SOTERIA

EFFECT OF NEUTRON FLUX ON THE MICROSTRUCTURE OF IRRADIATED RPV STEELS

A. Ulbricht, F. Bergner – HZDR

HZDR
HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF

Contributions from:
E. Meslin – CEA
M. Hernández-Mayoral, E. Oñorbe, M. González, P.P. Valdivieso – CIEMAT
A. Etienne, B. Radiguet – CNRS/Uni Rouen-GPM
H. Hein – Framatome
M. Brumovsky – UJV

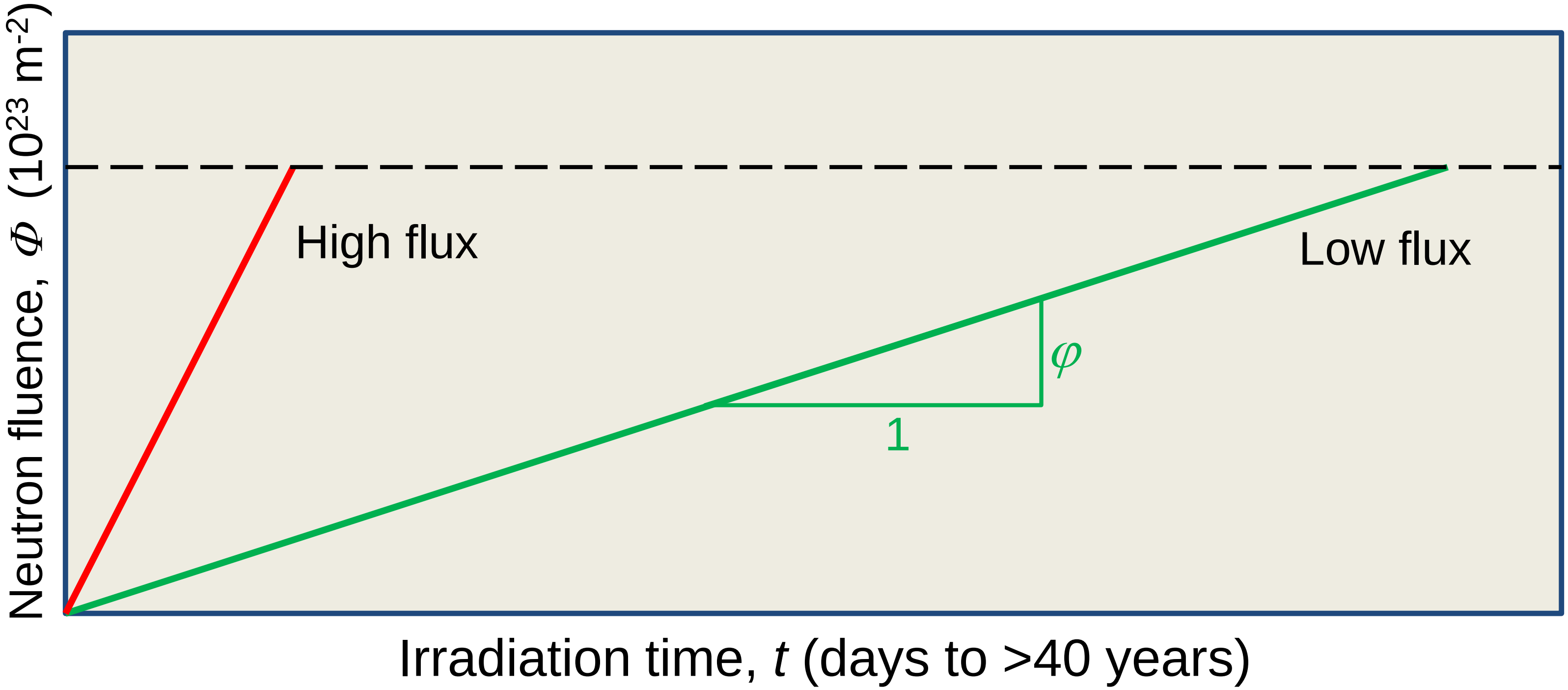
This project received funding under the Euratom research and training programme 2014-2018 under grant agreement N° 661913

Outline

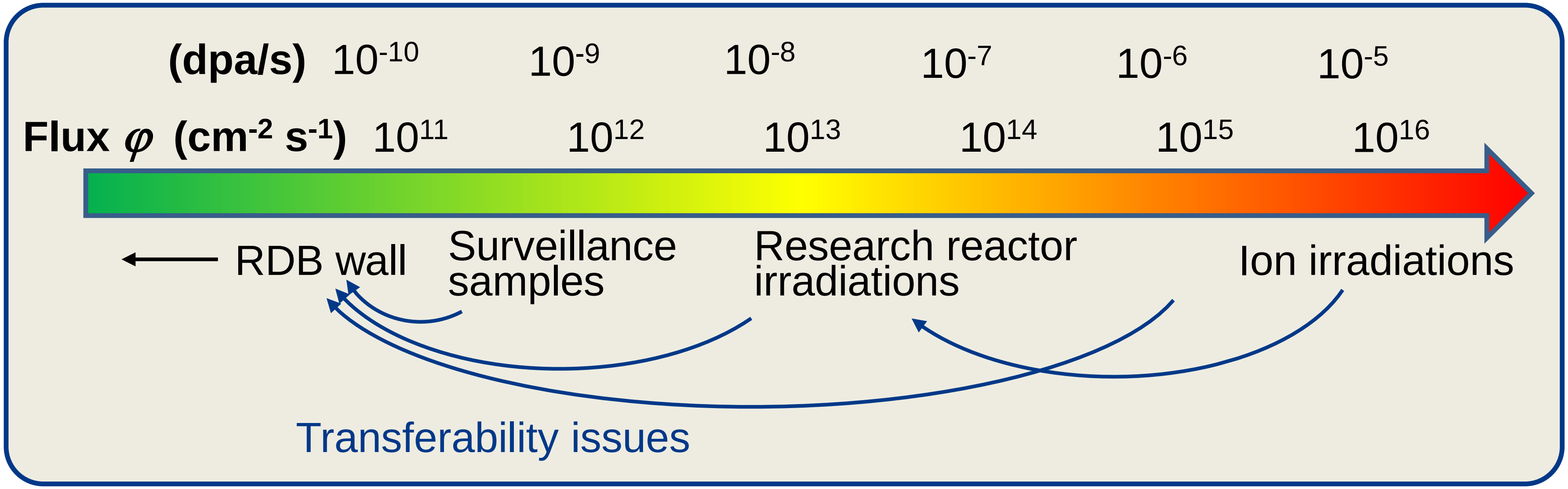
1. Introduction
   - Approaches to flux effects
   - Summary of reported flux effects on mechanical properties
2. Flux effect on microstructure
   - Transmission electron microscopy (TEM)
   - Atom probe tomography (APT)
   - Positron annihilation spectroscopy (PAS)
   - Small-angle neutron scattering (SANS)
   - Summary of experimental findings
3. Modelling of flux effects
   - Radiation-enhanced diffusion (rate theory model)
   - Precipitation kinetics (JMAK-type model)
   - Hardening (dispersed barrier hardening)
   - Embrittlement (LDO hypothesis)
4. Summary and conclusions

Introduction

- The natural variables to describe neutron exposure are irradiation time *t* and neutron flux φ.
- The primary parameter governing irradiation damage of materials is the neutron fluence, $\Phi = \varphi t$. → TT shift = f(Φ).
- Does the flux φ at which the <u>same</u> fluence $\Phi = \varphi t$ is accumulated make a difference? → TT shift = f(Φ, φ)?

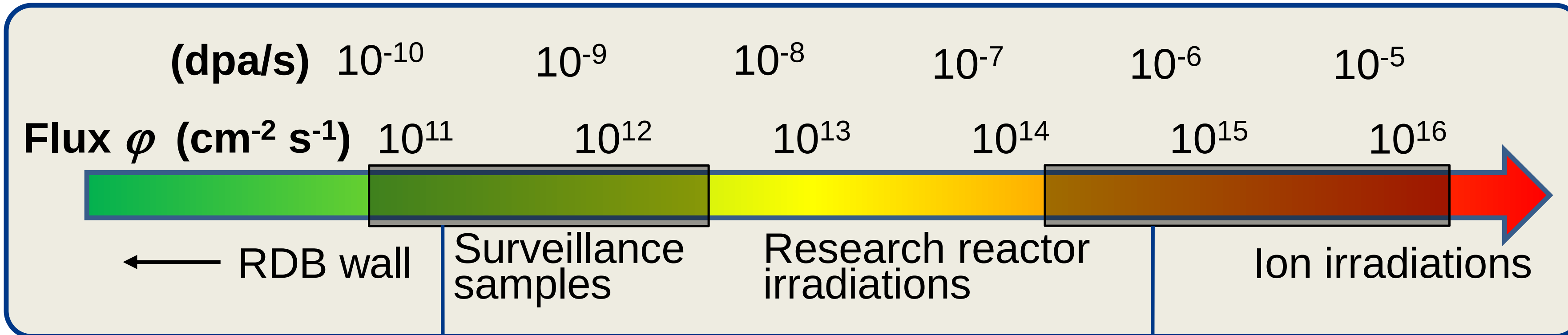


Introduction



- 1 dpa ≈ 0.7 x 10^{21} cm^{-2} (E > 1 MeV)
- 1 dpa ≈ 1 x 10^{21} cm^{-2} (E > 0.5 MeV)
- Spectrum effects and attenuation through the RPV wall (UJV contribution)

Introduction



EU project SOTERIA

CIEMAT, GPM/Uni Rouen, HZDR → This talk

CEA → Independent study

Approaches to flux effects

- How to isolate the effect of the secondary parameter **flux** from the effect of the primary parameter **fluence**? Two approaches:

(1)

Collect data with fluence and flux varied simultaneously in wide ranges and use statistics to isolate flux effects.

↓

Mechanical properties

(2)

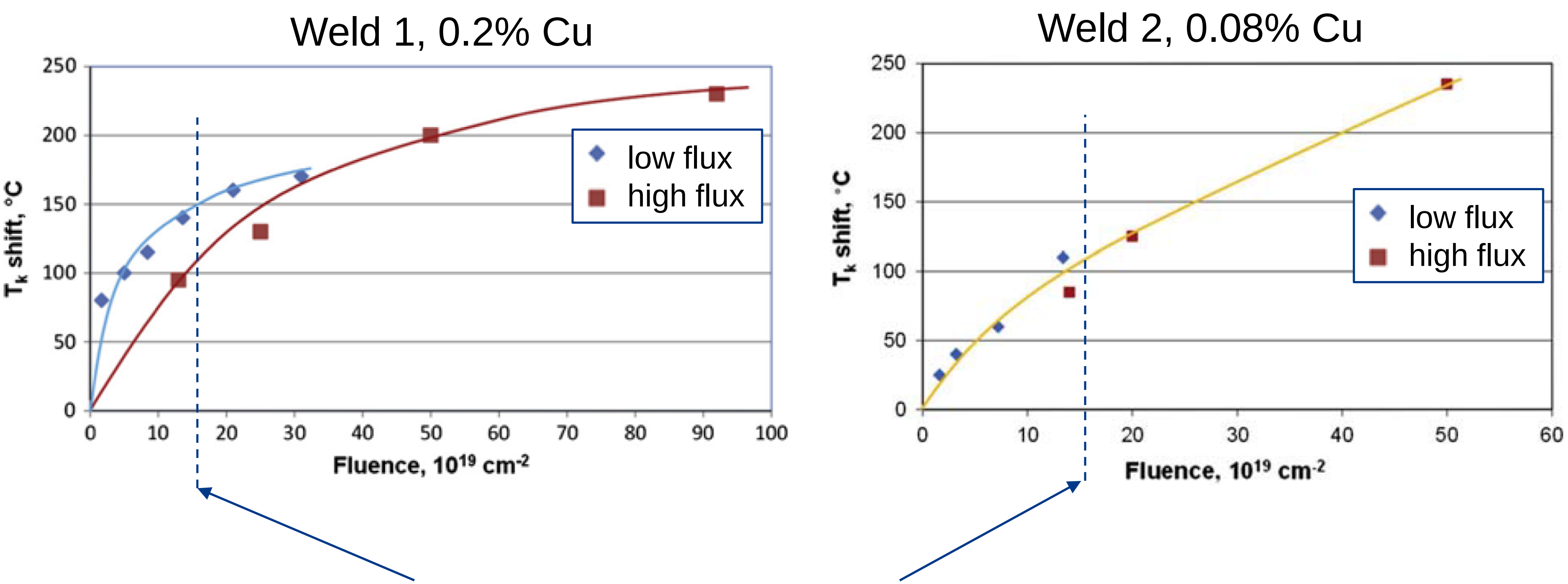
Characterize pairs of samples irradiated at different fluxes up to the <u>same</u> fluence.

↓

Microstructure

Reported flux effects on mechanical properties

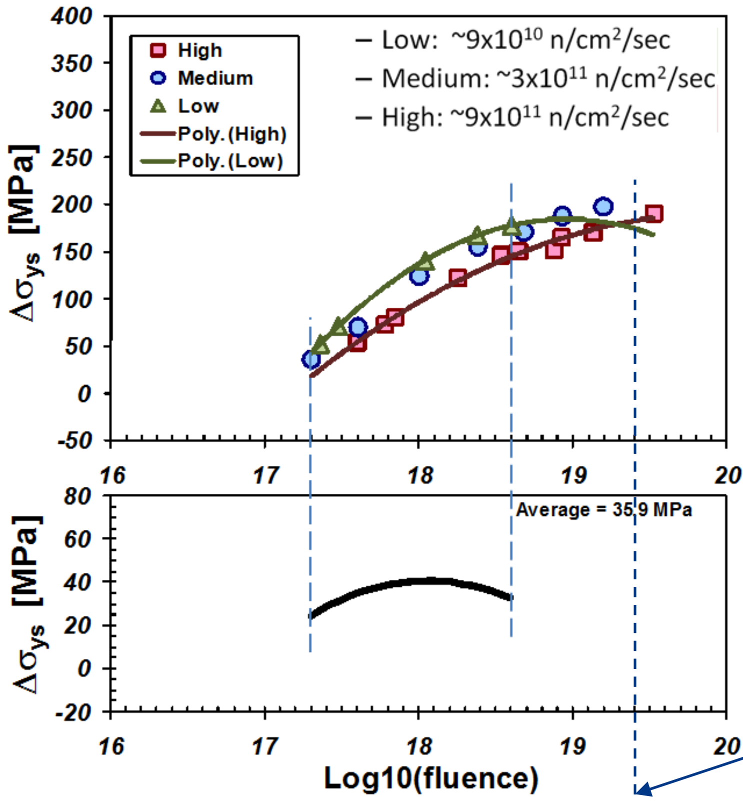
- Flux effect on brittle-ductile transition temperature shift (ΔT_k) for VVER-440 weld material
  [Kryukov et al., J. Nucl. Mater. 443 (2013) 171]



Design end-of-life fluence of VVER-440:
≈ 1.6 x 10^{20} cm^{-2} (≈ 0.24 dpa) (E > 0.5 MeV)

Reported flux effects on mechanical properties

- Flux effect on yield stress increase ($\Delta\sigma_y$) for western reactors
  [M. Kirk, In: Proc. of the IAEA Technical Meeting on Radiation embrittlement and Life Management of Reactor Pressure Vessels, Znojmo, 2010]



Single RPV steel (out of many)

Design end-of-life fluence of western RPVs:
$\approx 4 \times 10^{19}$ cm^{-2} ($\approx$ 0.06 dpa) (E > 1 MeV)

Reported flux effects on mechanical properties

Conclusions from previous work

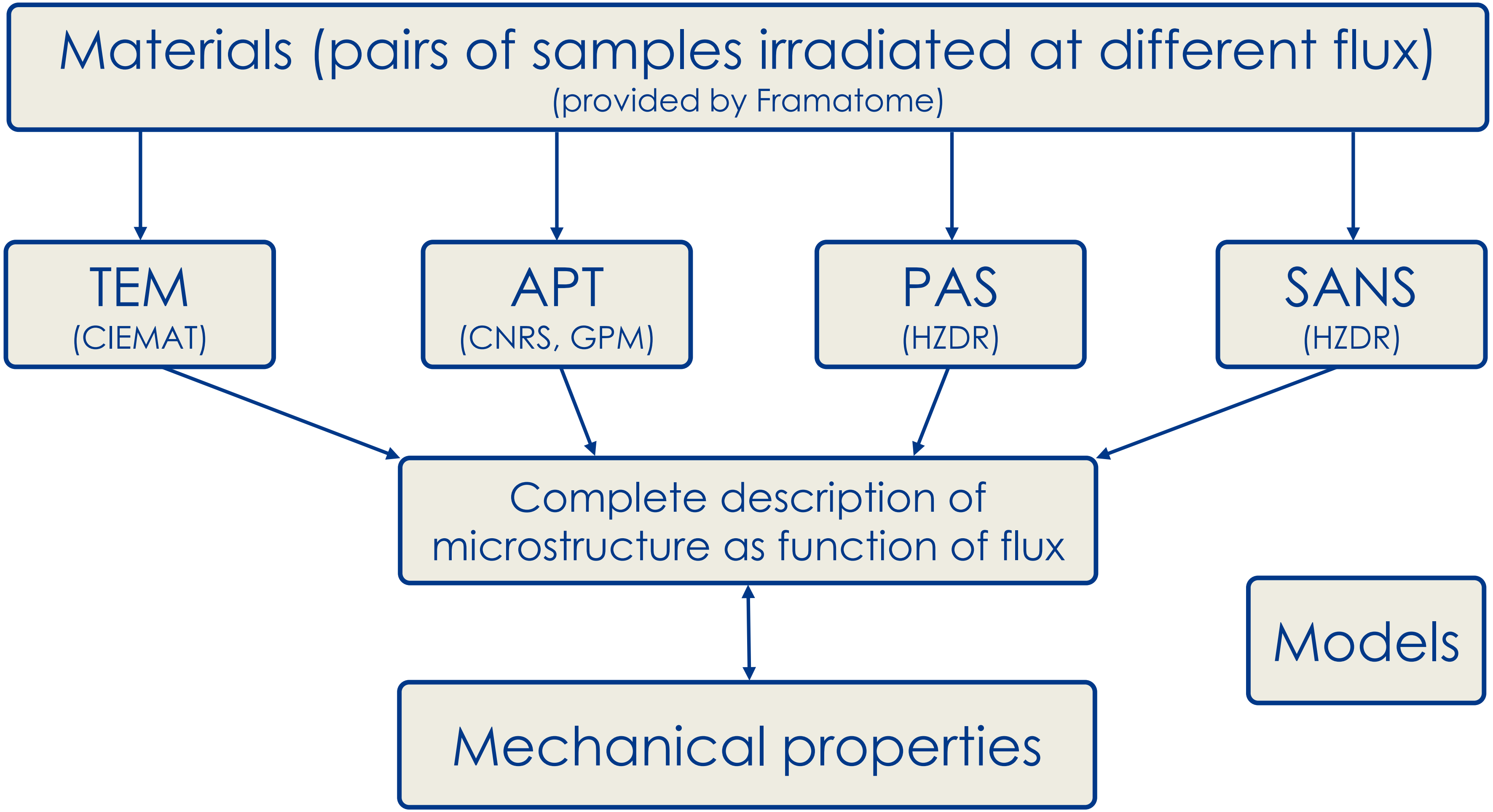
- The magnitude of the flux effect depends strongly on Ni and requires a minimum Cu to operate.
- Flux effect likely small for existing reactor steels.
- Flux effect likely unobservable for new reactor steels.

[*M. Kirk, In: Proc. of the IAEA Technical Meeting on Radiation embrittlement and Life Management of Reactor Pressure Vessels, Znojmo, 2010*]

Can flux effects be ignored?

- No, from the viewpoint of data scatter and uncertainties.
- No, from the viewpoint of microstructure evolution.
- No, from the viewpoint of multiscale modelling.

2. Flux effect on microstructure



Materials

Material		Composition (wt-%)								
		C	Mn	Si	Cr	Ni	Mo	V	P	Cu
Base	ANP-3	0.32	0.70	0.20	0.44	0.98	0.79	-	0.015	**0.12**
	ANP-10	0.18	0.81	0.15	0.40	0.96	0.53	< 0.01	0.006	**0.09**
Weld	ANP-6	0.05	1.41	0.15	0.07	1.69	0.46	0.004	0.012	**0.08**
	VFAB-1	0.063	1.66	0.21	0.14	0.90-1.47	0.8	0.01	0.016	**0.06**

Material		Irradiation conditions (E > 1 MeV)	
		Neutron fluence (10^{19} cm^{-2})	Neutron flux (10^{12} cm^{-2} s^{-1})
Base	ANP-3	**3.99**	**1.83 (high flux)**
	ANP-10	**3.38**	**0.047 (low flux)**
Weld	ANP-6	**5.70**	**2.51 (high flux)**
	VFAB-1	**5.87**	**0.082 (low flux)**

Flux factor

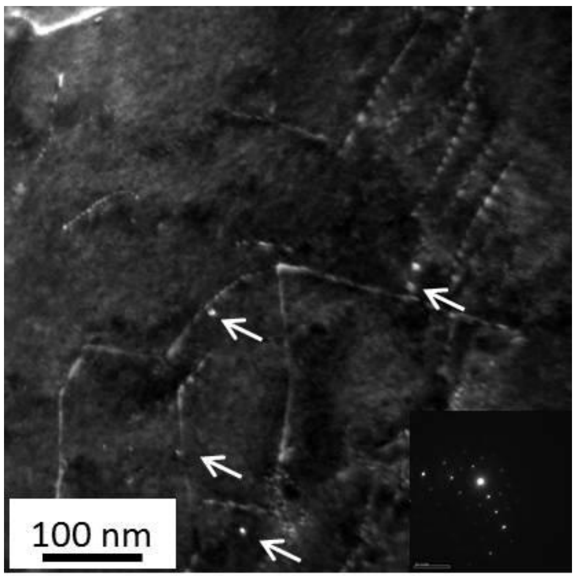
39

31

- Samples for TEM, APT, PAS and SANS provided by Framatome

TEM

- TEM was performed at CIEMAT.
- TEM is well suited to identify irradiation-induced dislocation loops.



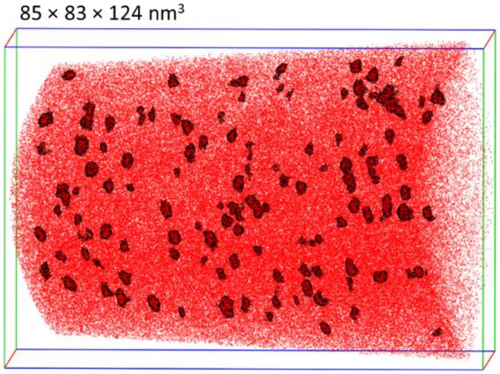
WBDF image for ANP-10
Arrows indicate loops.
(example for illustration)

Conclusions:

- ✓ the damage produced in the form of dislocation loops is **low** (≈ 10^{19} – 10^{20} m^{-3}, size ≈ 4 nm),
- ✓ **Base** metal: loop size similar for low and high flux, number density higher for low flux,
- ✓ **Weld** metal: loop size and number density similar for low and high flux.

APT

- APT was performed at GPM (Uni Rouen).
- APT provides information on the composition of irradiation-induced solute atom clusters (+ size, number density and volume fraction).



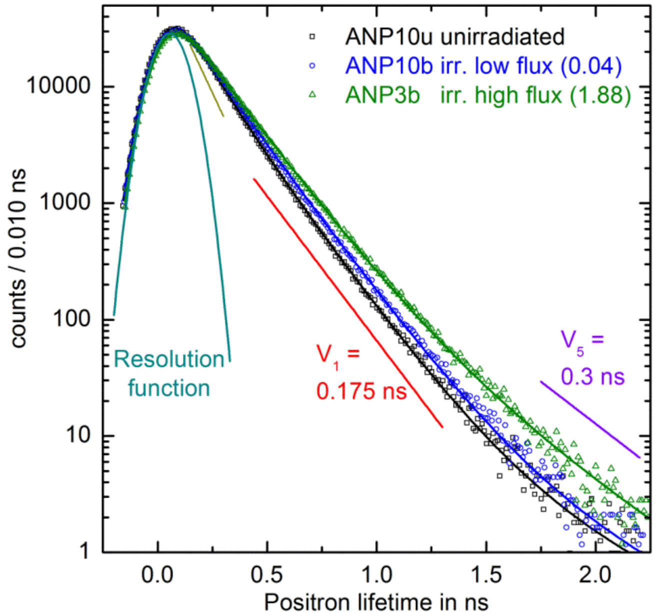
Atom map for weld metal ANP-6, low flux (example for illustration)

Conclusions:

- ✓ Solute atom clusters and segregated dislocations observed in all materials,
- ✓ segregations along dislocations lines and solute clusters are more enriched with solute atoms for low flux,
- ✓ solute clusters are slightly smaller in samples irradiated at high flux,
- ✓ cluster number density is in the same range for all samples.

PAS

- Gamma-induced Positron Spectroscopy (GiPS) was performed at the ELBE facility of HZDR.
- PAS provides information on open volume defects such as sub-nm vacancy clusters.



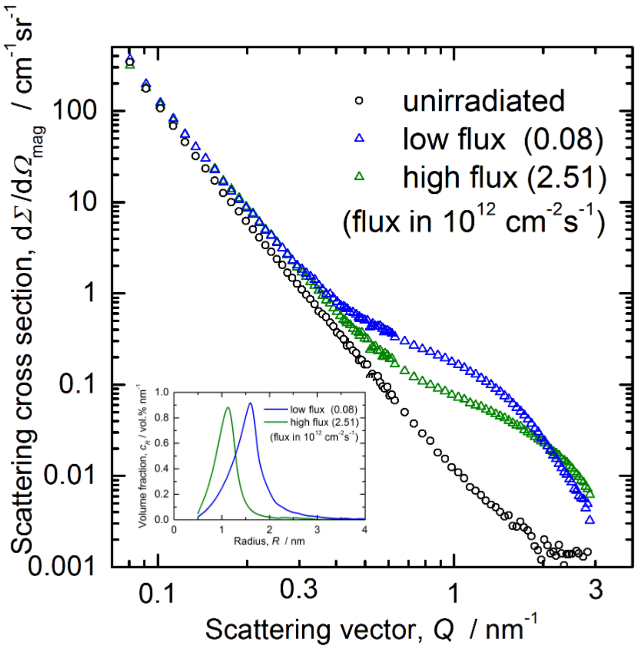
Lifetime spectra for ANP-3/-10 (example for illustration)

Conclusions:

- ✓ Irradiation-induced vacancies and sub-nm vacancy clusters (<10 vacs.) observed in all irradiated materials,
- ✓ Mean lifetime increases with increasing flux indicating a higher concentration of vacancies.

SANS

- SANS was performed by HZDR at beamline V4 of HZB Berlin.
- SANS provides macroscopically representative and statistically reliable averages of size and volume fraction of solute atom clusters.



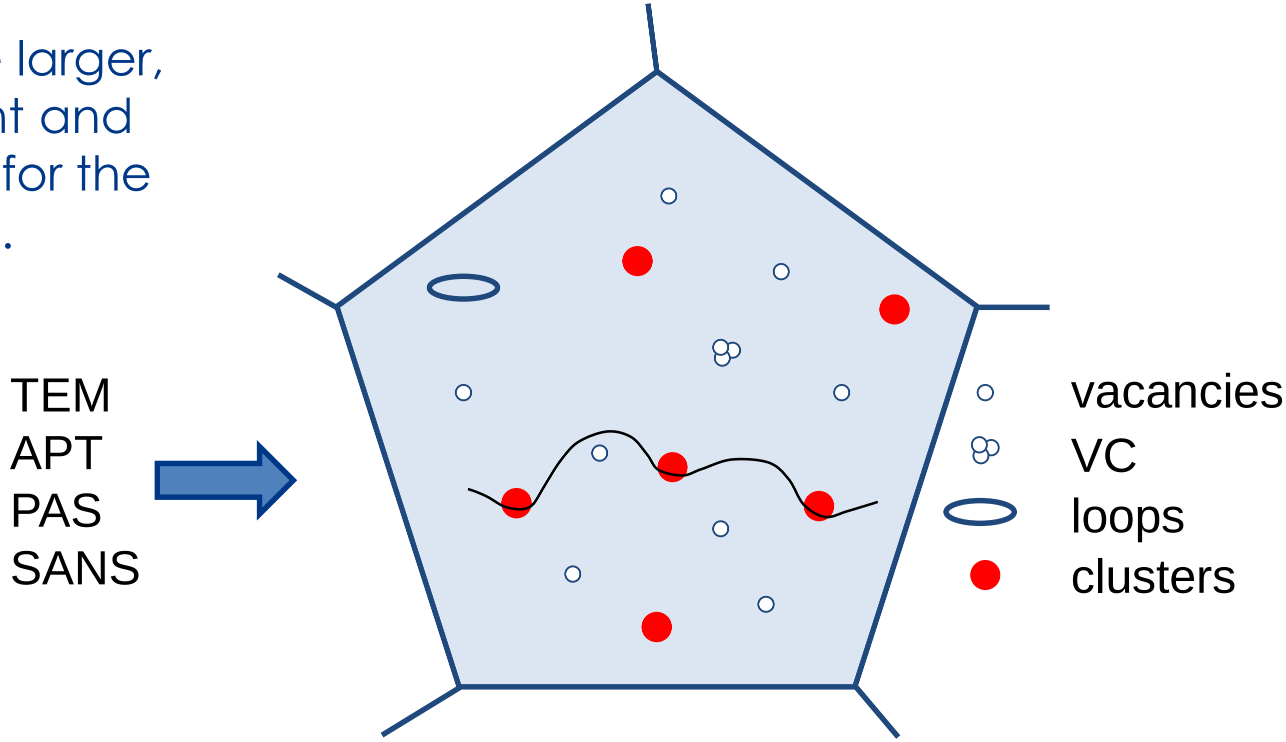
SANS: ANP-6 vs. VFAB-1

Conclusions:

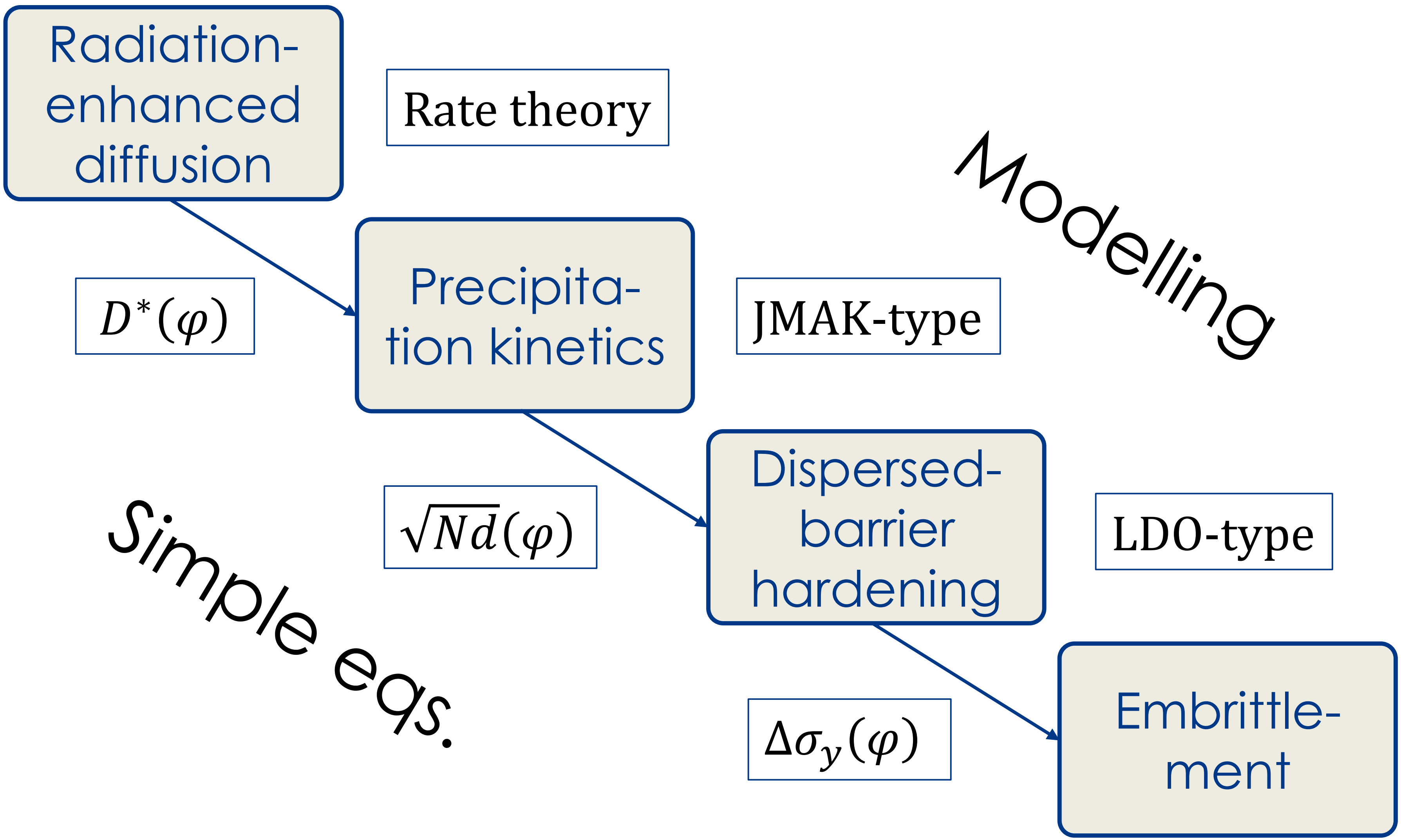
- ✓ Solute atom clusters observed in all materials (radius 0.8 – 1.2 nm, number density 10^{23} – 10^{24} m^{-3}),
- ✓ clusters larger for low flux,
- ✓ number density smaller for low flux,
- ✓ volume fraction slightly larger for low flux.

Summary of experimental findings

- We have observed irradiation-induced vacancies, VCs, loops and solute atom clusters (CRPs, MNPs and mixed forms).
- Vacancies and VCs are too small and loops are too rare to contribute significantly to hardening (but they play a role in cluster evolution).
- Clusters are larger, less frequent and less diluted for the lower fluxes.



3. Modelling of flux effects



Radiation-enhanced diffusion

- Point defect evolution (Harkness & Li 1971)
- Radiation-enhanced diffusion (Odette 1983)

Point defect evolution

$$\frac{dC_v}{dt} = G_v - \frac{4\pi r (D_v + D_i) C_v C_i}{\Omega_a} - K_v C_v$$

$$\frac{dC_i}{dt} = G_i - \frac{4\pi r (D_v + D_i) C_v C_i}{\Omega_a} - K_i C_i$$

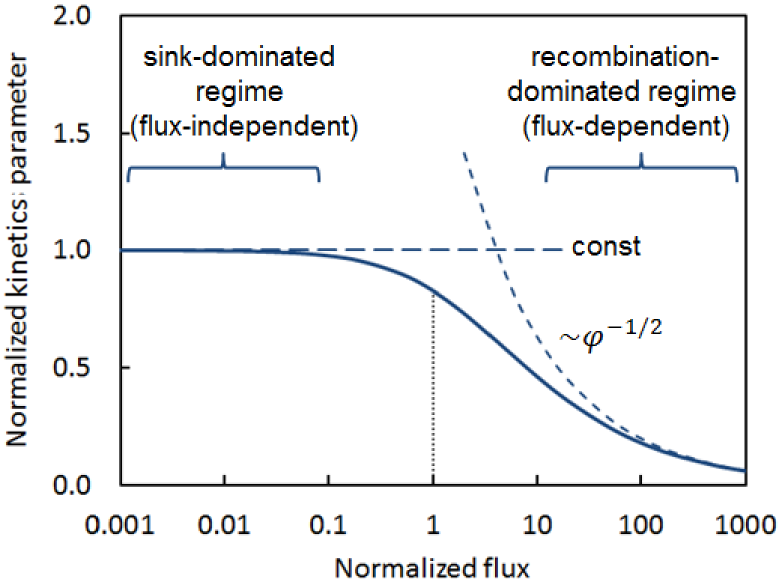
Steady state vacancy concentration

$$C_{vss} = 2.4 C_{vss,t} \left(\sqrt{1 + \frac{\varphi}{\varphi_t}} - 1 \right)$$

Radiation-enhanced diffusion

$$D^*_{Cu} = D_{Cu} \frac{C_{vss}}{C_{veq}}$$

$$k = D^*_{Cu} t \propto \frac{C_{vss}}{\varphi}$$

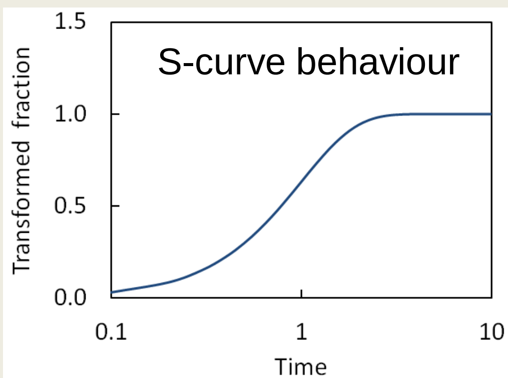


Further reading:
[*Odette et al., Phil. Mag. 85 (2005) 779*]
[*Eason et al., Report ORNL/TM-2006/530, 2006*]

JMAK-type behaviour

- Precipitation kinetics (Johnson-Mehl-Avrami-Kolmogorov, 1937-1940)

$$\frac{f_v(t)}{f_{eq}} = 1 - \exp\left[-c(Dt)^{3/2}\right]$$



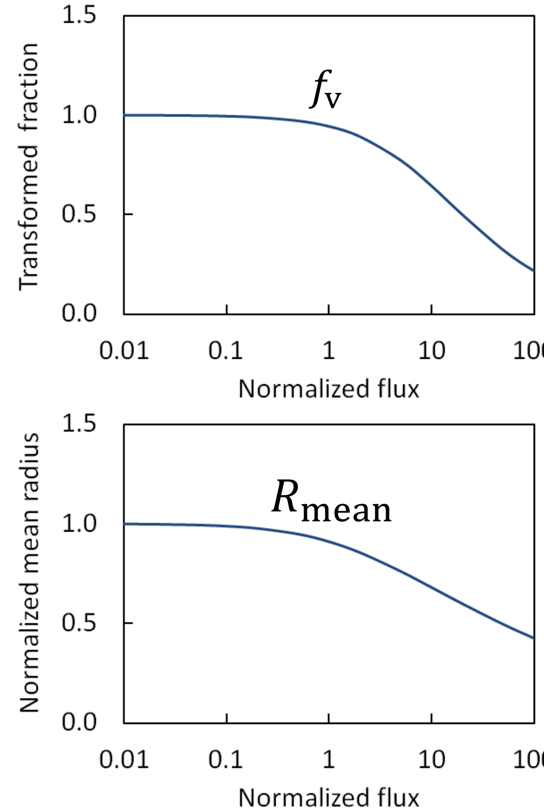
$$R_{mean} \propto \sqrt{Dt}$$

$$N \propto \frac{f_v}{(R_{mean})^3}$$

$$D \rightarrow D^*(\varphi)$$

$$t \rightarrow \frac{\Phi}{\varphi}$$

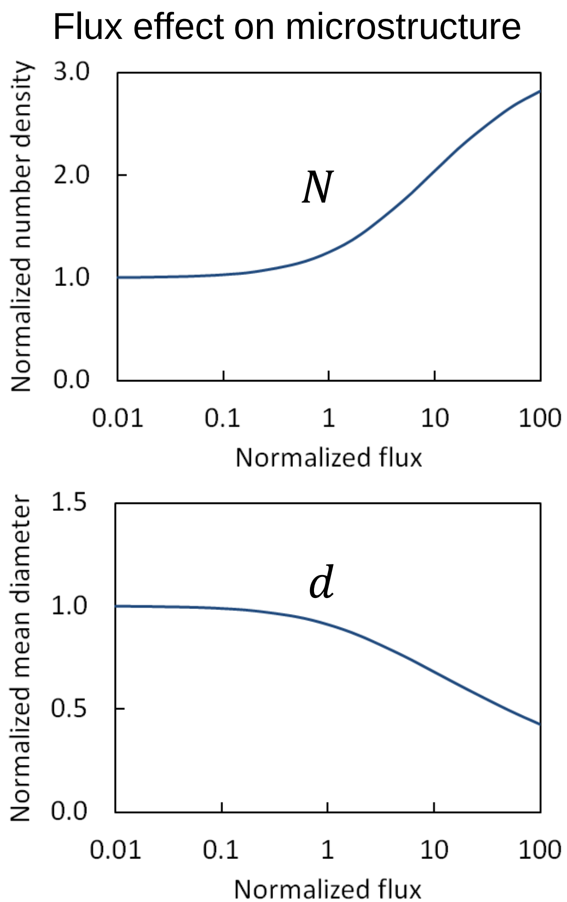
Flux effect on microstructure

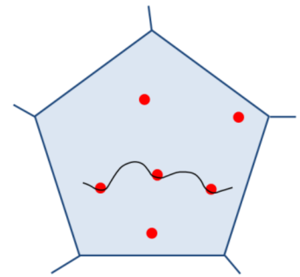


→ Consistent with SANS !

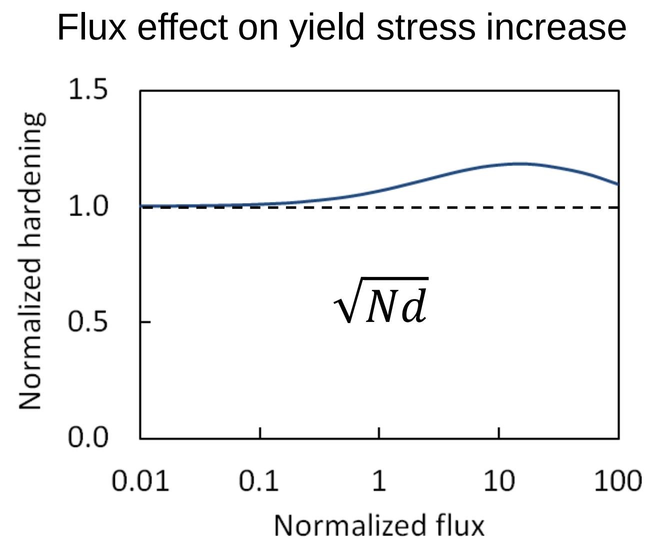
Dispersed-barrier hardening

- DBH model: Coherent zones (clusters) impede dislocation glide (Seeger 1959)





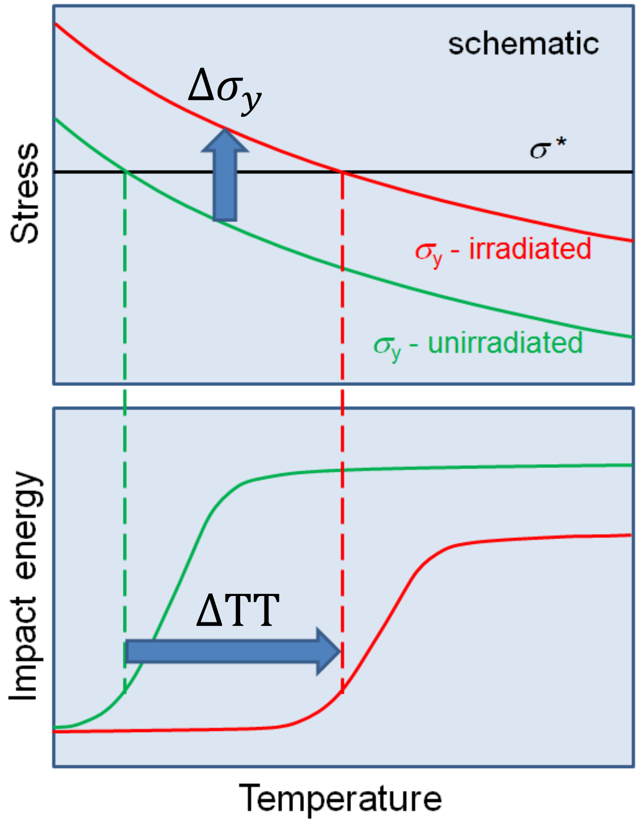
$$\Delta\sigma_y = \alpha M G b \sqrt{Nd}$$



Further reading:
[*Wagner et al., Acta Mater. 104 (2016) 131*]

Embrittlement versus hardening

- Ludwik-Davidenkov-Orowan (LDO) hypothesis: Brittle fracture if $\sigma_y > \sigma^*$



Microscopic cleavage stress σ^* (independent of temperature, unaffected by irradiation)

Yield stress σ_y (decreases at increasing *T*, raised by irradiation)

- ΔTT follows similar trends as $\Delta\sigma_y$
- Expectation: weak or no dependence on flux (except for non-hardening embrittlement)

4. Summary and conclusions

SOTERIA, WP2, Task 2.1

- RPV steel & weld irradiated at different flux up to the same fluence
- TEM, APT, PAS and SANS → complete description of the irradiated microstructure

Conclusions

- Vacancy concentration, loop density, cluster size, cluster density and cluster composition → depend on flux
- Hardening is governed by cluster size and density
- Mechanical properties → no or minor dependence on flux
- This is because:
  - … there is a flux-independent sink-dominated regime ($\varphi \ll 10^{12}\ \mathrm{cm^{-2}s^{-1}}$).
  - … in the flux-dependent recombination-dominated regime, the effects of cluster number density and size on hardening partly cancel out.