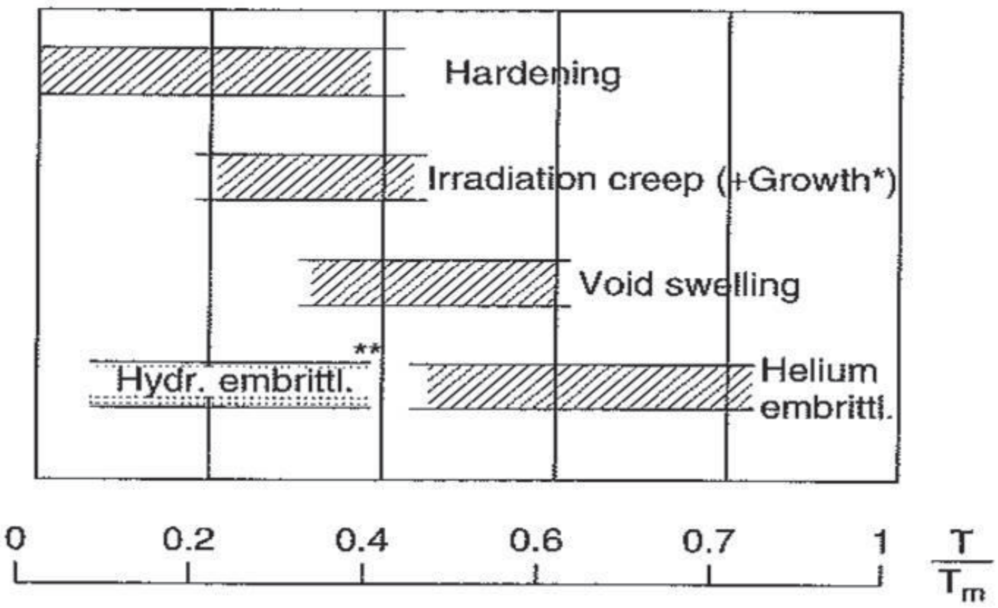
Hydrogen: very mobile

- e.g. for steel: large fraction leave the material (> 250 °C)
- in metals which form hydrids (W, Ta) →H-embrittlement

Helium: mobile

- induce embrittlement particularly at high temperatures
- influence on swelling, can stabilize voids
- formation of blisters and exfoilation on the surface

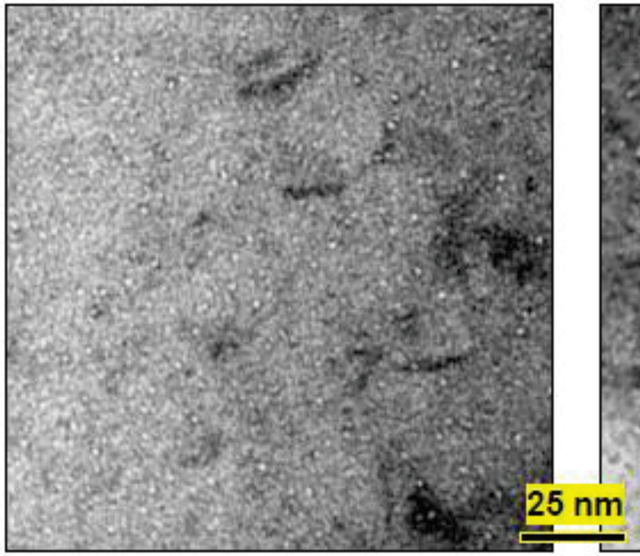
→ lots of studies with He beam/implantation



T_m: melting temperature

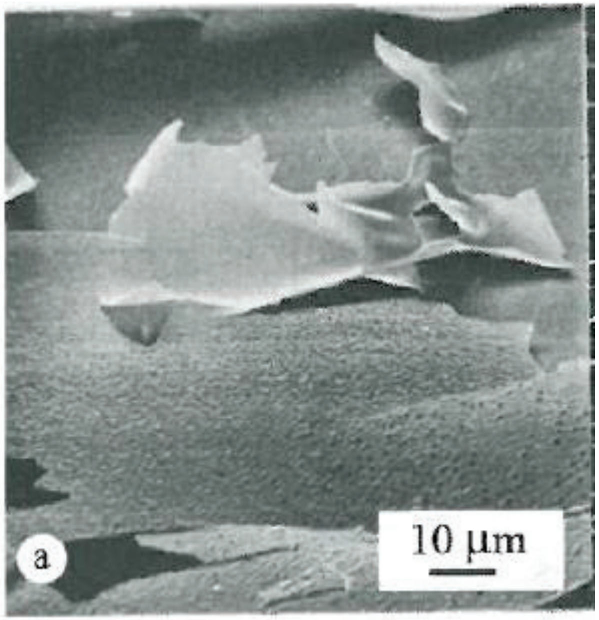
11 DPA, 1020 appm He, 353 °C

TEM

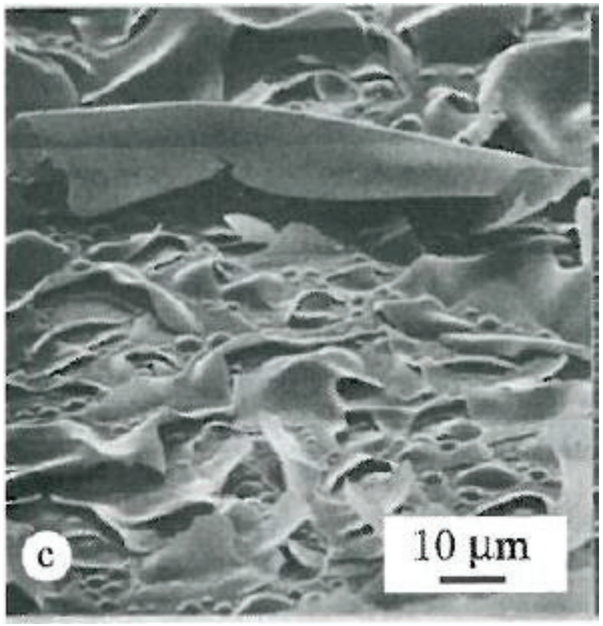


Blistering/Exfoliation due to Helium

$2*10^{18}$ ^{4}He/cm^{2}, 100 keV,
normal incidence

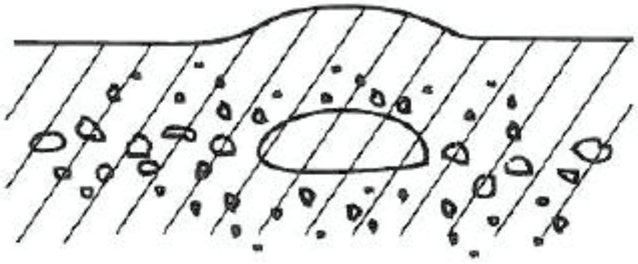


$1*10^{19}$ ^{4}He/cm^{2}, 100 keV
normal incidence



polycrystalline
nickel

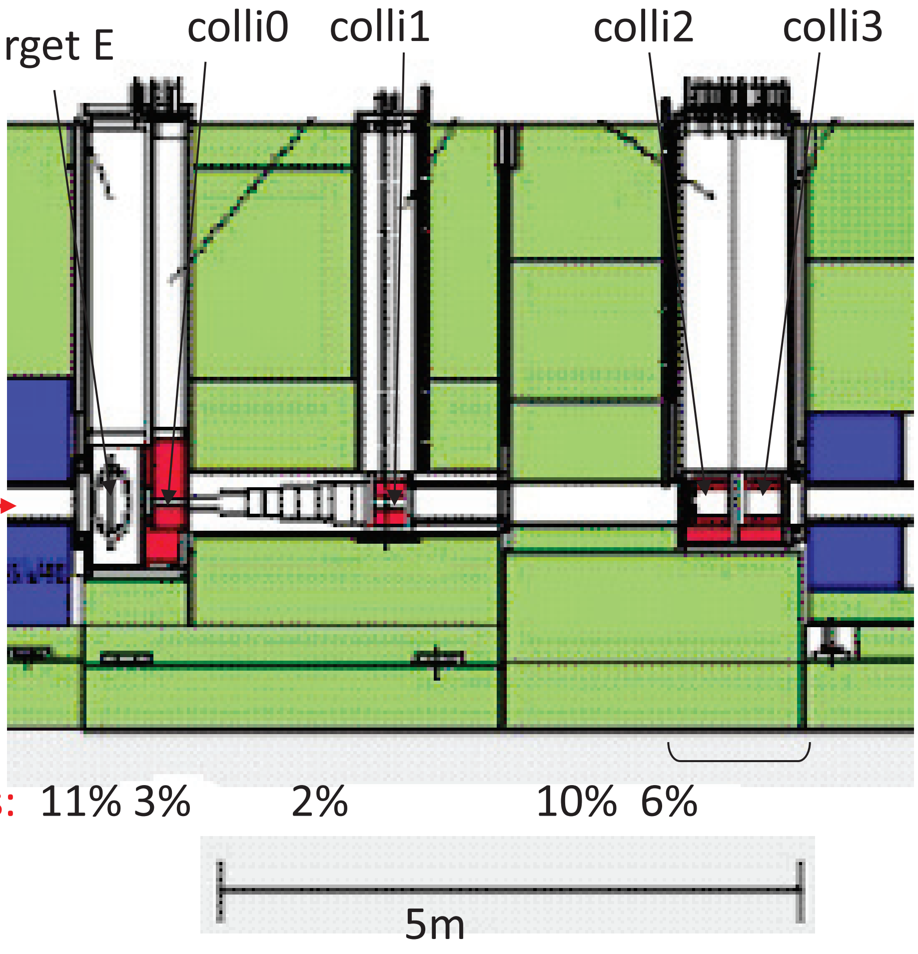
G. Was, Fundamentals of Radiation Material Science, Springer, 2007



Bubbles built-up larger bubbles (coalescence)
Pressure between bubbles causes stress
→ not balanced by the surface tension
→ fractures on the surface

Collimator system behind Target E

Beamline:



beam loss: 11% 3% 2% 10% 6%

Target E:

4 cm graphite wheel

→ beam spread (~ 6 mrad) due to multiple scattering

collimator system:

OFHC Cu

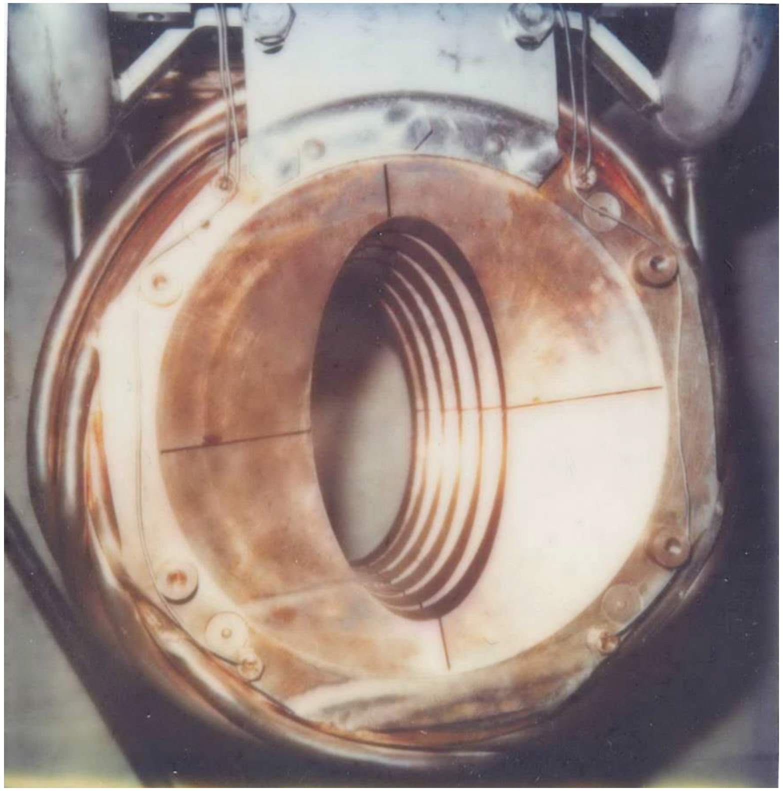
Protection of the beam line:
Reducing beam losses and subsequent activation
→ high power deposition

→ about 30% beam loss
(depends on actual beam optics)

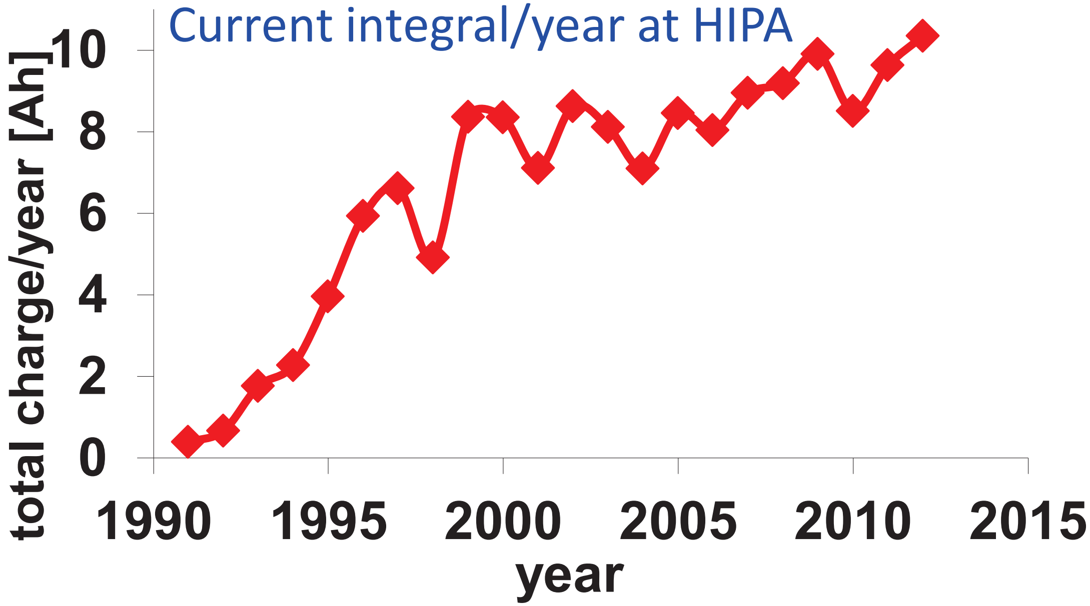
KHE2: Overview

Operation: from 1990-2012
on Target E: 147 Ah
absorbed by KHE2: ~ 10%

KHE2: 1990 before installation

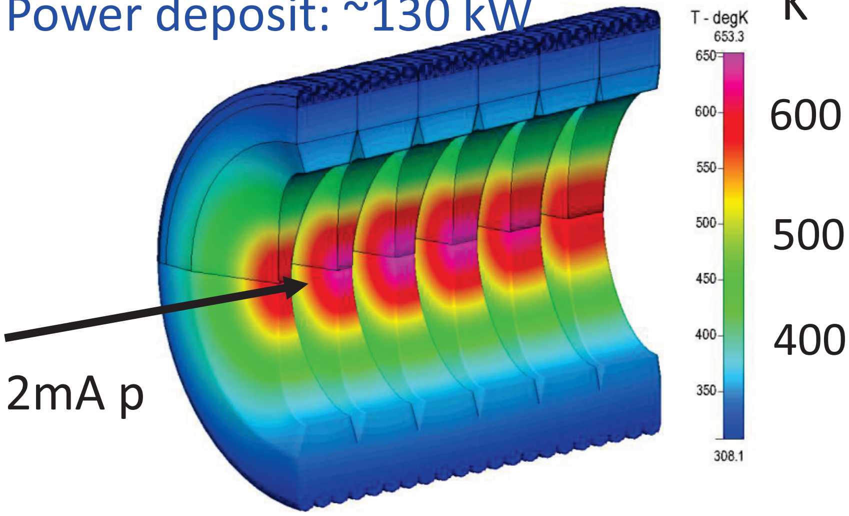


6 sections, cooling pipes

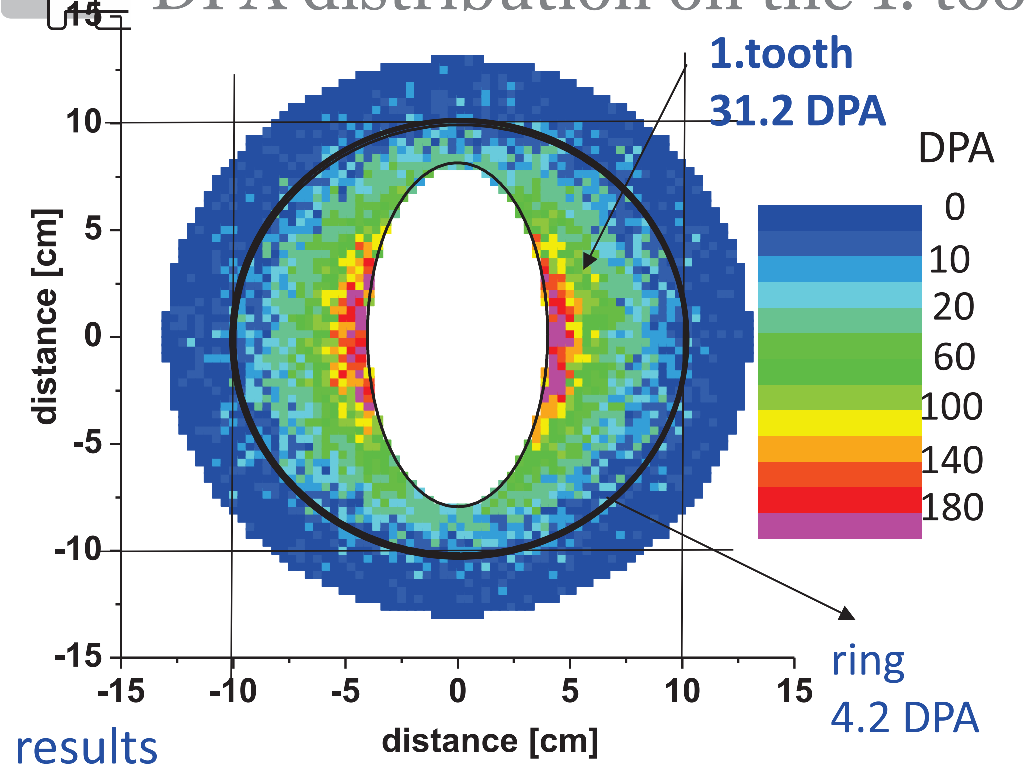


Current integral/year at HIPA

Temperature distribution: max: 380 °C
Power deposit: ~130 kW



DPA distribution on the 1. tooth of KHE2

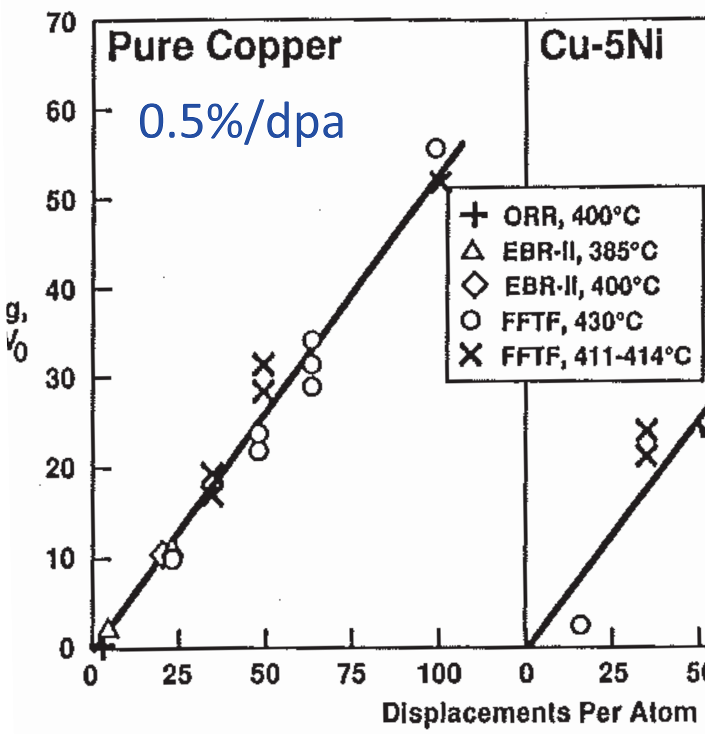


1.tooth
31.2 DPA

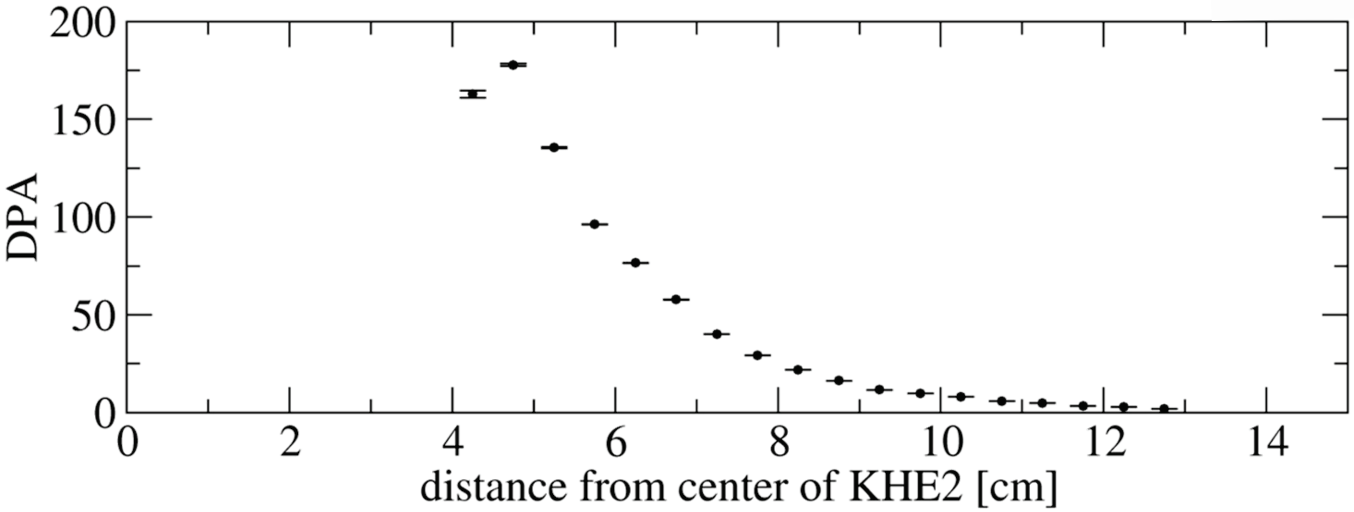
ring
4.2 DPA

MARS results

Swelling %$\Delta V/V_o$



0.5%/dpa



Radial dependence of DPA on 1. tooth

very high DPA at the inner side

Results after inspection in hot cell

Measurement of the horizontal opening via 2 laser distance meters:
result: very close to original values
accuracy: 0.5 mm
→ no swelling observed

expected: $\Delta V/V=0.5\%/\text{DPA}$
$V=10\text{x}10\text{x}10\text{mm}^3$, 80 DPA → $\Delta l = 2\text{x}1.2$ mm

Blisters on the surface at
vertical & horizontal direction,
but not in between

Possible explanation:
thermal expansion/movement close
to the slits, highest stress

Damage might be due to gas production:

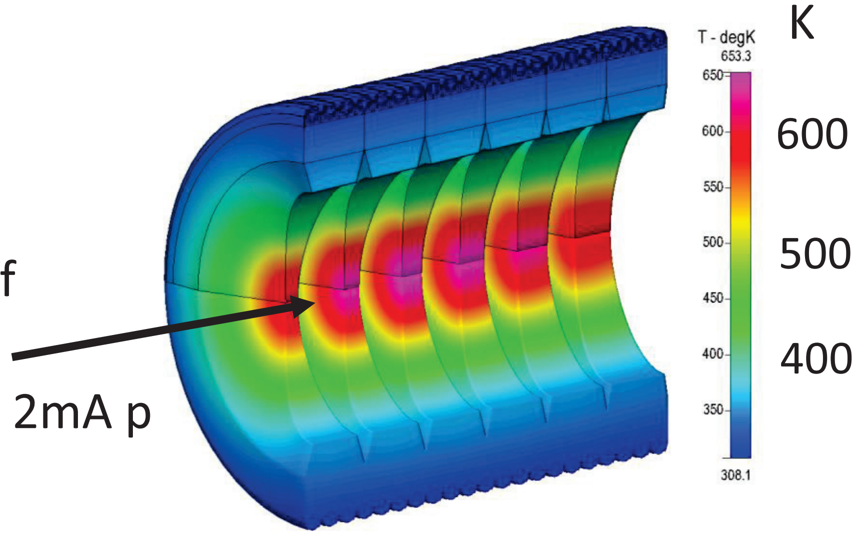
He ~ 240 appm H ~ 5050 appm	MCNPX: Bertini-Dresner, mean values of KHE2 (inner ring)

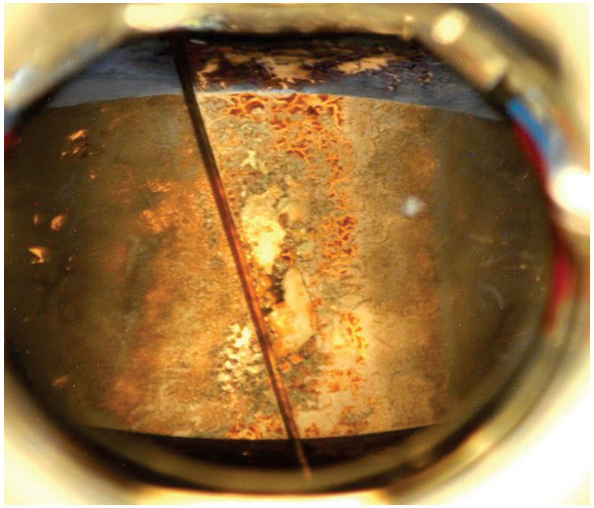
Results after inspection in hot cell

- vertical: 80 - 100 °C (2mA)
  different appearance at entry & exit:

- some pieces (1-2 mm height) peel off

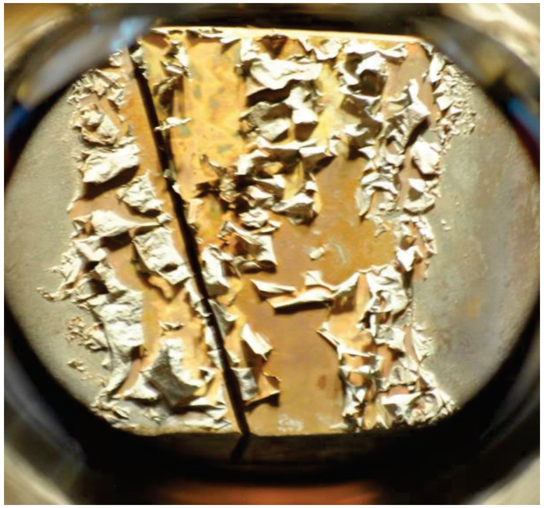
- grey surface
  → guess: errosion + dirt





entry

'rusty': erosion?, blisters peeled off?



exit

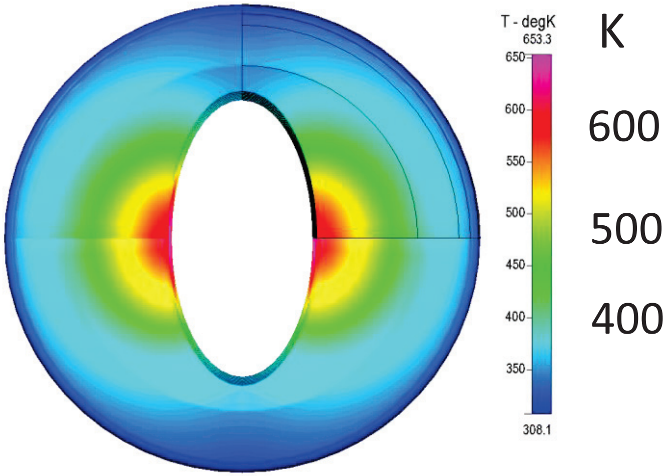
Dirt from Target E (graphite)?

Results after inspection in hot cell

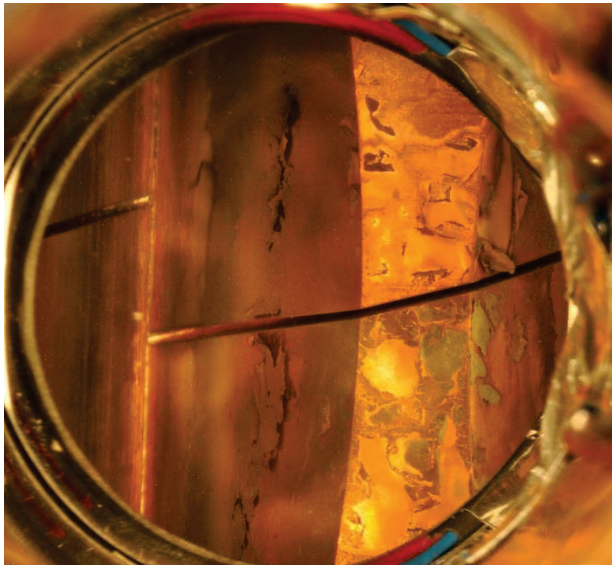
- horizontal: 350 - 400 °C (2 mA)
  main damage at entry
  → higher temperature, more DPA

- large pieces peel off

- no swelling or deformation at slits

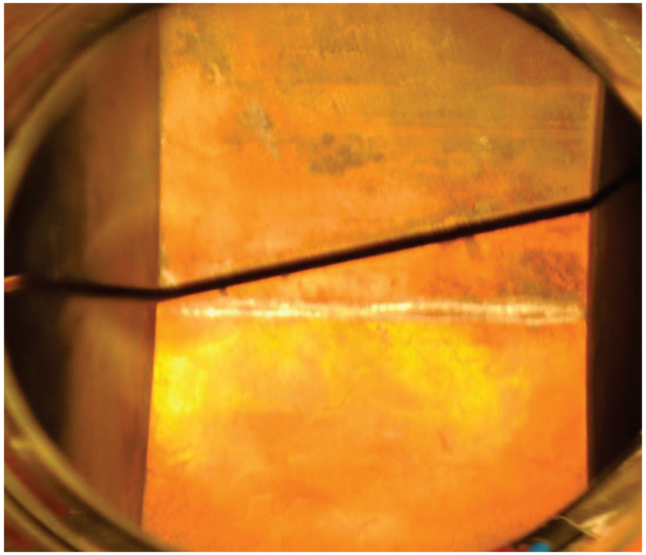


entry



slits completely intact !

exit



no damage seen here and at the back:
(much less He production)

Summary/Conclusion

- Derivation & calculation of DPA
  DPA: not a realistic measure of defects, just for quantification,
  neglects many details of the irradiation
- Athermal & thermal heating effect is significant
  - Healing depends on the density of defects & damage signature
  - Annealing depends on the time, temperature (→ migration)
- Visualizing of defects: TEM
  → Counting of the number of defects (many different kinds)
- Effect of Hydrogen/Helium: Embrittlement, Swelling, Blistering

- Large database from measurements in reactors has to be used with care to predict radiation damage at high-power accelerators .
- Macroscopic effects might be very different under different conditions, particularly for large DPA.
- Database for radiation damage from high energetic particles scarce.

How to transfer mechanical/physical property changes measured on thermal/fission reactor neutrons to high-energy particle beams?

→damage correlation

→very complex problem

Irradiation test experiments are needed!