FLUX EFFECT ON RPV MATERIALS

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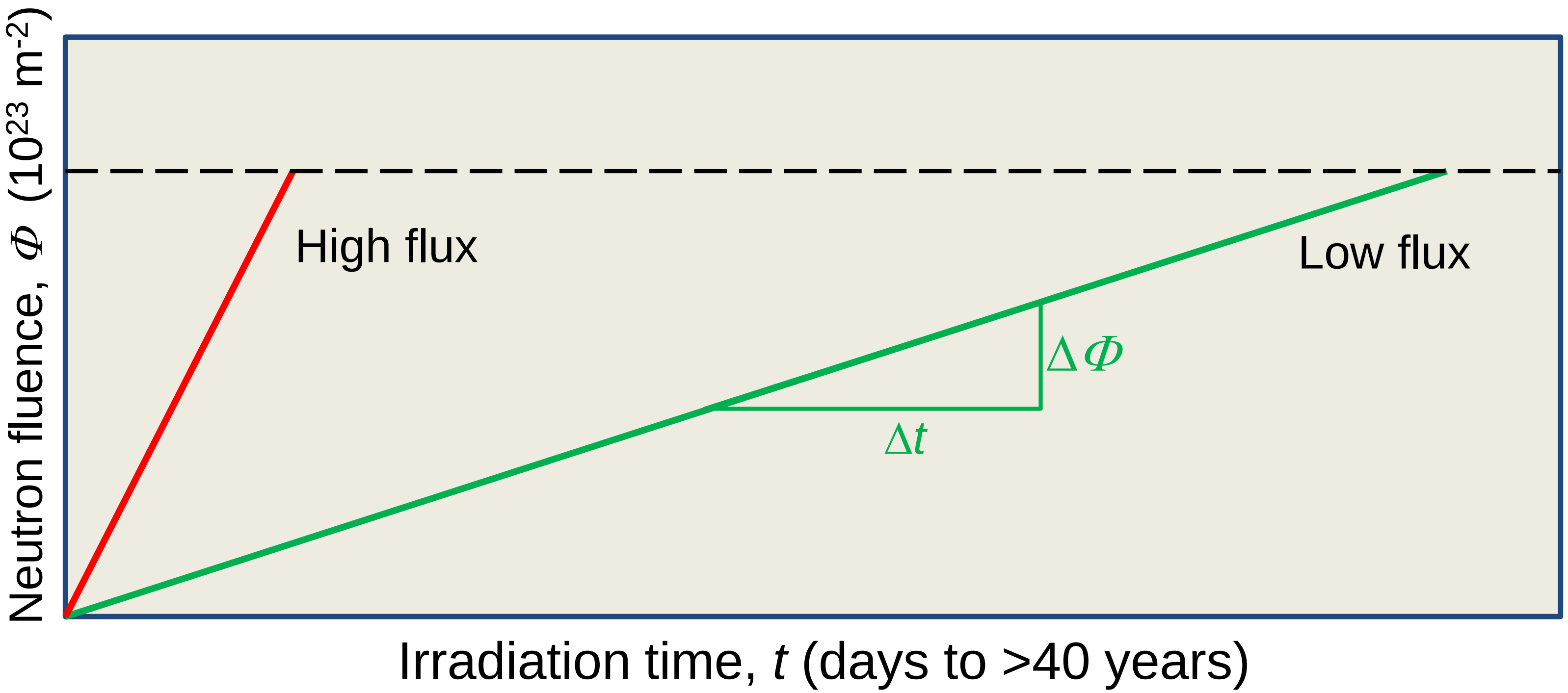
Introduction

- 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

❑ Approaches to flux effects

How to isolate the effect of the secondary parameter **flux** from the effect of the primary parameter **fluence**?

Two approaches:

Collect data with fluence and flux varied simultaneously in wide ranges and use statistics to isolate flux effects.	Mechanical properties
Characterize pairs of samples irradiated at different fluxes up to the same fluence.	Microstructure

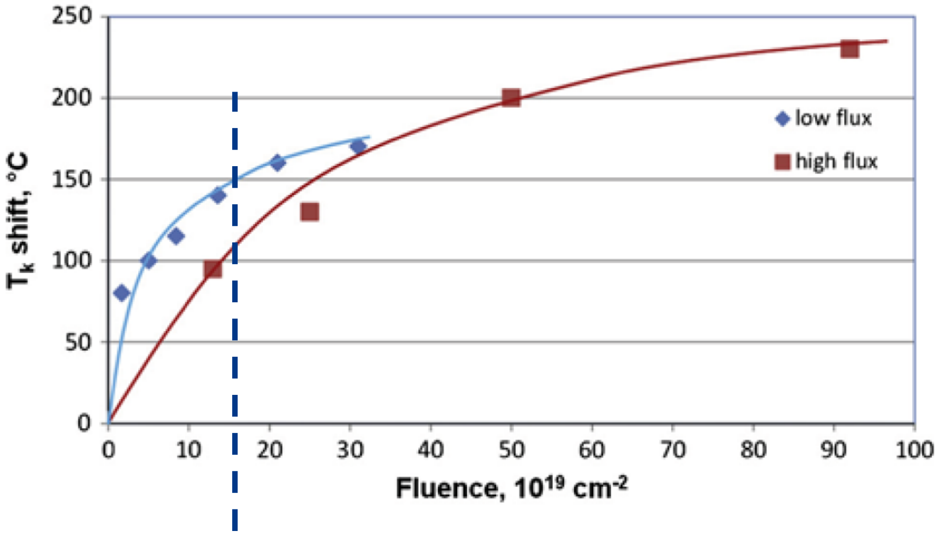
Introduction

❑ Reported flux effects on mechanical properties (example 1)

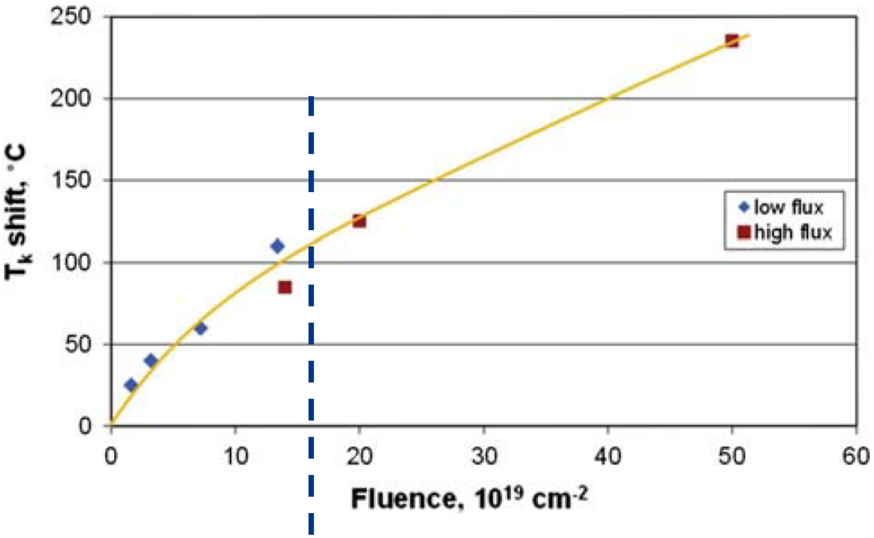
Brittle-ductile transition temperature shift (ΔT_k) for VVER-440 weld material

[*Ref.: A. Kryukov et al. J. Nucl. Mater. 443 (2013) 171*]

Weld 1, 0.2% Cu



Weld 2, 0.08% Cu



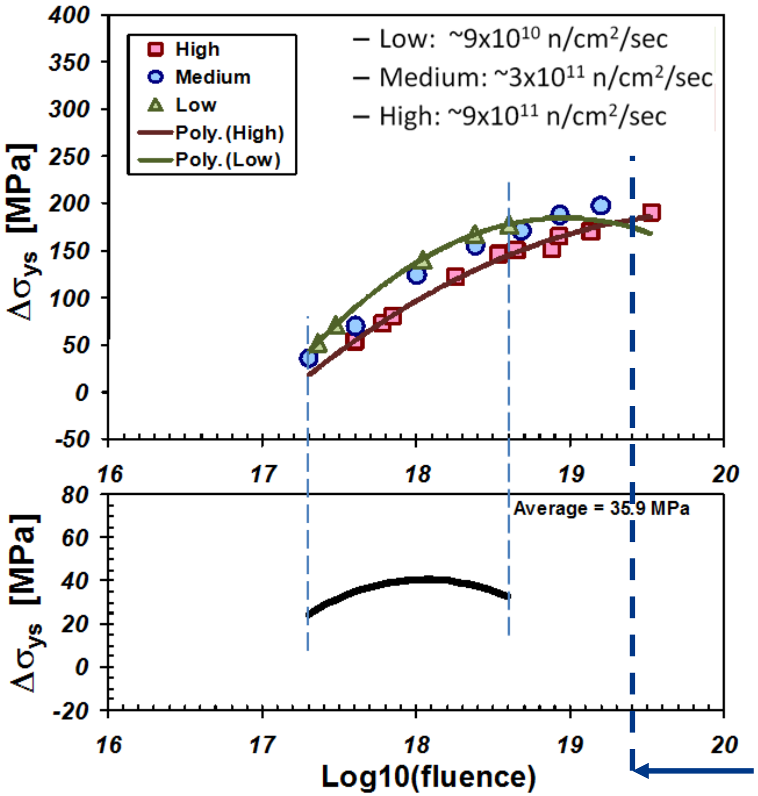
Design end-of-life fluence of VVER-440: ≈ 1.6 x 10^{20} cm^{-2} (E_n > 0.5 MeV) ≈ 0.16 dpa

Introduction

❑ Reported flux effects on mechanical properties (example 2)

Yield stress increase ($\Delta\sigma_y$) for western reactors

[*Ref.: M. Kirk, In Proc. of the IAEA Technical Meeting on Rad. Embrittlement and Life Management of RPVs, Znojmo, 2010*]



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.

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

Introduction

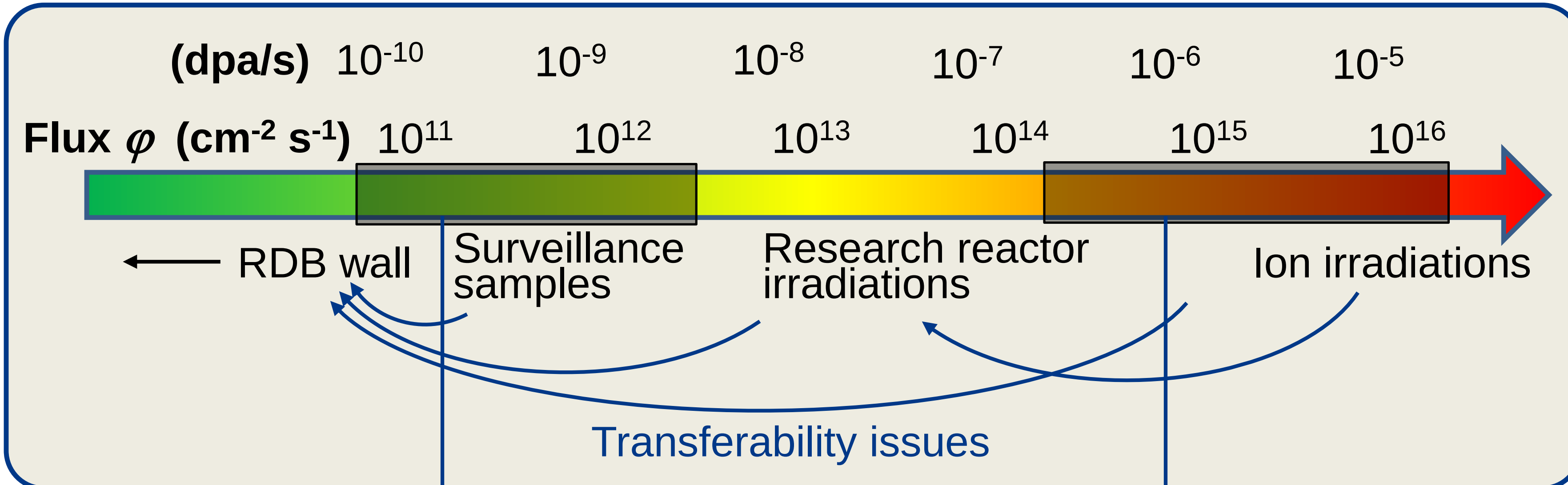
- **Can flux effect be ignored?**
  - No, from the viewpoint of data scatter and uncertainties.
  - No, from the viewpoint of microstructural evolution.
  - No, from the viewpoint of multiscale modelling.

➡ Statistically more reliable microstructural data are needed, in particular for low-Cu RPV steels irradiated up to high fluence.

- **Objectives**
  - Identification of pairs of samples from one and the same material, irradiated at different flux up to the same fluence.
  - Characterization of nanofeatures by complementary techniques.
  - Rationalization of the flux effect on mechanical property changes in terms of irradiation-induced nanofeatures.

Introduction

- 1 dpa ≈ 1 x 10^{21} cm^{-2} (E_n > 0.5 MeV)
- 1 dpa ≈ 0.7 x 10^{21} cm^{-2} (E_n > 1 MeV)



Microstructure changes investigated in WP2.1 by:

CIEMAT
CNRS
HZDR

CEA

- Spectrum φ_E effects on TT shift (**UJV** contribution)

Experiments

❑ Materials and irradiation conditions

- Framatome provided samples for microstructure investigations.

Material		Composition (wt-%)								
		C	Mn	Si	Cr	Ni	Mo	V	P	Cu
Base	ANP-3	0.23	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	1.47	0.8	0.01	0.016	**0.06**

Material		Irradiation conditions ($E_n > 1$ MeV)	
		Neutron fluence (10^{19} cm^{-2})	Neutron flux (10^{12} cm^{-2} s^{-1})
Base	ANP-3	**3.89**	**1.83 (high flux) HF**
	ANP-10	**3.38**	**0.047 (low flux) LF**
Weld	ANP-6	**5.70**	**2.51 (high flux) HF**
	VFAB-1	**5.87**	**0.082 (low flux) LF**

Flux factor

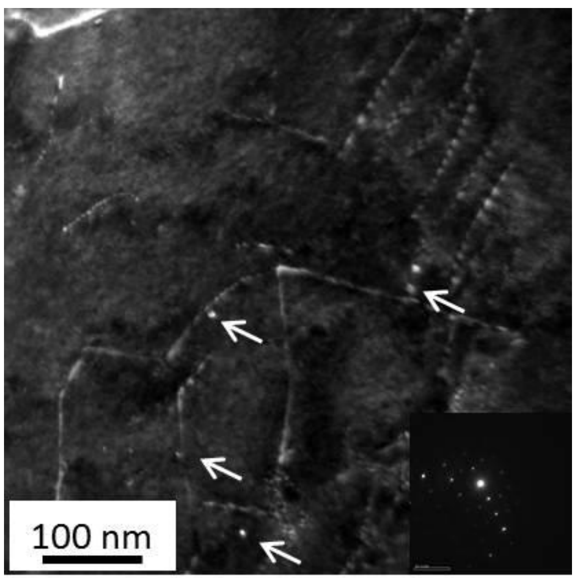
39

31

Results: TEM

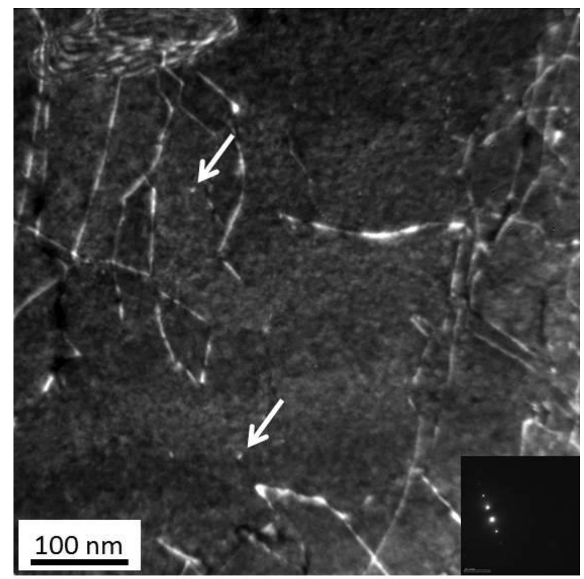
- TEM was performed at CIEMAT.
- TEM is well suited to identify irradiation-induced dislocation loops.

<u>Example</u>: base metal – low flux



WBDF image for ANP-10.
Arrows indicate loops.

<u>Example</u>: weld metal – low flux



WBDF image for VFAB-1.
Arrows indicate loops.

Results: TEM

Material		Dislocation loop	
		Mean diameter (nm)	Density (m^{-3})
Base	ANP-3 (HF)	3.8 ± 0.3	$(8 \pm 4) \times 10^{18}$
	ANP-10 (LF)	4.0 ± 0.2	$(2.0 \pm 0.3) \times 10^{20}$
Weld	ANP-6 (HF)	3.8 ± 0.1	$(1.7 \pm 0.5) \times 10^{20}$
	VFAB-1 (LF)	3.4 ± 0.9	$(1.2 \pm 0.5) \times 10^{20}$

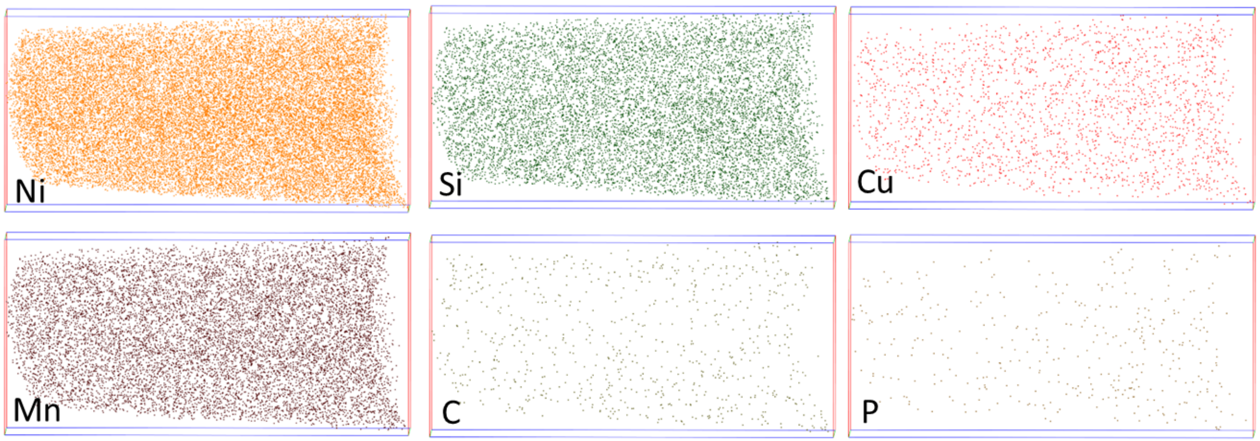
Findings:

- The damage produced in the form of dislocation loops is **low** ($\approx 10^{19} - 10^{20}$ 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.

Results: APT

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

Microstructure of unirradiated reference sample (ANP-3)



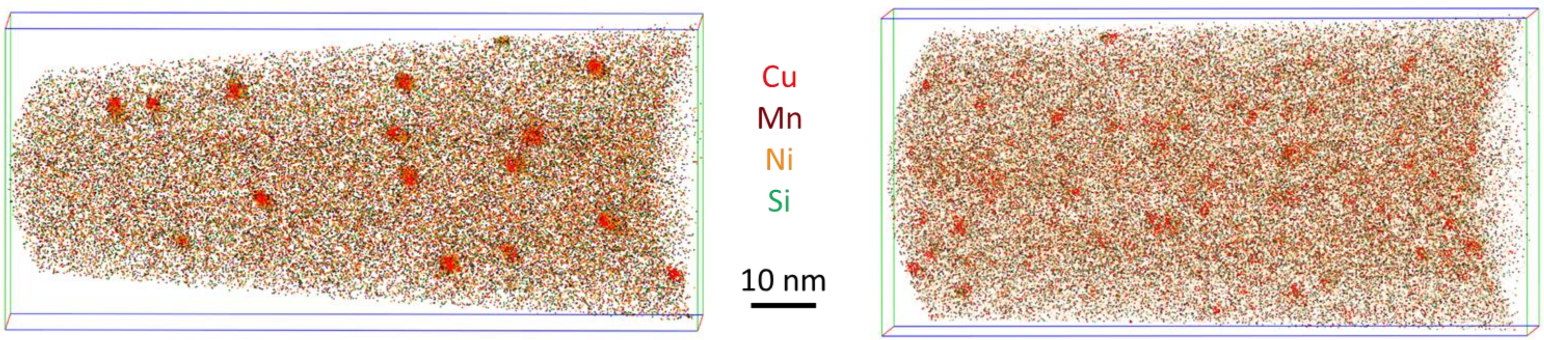
$36 \times 36 \times 76\ nm^3$

- Distribution of solute atoms: no clustering, no segregation
- Lower concentration in C, Mo and Mn than nominal composition
  → presence of carbides but not intercepted in APT

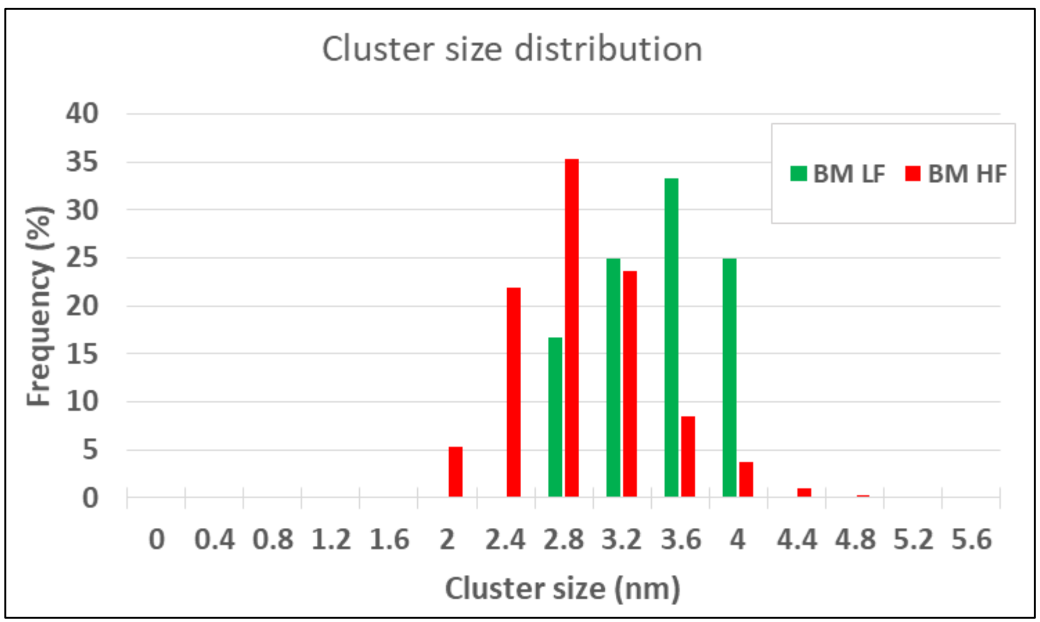
Results: APT

❑ Microstructure after irradiation – base metal

LF



HF



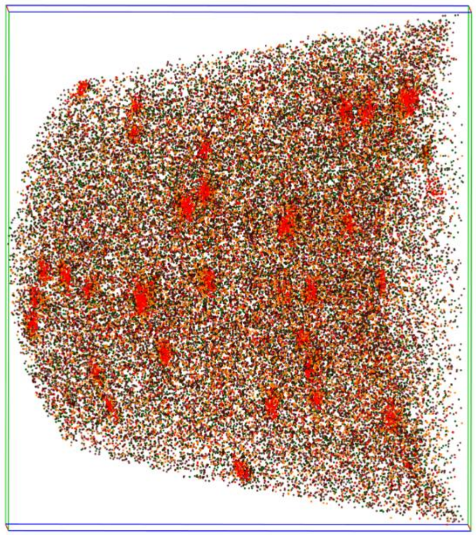
Solute cluster	LF	HF
Density 10^{23} m^{-3}	1.1	3.7
Size nm	3.2	2.7

- Higher density at HF
- Smallest size at HF

Results: APT

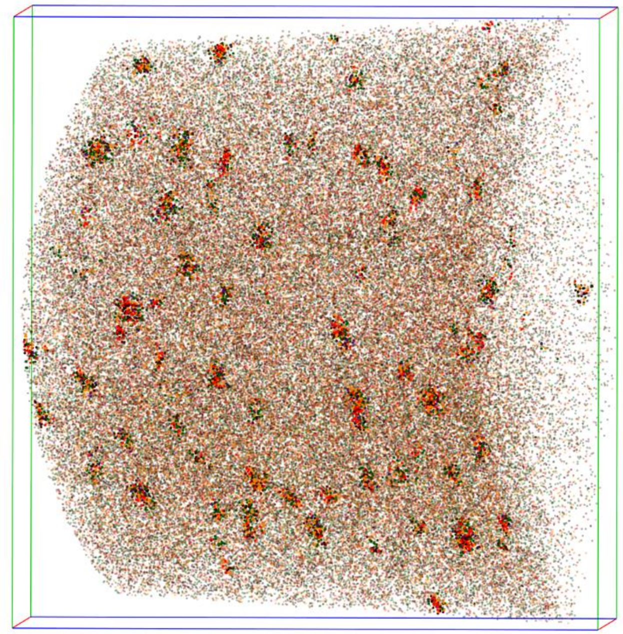
❑ Microstructure after irradiation – weld metal

LF

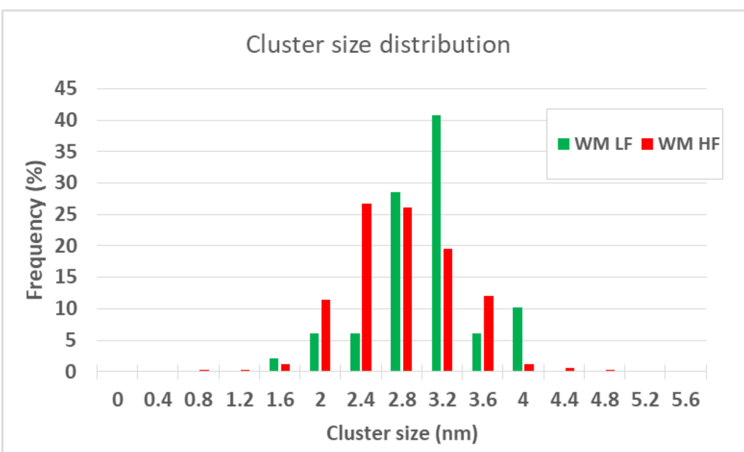


Cu
Mn
Ni
Si

10 nm



HF



Solute cluster	LF	HF
Density 10^{23} m^{-3}	3.0	3.1
Size nm	2.8	2.6

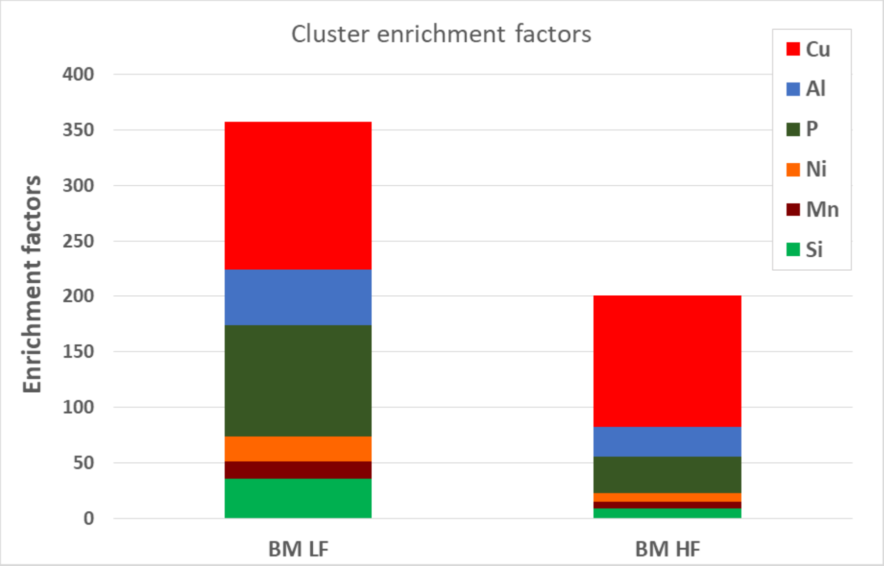
- No difference in density
- Smallest size at HF

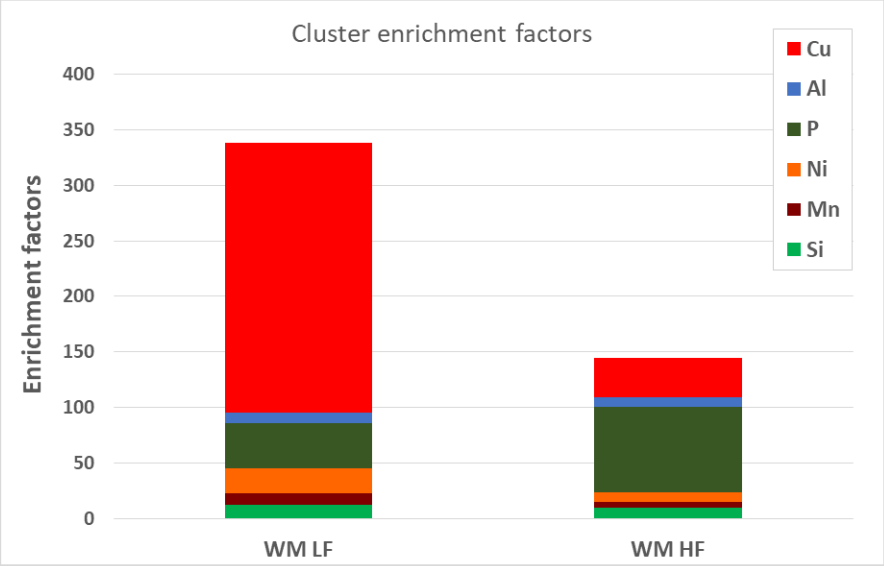
Results: APT

❑ Cluster composition

Base metal

weld metal





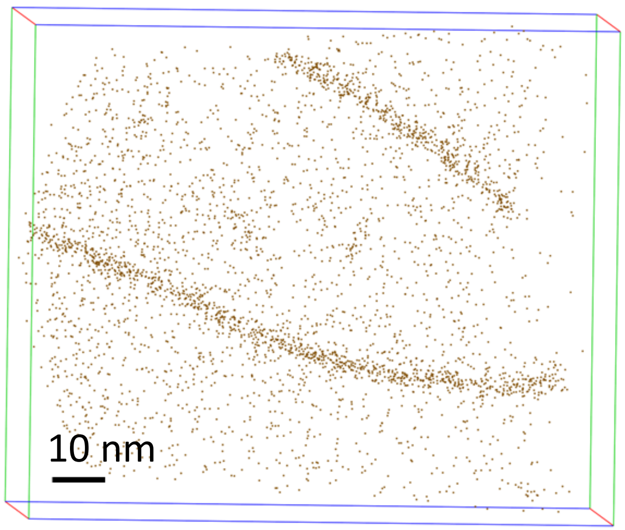
Low flux high flux Low flux high flux

- More enriched cluster at LF for both base and weld metal

Results: APT

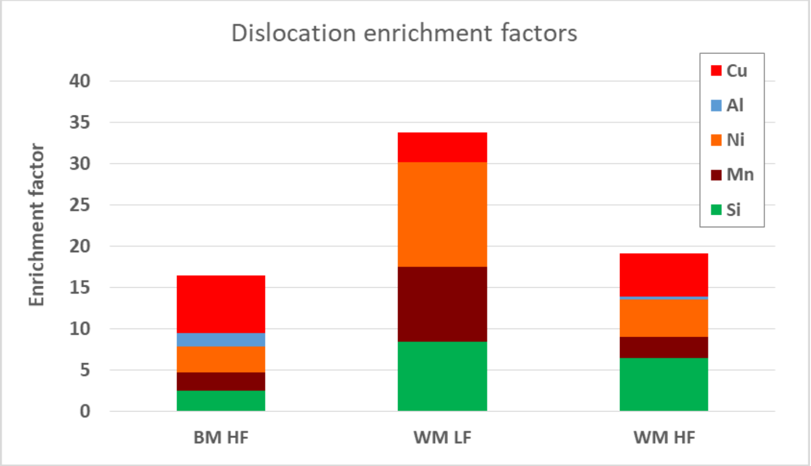
- Segregation at dislocations

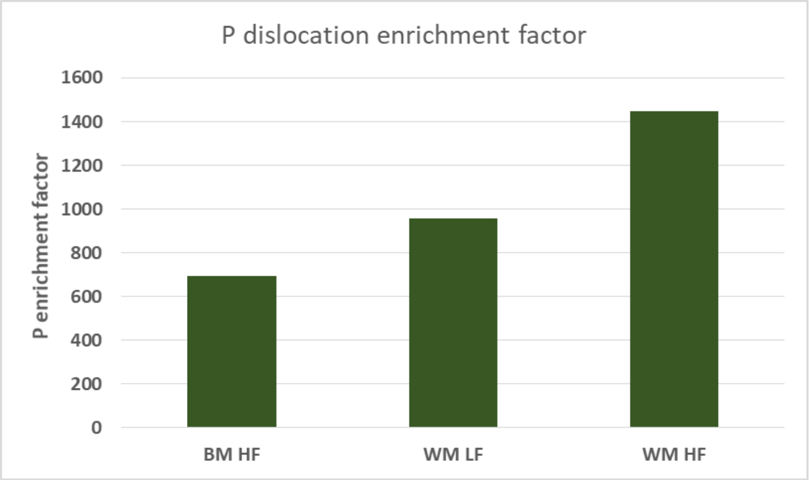
weld metal (HF)



- Density of dislocations ~ 10^{14} m^{-2}
- + Cluster at dislocations (BM HF, WM LF)

No dislocation observed in BM LF





BM HF WM LF WM HF

Results: APT

Findings:

- Solute atom clusters and segregated dislocations observed in all materials.
- Segregation along dislocation 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 higher for high flux (base metal) and in the same range for low and high flux (weld metal)

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.

Positron-Annihilation Lifetime measurement using Electron Bremsstrahlung

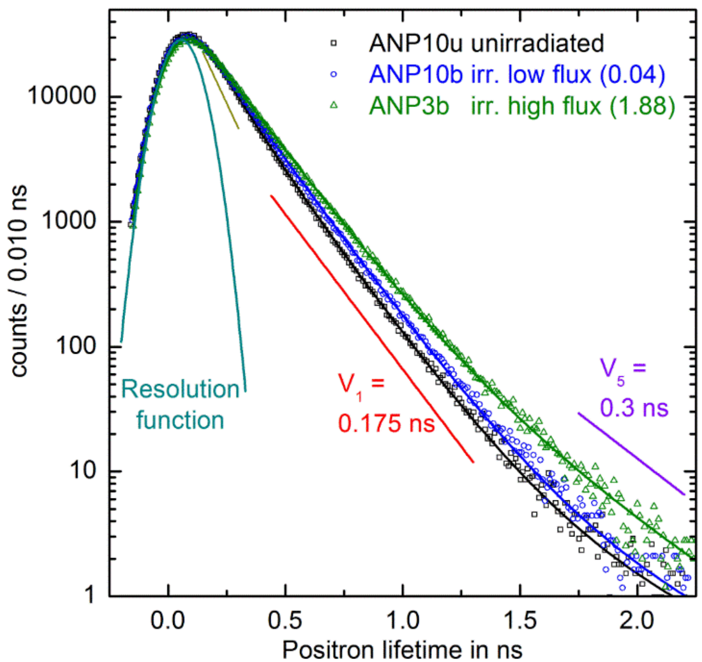
Principle:
pulsed γ-beam (pulse length 10 ps, repetition rate 13 MHz) induced electron-positron pair formation in the sample, positrons are used for LT

Advantages:
- using of extended bulk samples (~1 mm thickness)
- reduction of background and surface effects
- suited for radioactive samples in particular for ^{60}Co activities above 10^6 Bq.

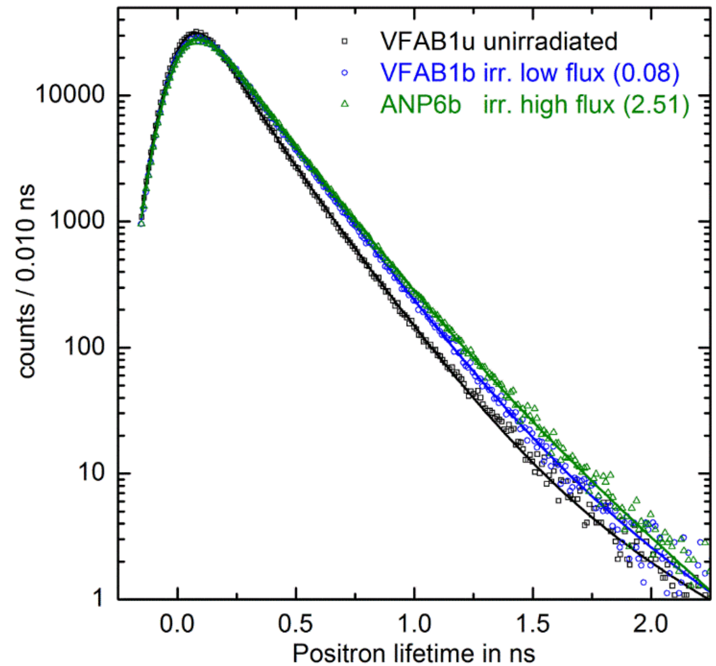
Results: PAS

Lifetime spectra of

Base metal



Weld metal



- The high-quality and high-resolution lifetime spectra allow three lifetime components to be identified.

Results: PAS

Material condition	LT 1 (108 ps)	LT 2 (175 ps)	LT 3	Mean LT
ANP-10 unirr.	45.02 %	54.66 %	0.32 % (492 ps)	146 ps
ANP-10 (BM-LF)	**22.49 %**	**77.07 %**	**0.44 % (462 ps)**	**161 ps**
ANP-3 (BM-HF)	**3.82 %**	**92.35 %**	**3.83 % (347 ps)**	**179 ps**
VFAB-1 unirr.	38.76 %	60.64 %	0.60 % (445ps)	151 ps
VFAB-1 (WM-LF)	**0 %**	**98.56 %**	**1.44 % (374 ps)**	**178 ps**
ANP-6 (WM-HF)	**0 %**	**92.08 %**	**7.92 % (282 ps)**	**183 ps**

- LT 1: annihilation in bcc Fe lattice
- LT 2: annihilation in single vacancies
- LT 3: annihilation in vacancy clusters (<10 vacancies)

Results: PAS

Findings:

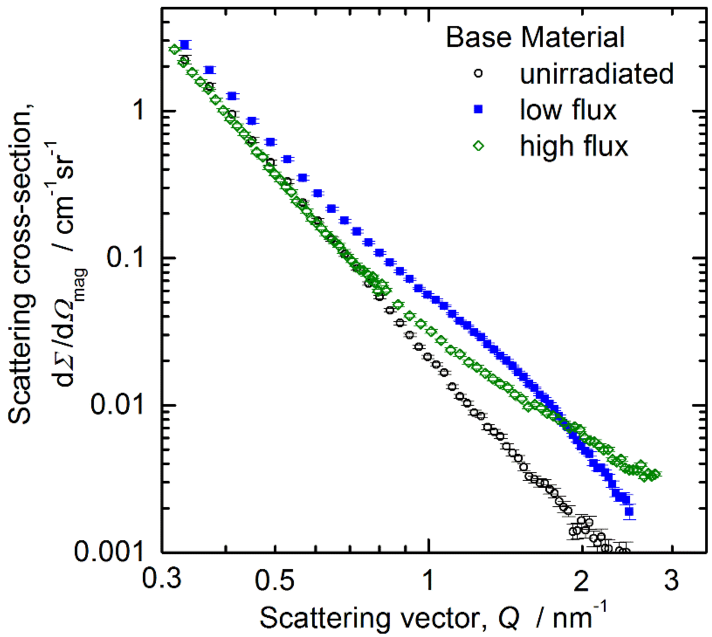
- Annihilation in single vacancies is dominant.
- Base metal: single vacancy concentration increases with flux.
- Weld metal: saturation effect (no annihilation in Fe matrix)
- More vacancy clusters for the higher flux.

Results: SANS

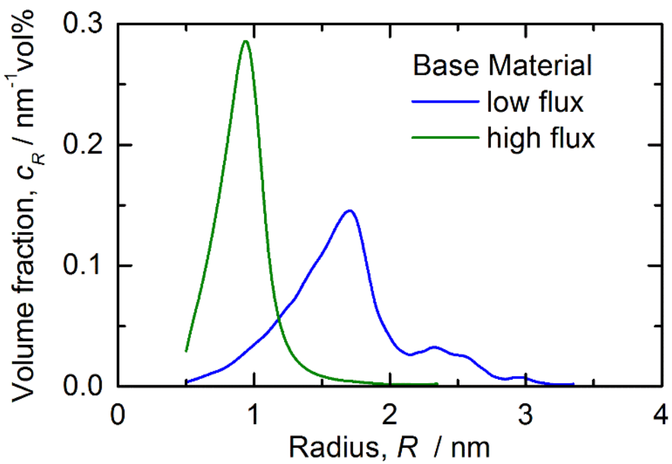
- SANS was performed by HZDR at beamline V4 of HZB Berlin.
- SANS provides macroscopically representative and statistically reliable averages of size and volume fraction of solute atom clusters.

Base metal

Scattering intensities



Size distributions

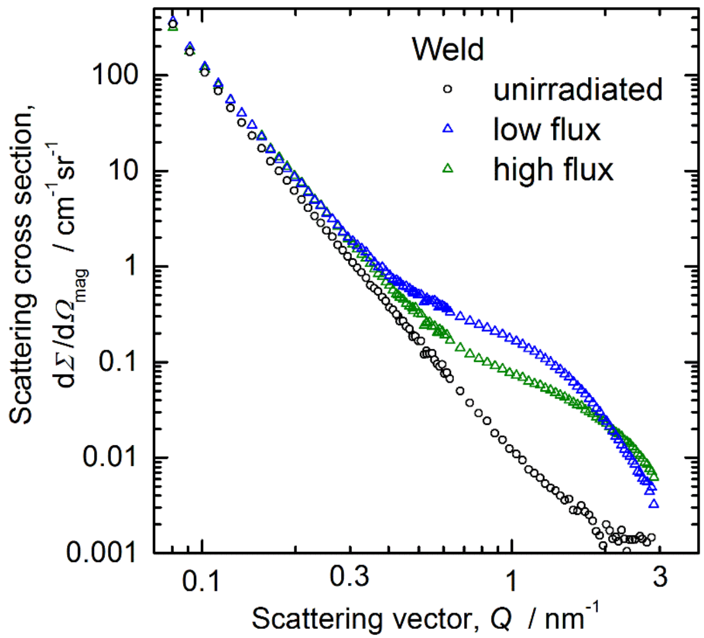


- A clear effect of decreasing mean radius of clusters with increasing flux

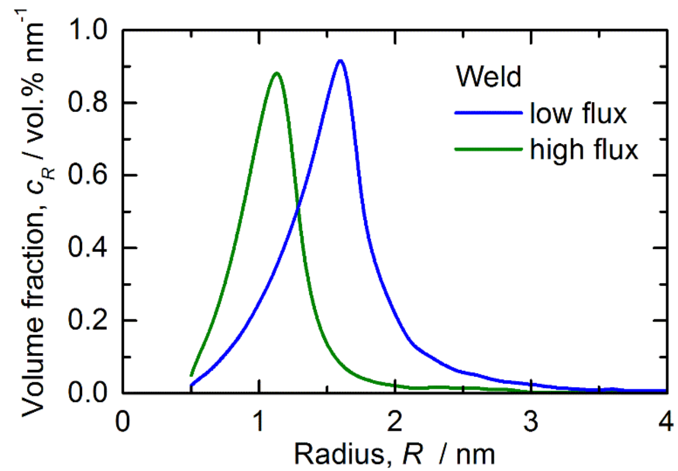
Results: SANS

Weld metal

Scattering intensities



Size distributions

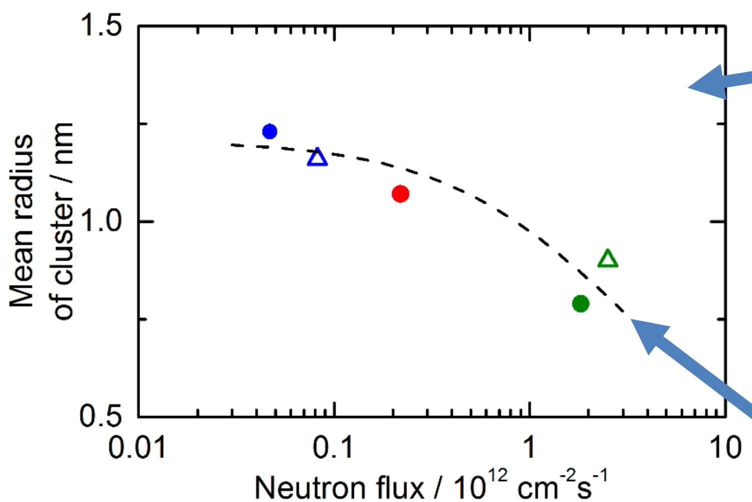


- A clear effect of decreasing mean radius of clusters with increasing flux

Results: SANS

❑ Summary of measured characteristics

Material condition	c in vol%	*N* in 10^{16} cm^{-3}	$\bar{R}$ in nm	A-ratio
ANP-10 (BM-LF)	0.120±0.014	11 ± 0.5	1.23±0.03	2.5±0.1
ANP-3 (BM-HF)	0.124±0.005	50 ± 4	0.79±0.02	2.6±0.1
VFAB-1 (WM-LF)	0.710±0.002	75 ± 3	1.16±0.02	2.2±0.1
ANP-6 (WM-HF)	0.534±0.016	130 ± 15	0.90±0.03	2.2±0.1



dots (BM); triangles (WM)

- Constant A-ratio for both flux condition indicates that mean cluster composition is similar.
- A-ratio clearly above 1.4 implies that scattering features are not vacancy or vacancy-rich clusters.
- Model fit according to deterministic growth model. [Wagner et al., Acta Mater. 104 (2016) 131]

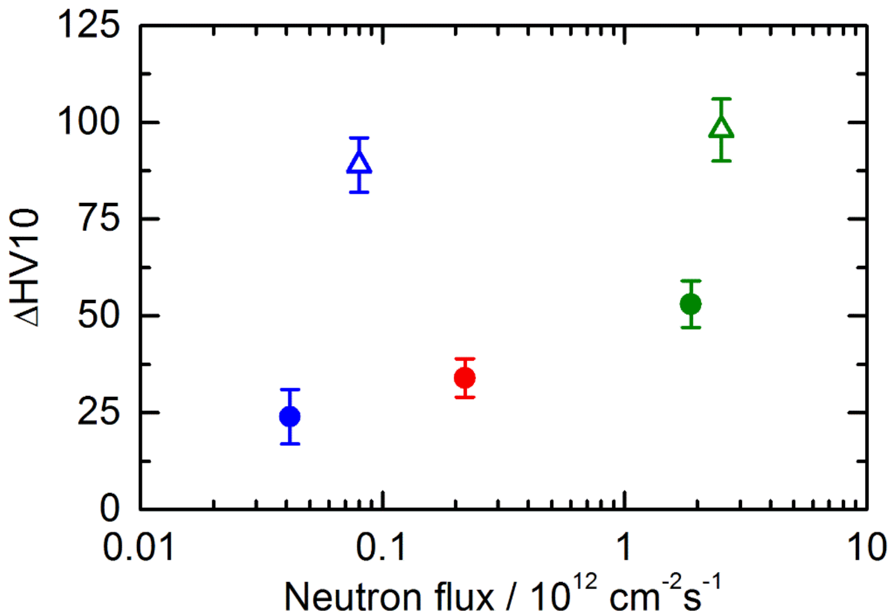
Results: SANS

Findings:

- Solute atom clusters observed in all irradiated conditions.
  - Radius 0.8 – 1.2 nm
  - Number density $10^{23} – 10^{24}$ m^{-3}
- Cluster are larger for low flux condition.
- Number density of clusters increases with flux.
- Volume fraction is independent of flux (base metal) or slightly larger for low flux (weld metal)

Results: Vickers hardness

- Irradiation-induced Vickers hardness increase ΔHV10 vs. neutron flux for RPV base and weld metal:

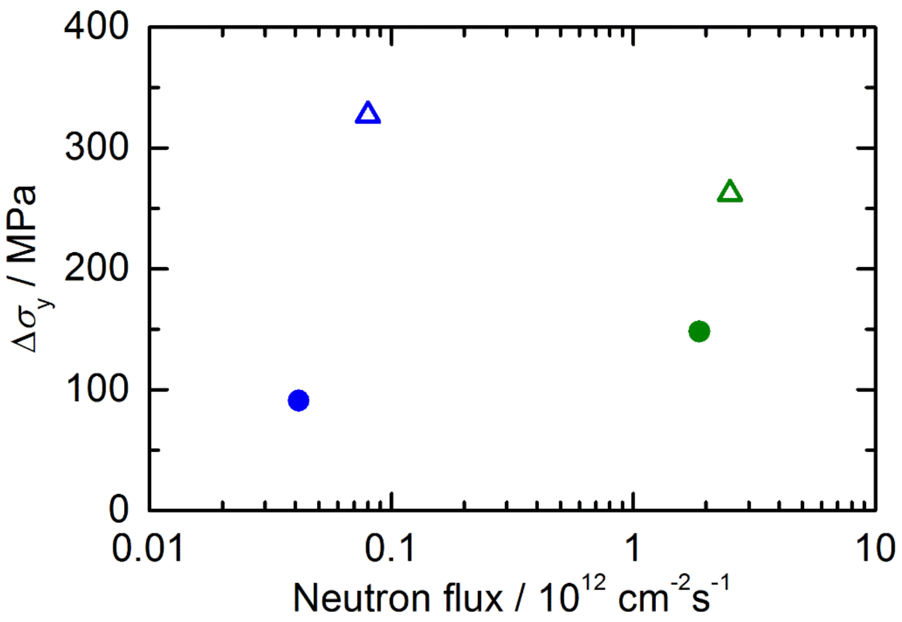


dots (BM); triangles (WM)

- base metal:
  ΔHV10 increases with increasing flux
- weld metal:
  ΔHV10 seems to be independent to flux

Results: Yield stress

- Irradiation-induced yield stress increase $\Delta\sigma_y$ vs. neutron flux for RPV base and weld metal:



dots (BM); triangles (WM)

- Opposing trends of yield stress increase with neutron flux for base and weld metal

Conclusions

- Using TEM, APT, PAS and SANS, different kinds of irradiation-induced nanofeatures were detected:
  - Dislocation loops,
  - Segregation to dislocations,
  - Vacancies and sub-nm vacancy clusters,
  - Solute atom clusters of different composition.
- An increase of neutron flux tends to produce:
  - Higher vacancy concentrations,
  - Less loops (base metal),
  - More, smaller and more diluted clusters.
- Based on the observed opposing trends of size *d* and number density *N* of solute atom clusters as functions of flux, the hardness increase and the yield stress increase ($\sim\sqrt{Nd}$) can be insensitive to flux (weld metal).

The SOTERIA Consortium

SOTERIA

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Effect on neutron energy spectrum

- Analysis of existing databases (IAEA CRP) show that some effects of neutron spectra on trend curves of TT shifts exists. It depends on energy threshold used for the referred fluence.
- Values with E_n > 1 MeV seems to be non-sensitive to this effect while fluences with E_n > 0.5 MeV as well as the parameter dpa show some non-negligible effects.
- TT shifts in out of core positions were generally lower than after irradiation in the core.
- TT shifts of VVER welds in surveillance position were lower than in experimental reactors.
- Irradiation in positions with larger ratio $\Phi_{0.5MeV}/\Phi_{1MeV}$ caused smaller TT shifts.

APT on ion-irradiated binary model alloys

- The flux effect in terms of radiation-induced segregation phenomenon in Fe-ion irradiated model alloys of Fe-3Mn and Fe-3Ni were studied by APT at CEA.
- Irradiation conditions: 2 MeV Fe^{3+} and 27 MeV Fe^{9+} ions with fluxes of 1.5×10^{12} and 1.5×10^{11} $cm^{-2}s^{-1}$ up to fluences of 3.8×10^{15} and 2.5×10^{16} cm^{-2}.
- APT analysis show that decreasing flux induces larger, more concentrated but less numerous clusters.