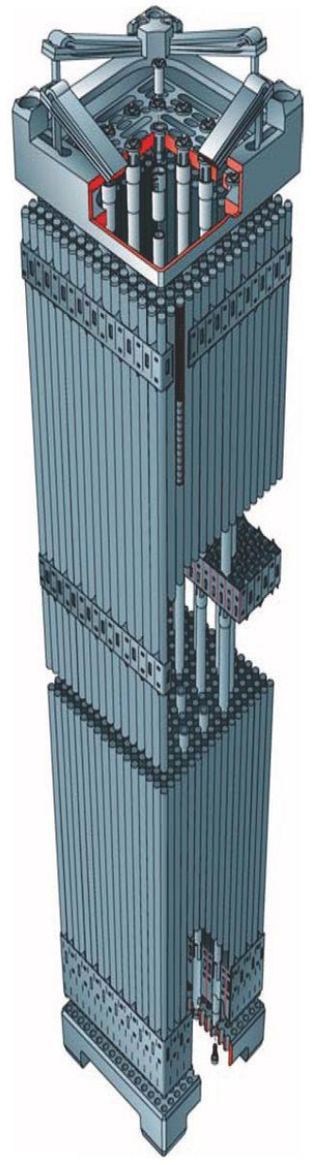
Investigating the Effects of Proton-Irradiation Damage in Zirconium Alloys

Matthew Topping



Acknowledgements:
Michael Preuss, Philipp Frankel, Allan Harte, Tom Seymore, Elisabeth Francis, Felicity Baxter, Tamas Ungar, Simon Pimblott and his team.

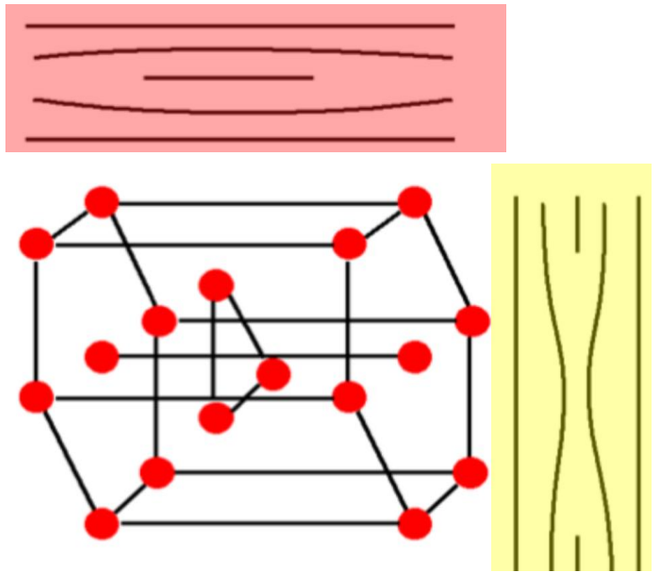
Background

- Zr-alloys used for encapsulating fuel and as structural materials in fuel assemblies for LWRs due to low neutron cross section and good corrosion performance under normal operating conditions.

- Main material degradation
  - Corrosion including hydrogen pick up impacting mechanical performance
  - *Irradiation induced/enhanced dimensional instability (growth, creep)*
  - Irradiation hardening
  - Pellet Cladding Interaction (PCI)

Irradiation Induced Growth

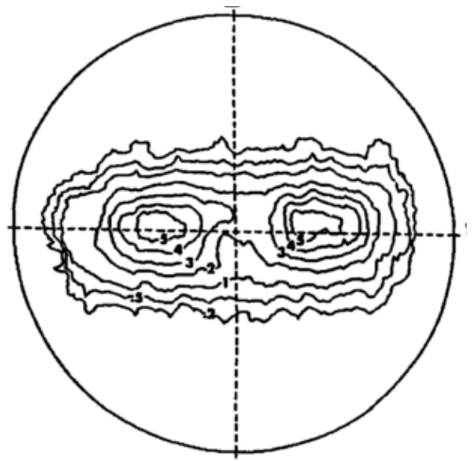
<a>-loops



<c>-loops

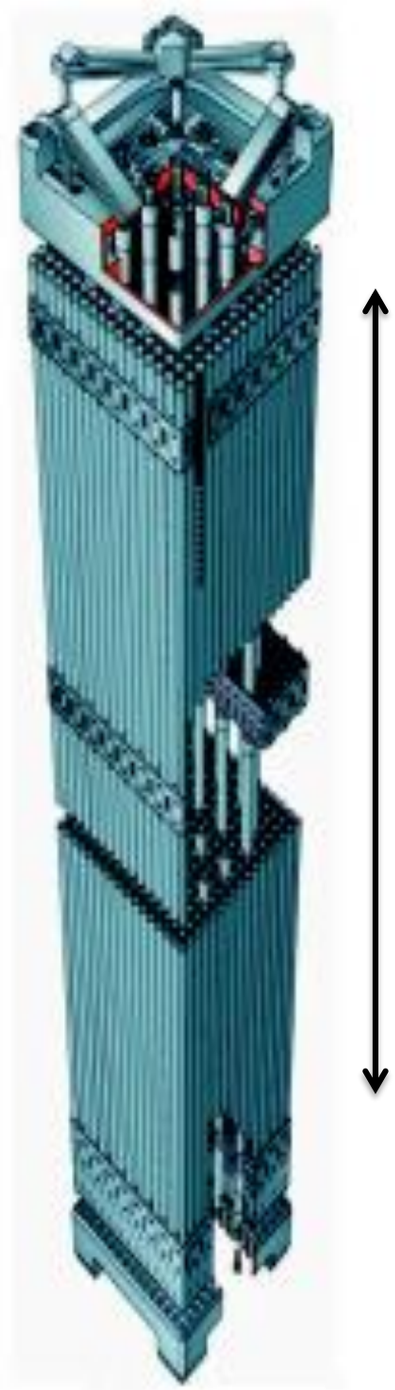
(0002) pole figure

Axial

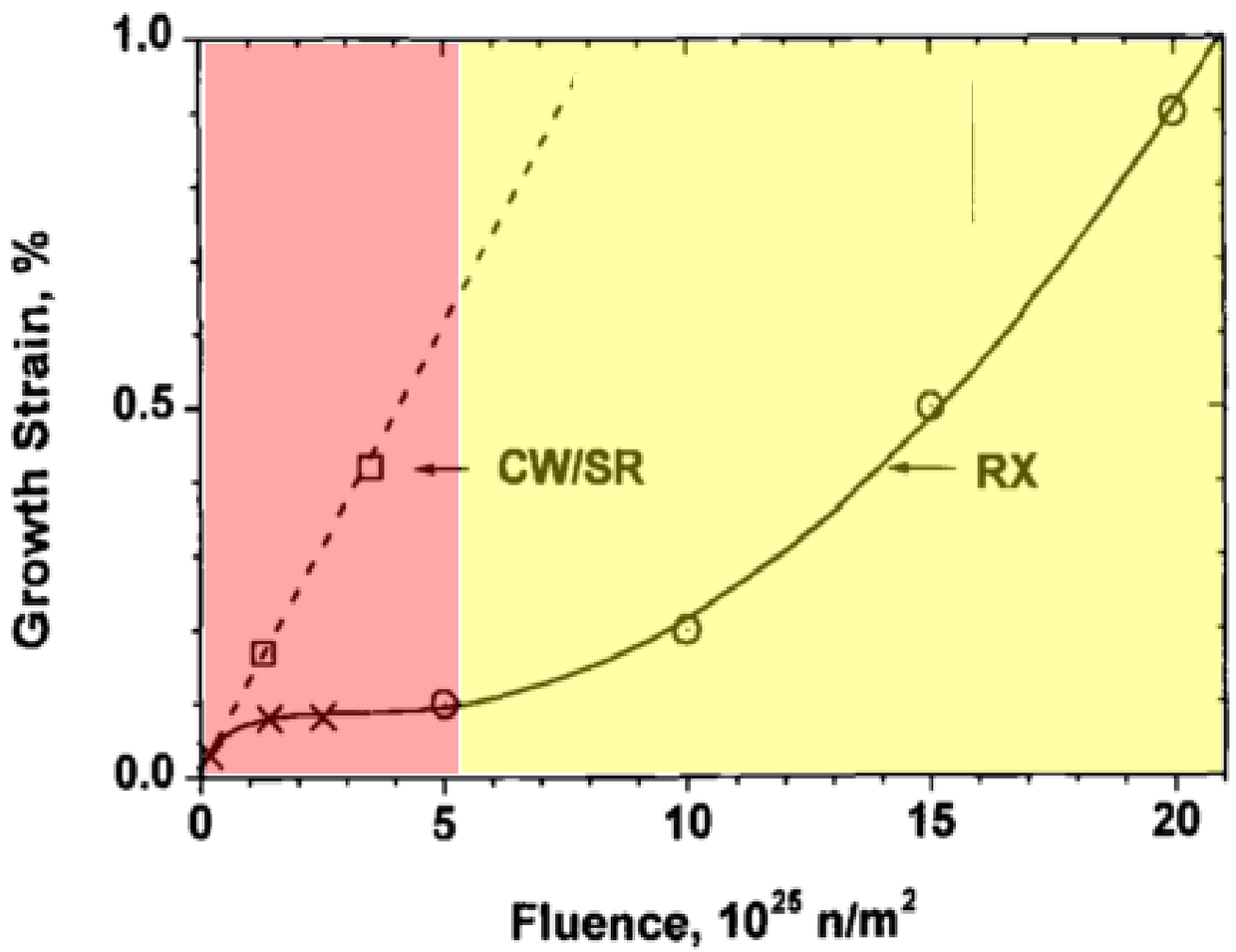


Hoop

Shrinkage in the radial direction

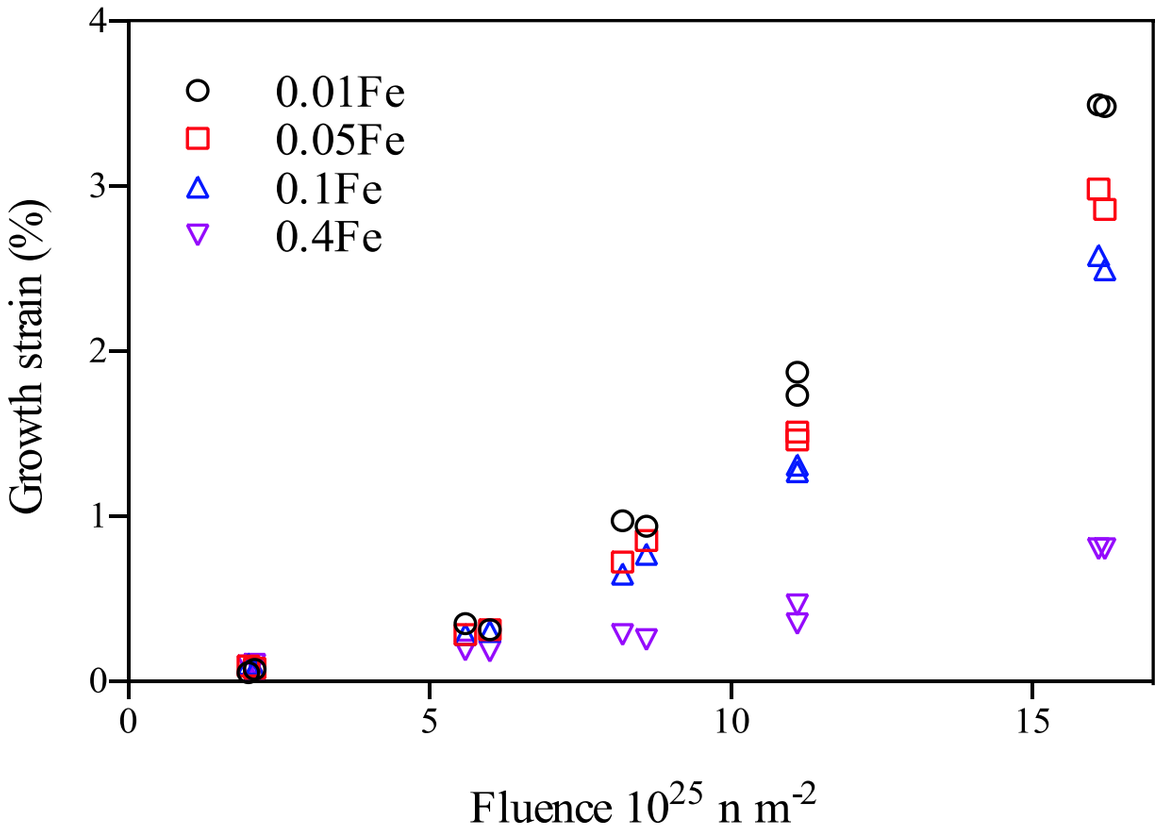


Elongation in the axial direction

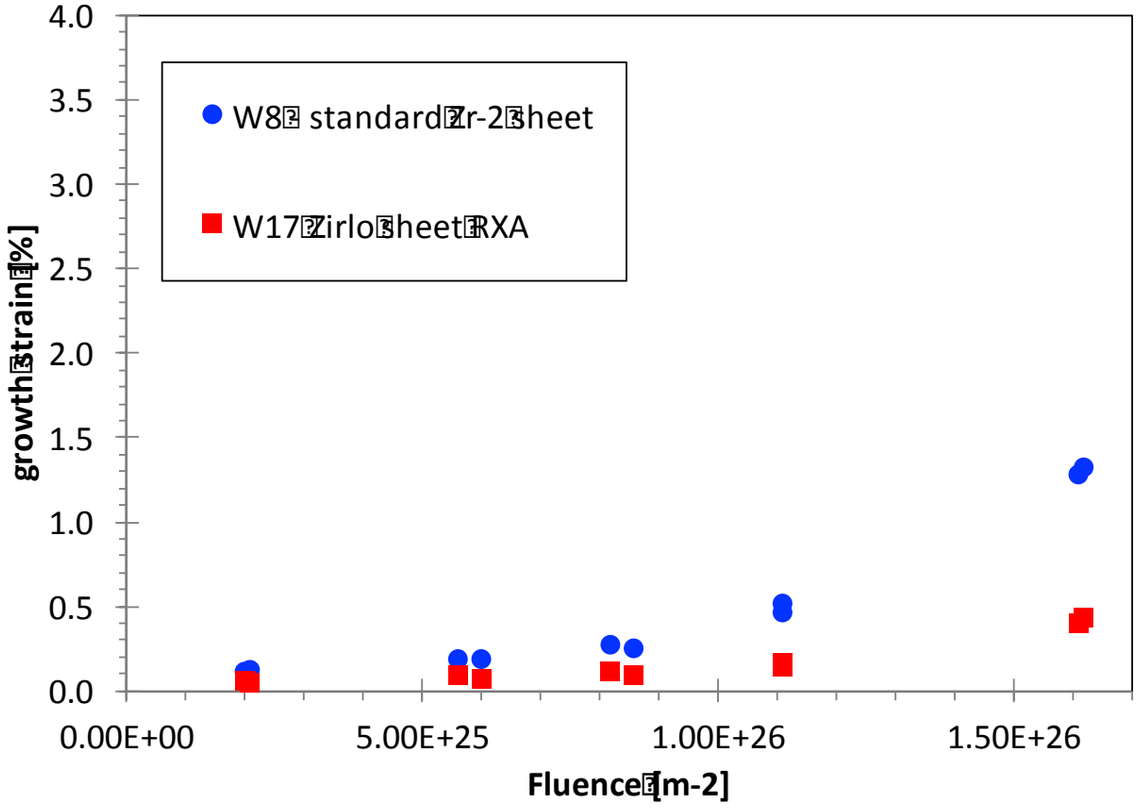


Effect of Alloy Chemistry on IIG

Effect of Fe
Zr-1Sn-XFe



Effect of Nb
ZIRLO vs Zircaloy-2



S. Yagnik, R.B. Adamson, G. Koblyansky, J.H. Chen, D. Gilbon, S. Ishimoto, et al., Zirconium in the Nuclear Industry: 18th International Symposium, 2016.

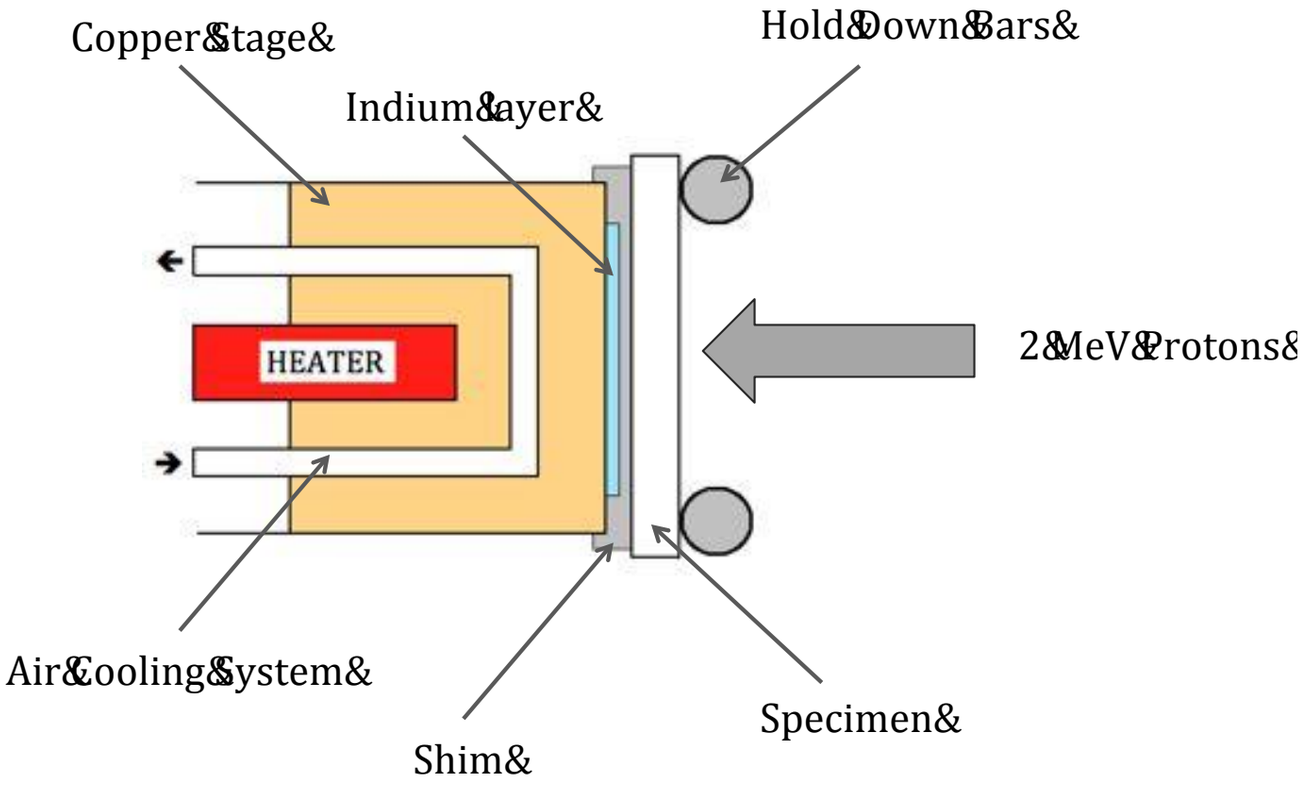
All samples irradiated at BOR-60

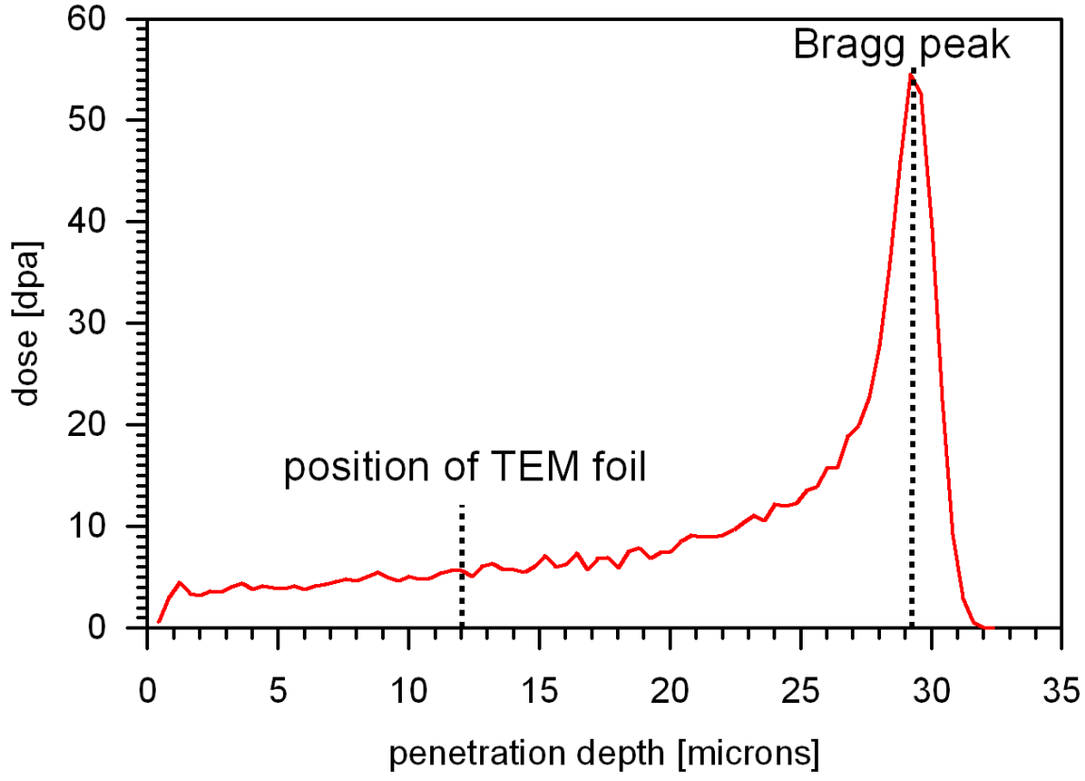
Proton Irradiation

DCF @ University of Manchester, UK

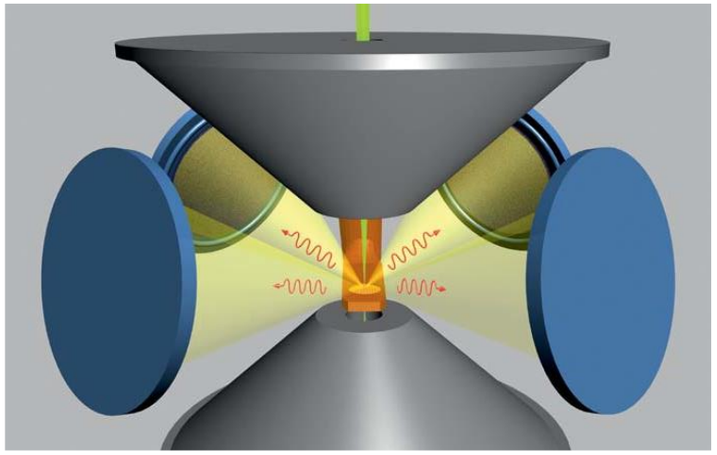
Typical irradiation parameters:

- 350 ±9 ° C
- 2 MeV ~0.2 μA mm^{-2}, 6.7 $x10^{-6}$ dpa s^{-1} (about 100 faster than in a commercial reactor)
- Up to 7 dpa
  - at 40% penetration depth
  - quick Kinchin-Pease calculation in SRIM



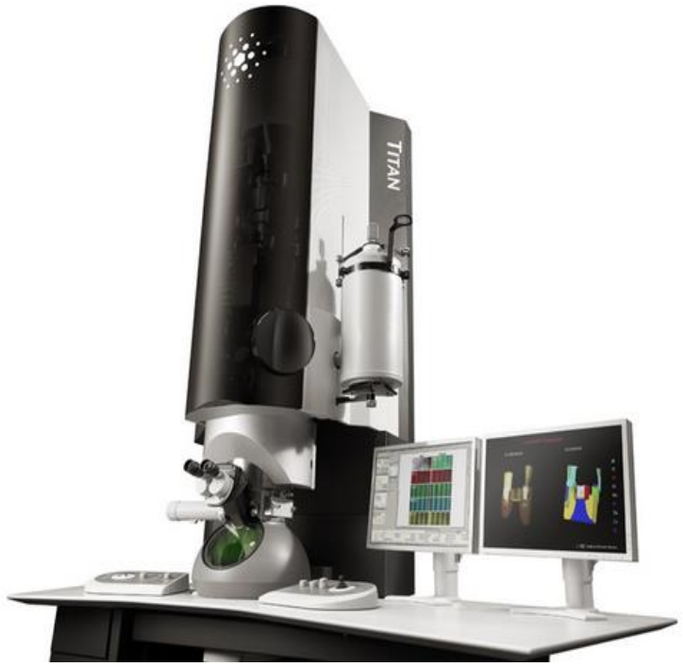


Analytical Electron Microscopy

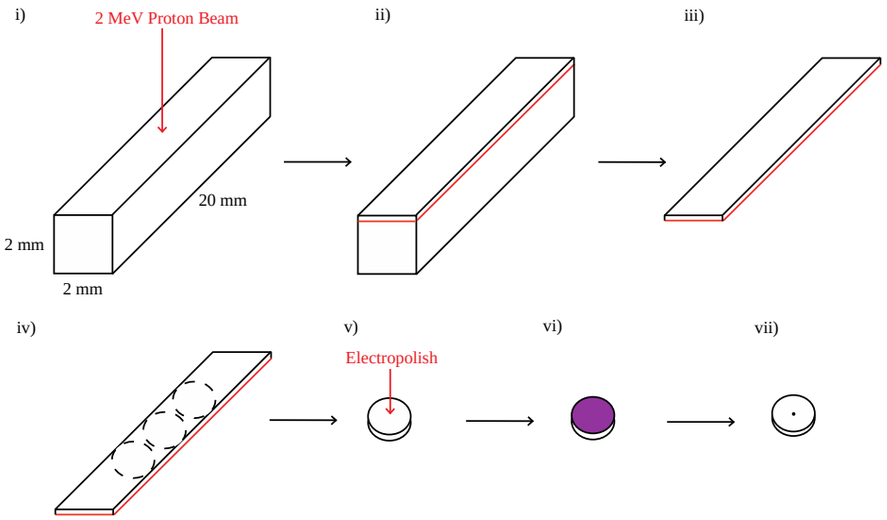


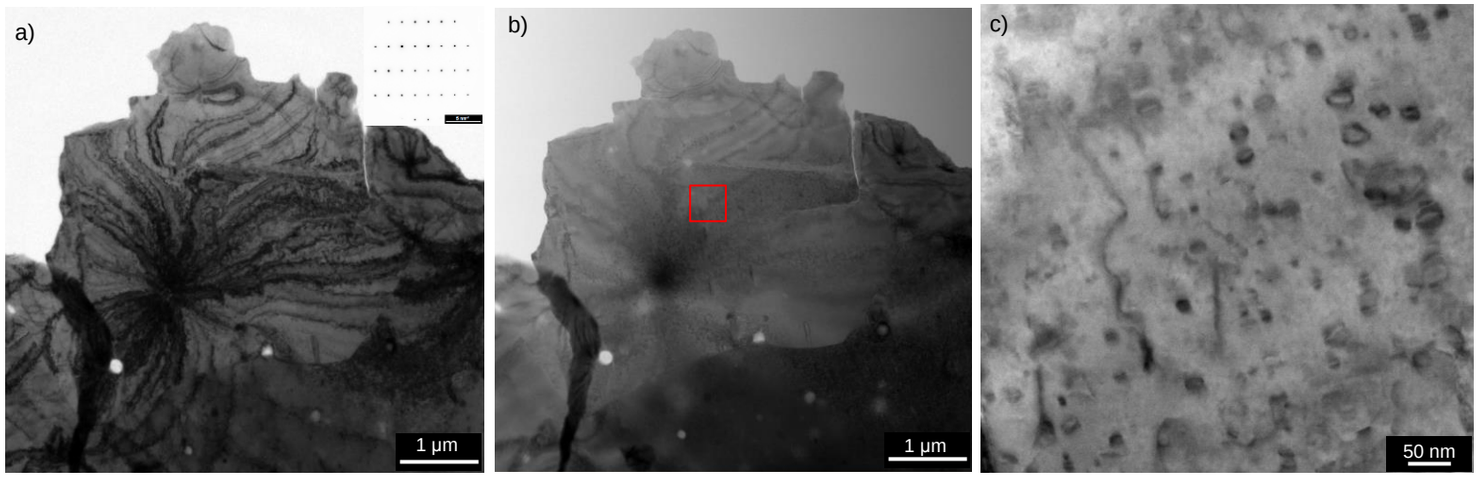
Chemical microanalysis by aberration-corrected scanning TEM:

- Bright electron source and C_s-correction
- Windowless energy dispersive X-ray detectors of a large collection angle



We avoid FIBing as it would be difficult to distinguish between proton irradiation damage and FIB damage.





Dislocation analysis by on-axis bright-field STEM

- Convergent scanning beam reduced matrix strain contrast
- Dislocations observed from perpendicular to their habit plane

Convolutional Multiple Whole Profile analysis

$$I_{measured} = I_{size} * I_{strain} * I_{fault} * I_{instrument}$$

Theoretical broadening for single peak:

$$A_L^D = \exp\left[-\rho\left(\frac{\pi b^2}{2}\right)L^2 f(\eta) g^2 \bar{C}\right]$$

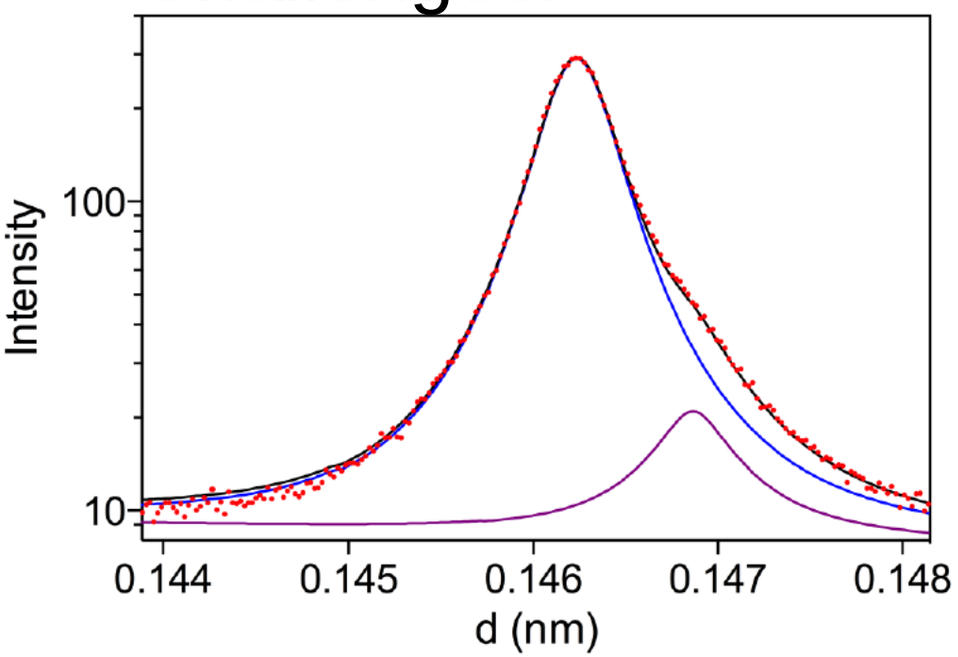
Average contrast factor for particular dislocation:

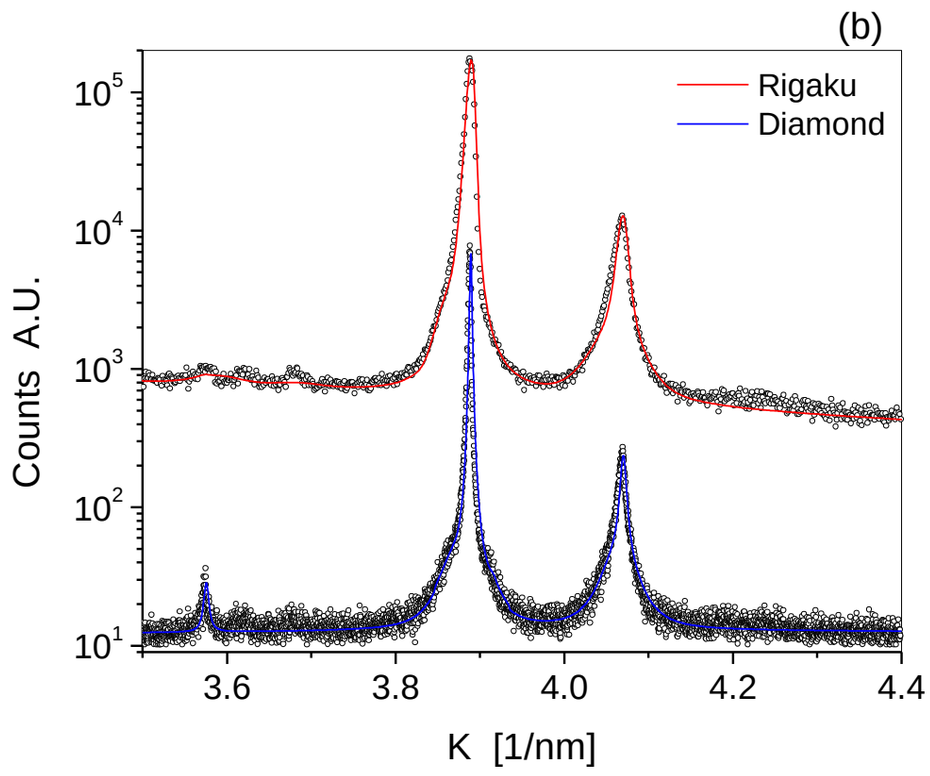
$$\bar{C}_{hkil} = \bar{C}_{hki0}(1 + q_1 x + q_2 x^2)$$

q1 and q2 have been calculated for the different loop types treating the loop as a polygon with N sides, where N is a large number

Equipment	Crystallite size [nm]	Dislocation density [10^{14} m^{-2}]	Dipole character parameter: $M=R_e\sqrt{\rho}$	Contrast parameters: a_1, a_2
Rigaku	300	99	0.49	-0.28/-0.31
Diamond	>> 1 μm	85	0.44	-0.25/-0.3

Challenges:





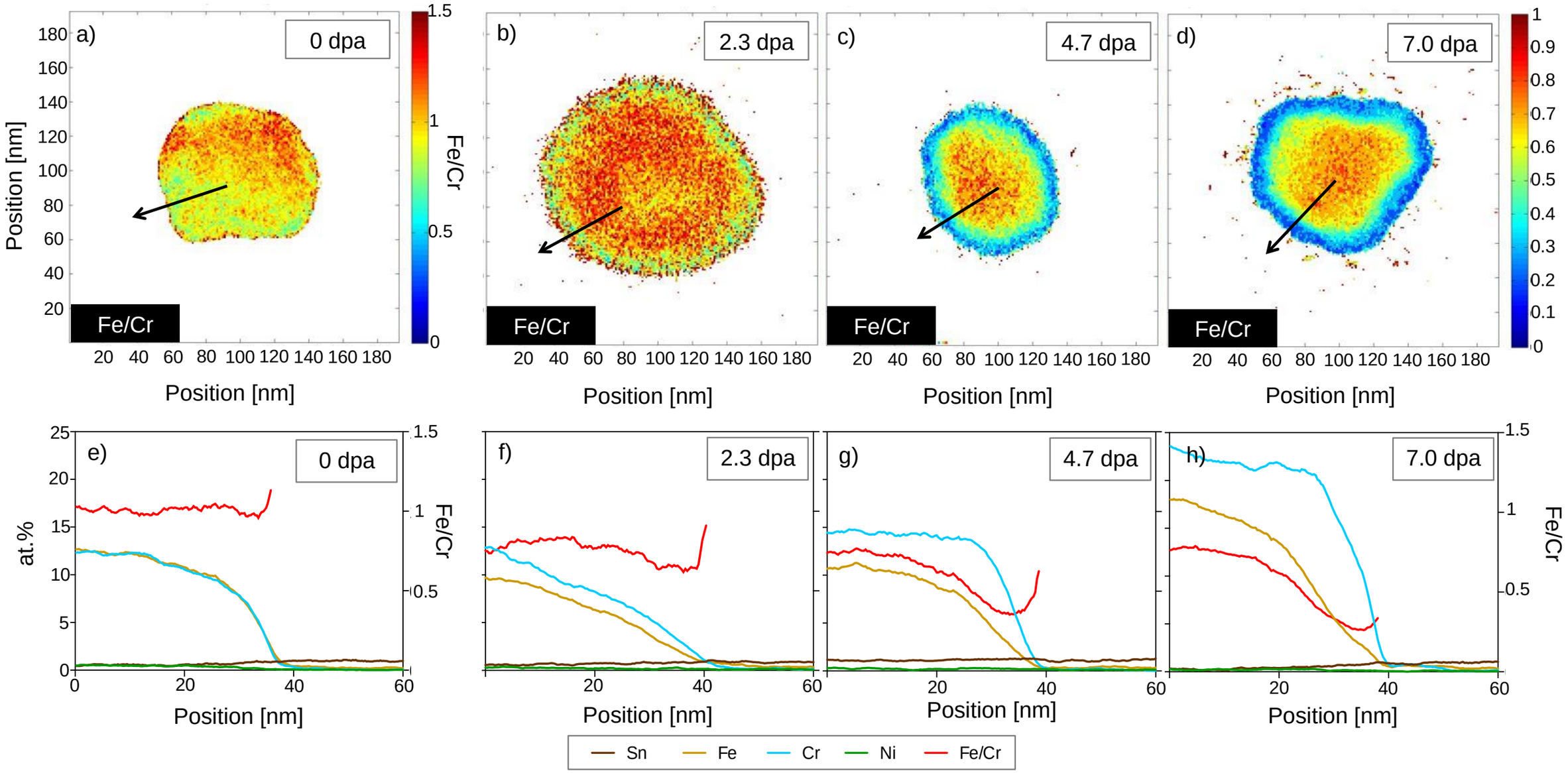
Second Phase Particle and their evolution during Irradiation

Question 1: Does proton irradiation create the same leaching out of particularly Fe as observed for neutron irradiated Zr alloys?

Second Phase Particle Evolution

Zircaloy-2: common SPPs are $Zr(Fe,Cr)_2$ and $Zr_2(Fe,Ni)$

$Zr(Fe,Cr)_2$

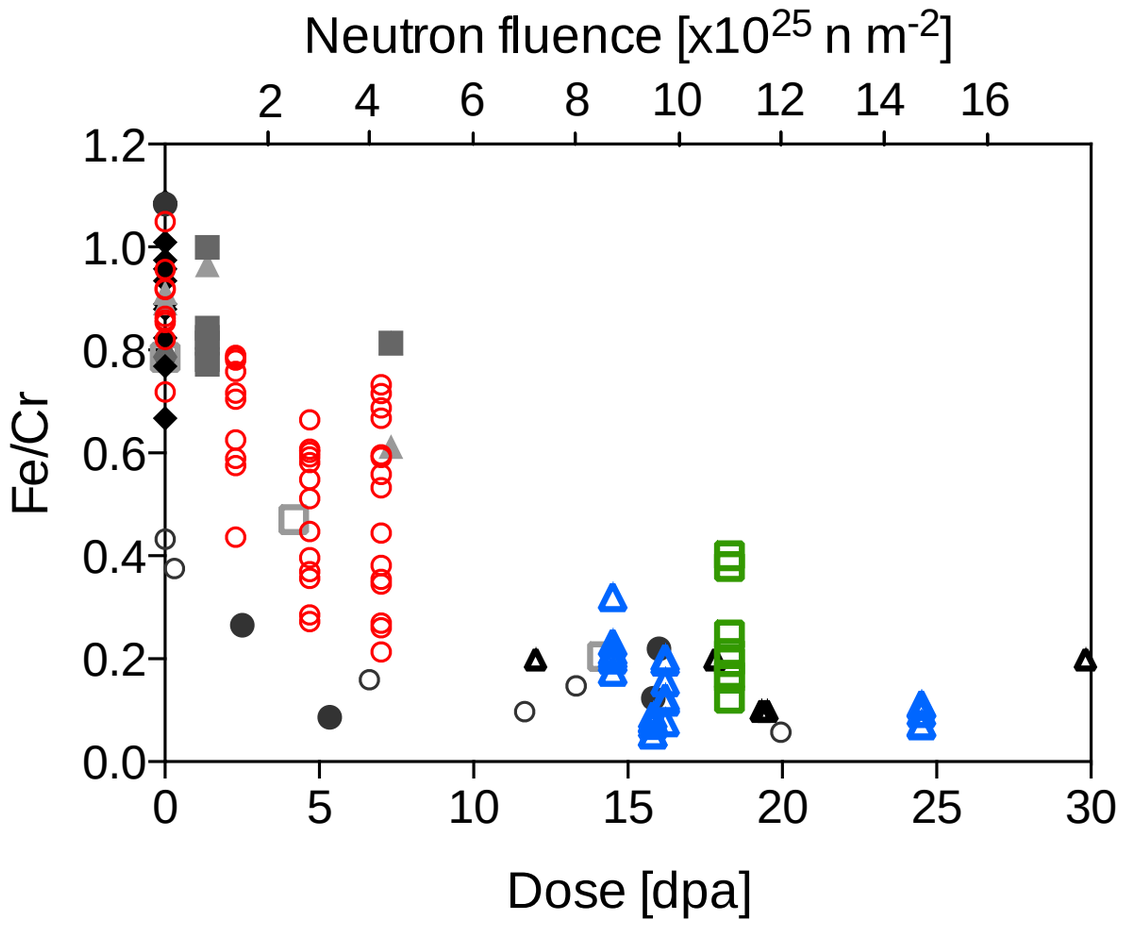


Fe leaches out more quickly than Cr and Ni in $Zr(Fe,Cr)_2$ and $Zr_2(Fe,Ni)$

Comparing proton and neutron irradiation

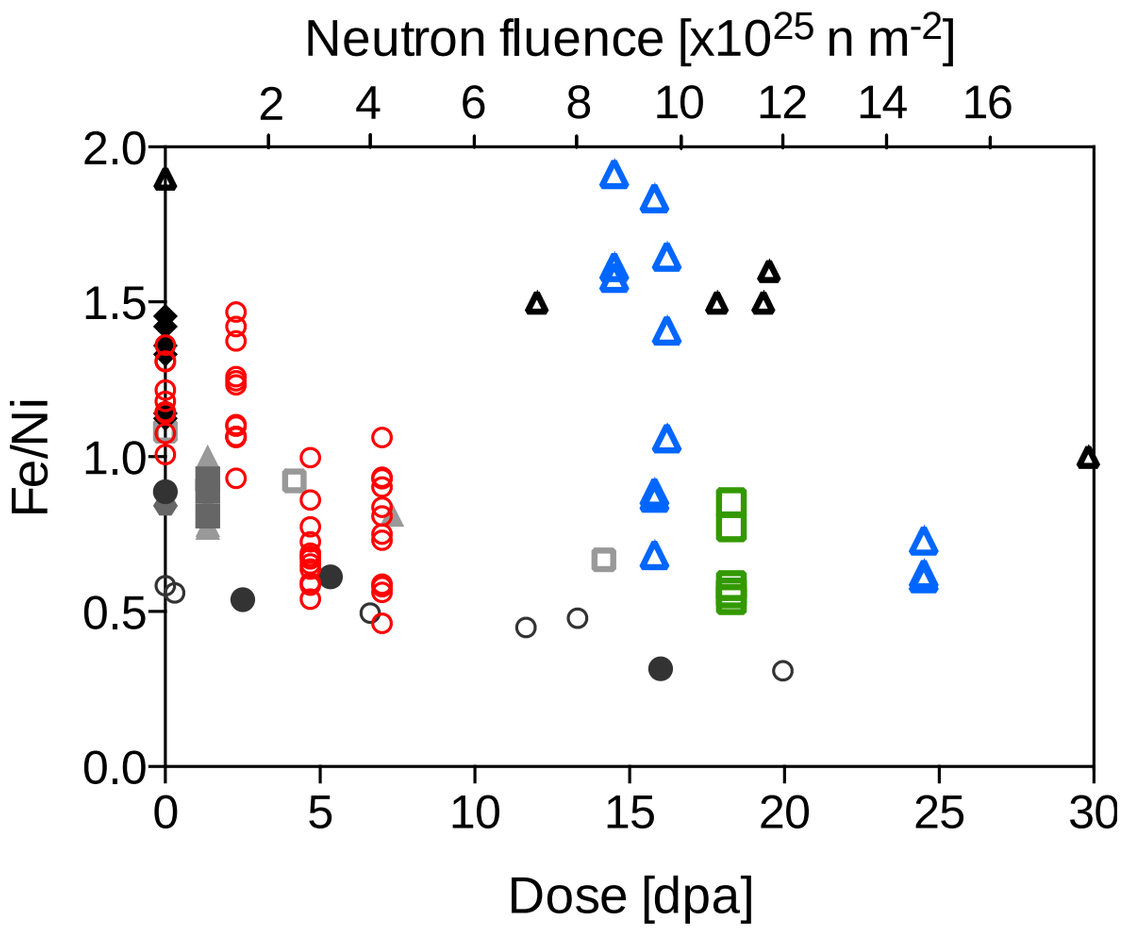
$Zr(Fe,Cr)_2$

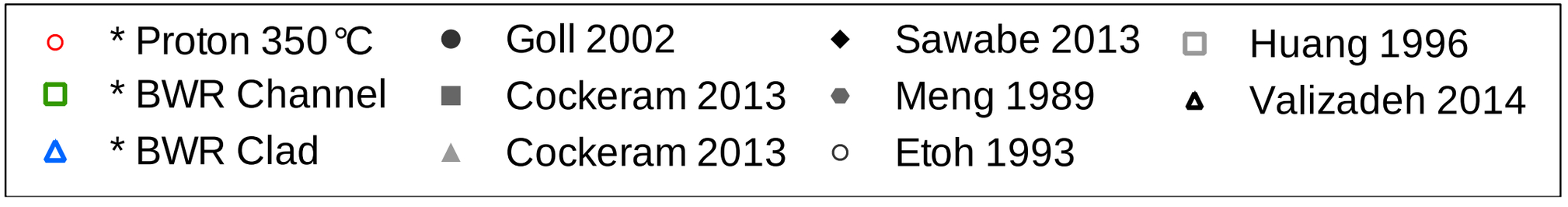
a) Zircaloy-2 SPP Fe/Cr Evolution



$Zr_2(Fe,Ni)$

b) Zircaloy-2 SPP Fe/Ni Evolution





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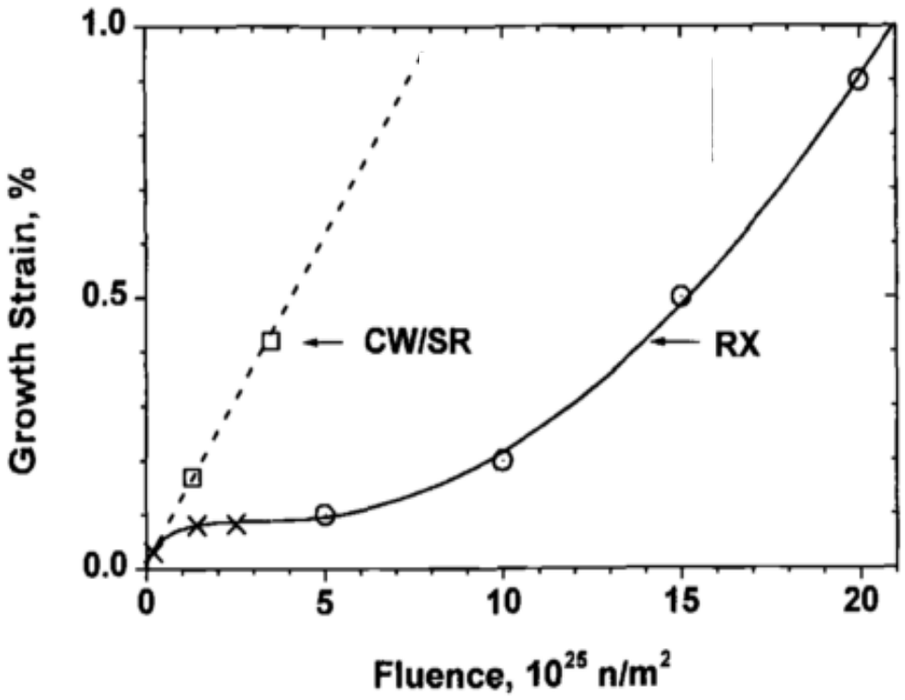
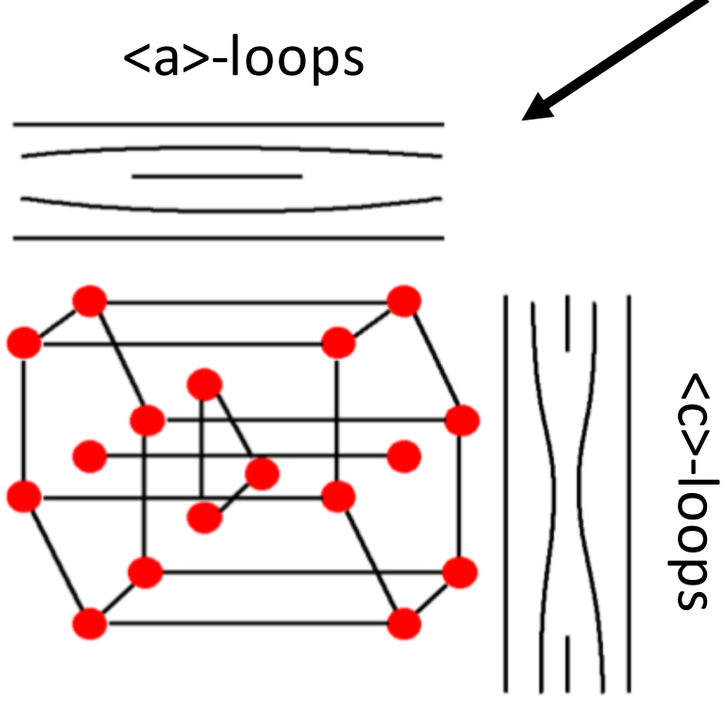
Dislocation loop analysis

Question: What might affect c-loop formation



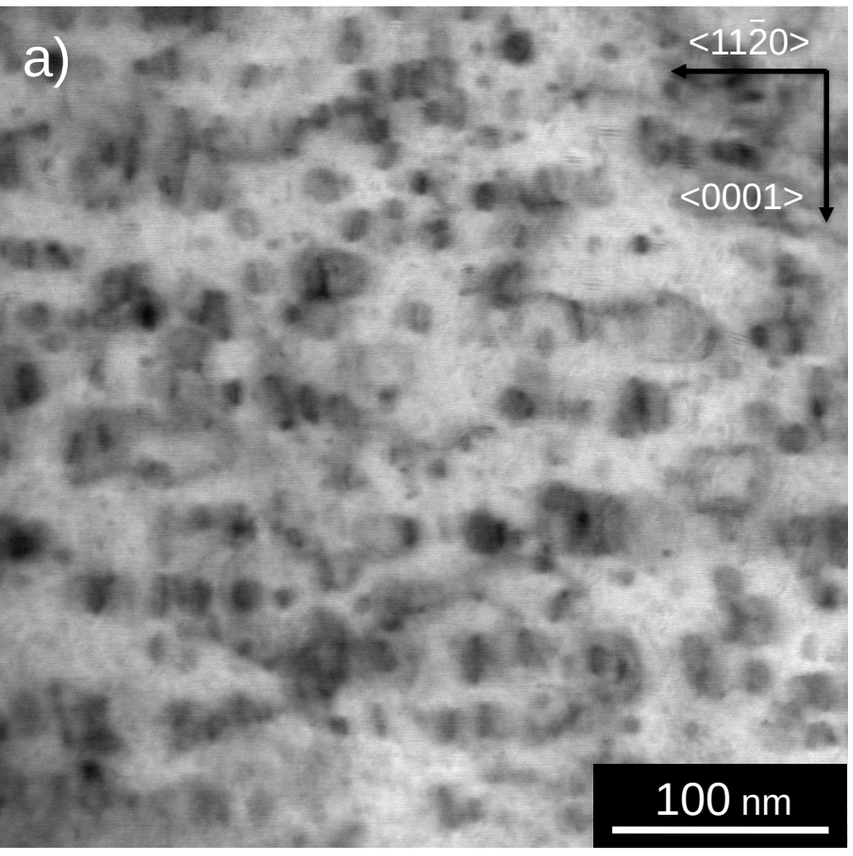


only vacancy loops

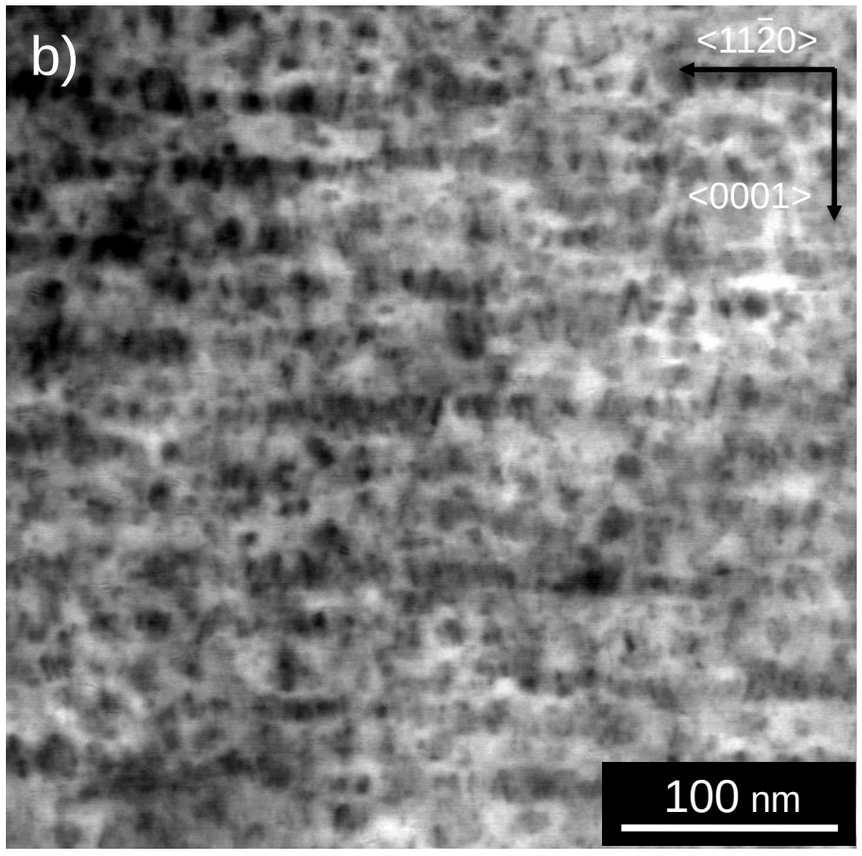
Proton-irradiated Zr-2

Evolution of a-loops

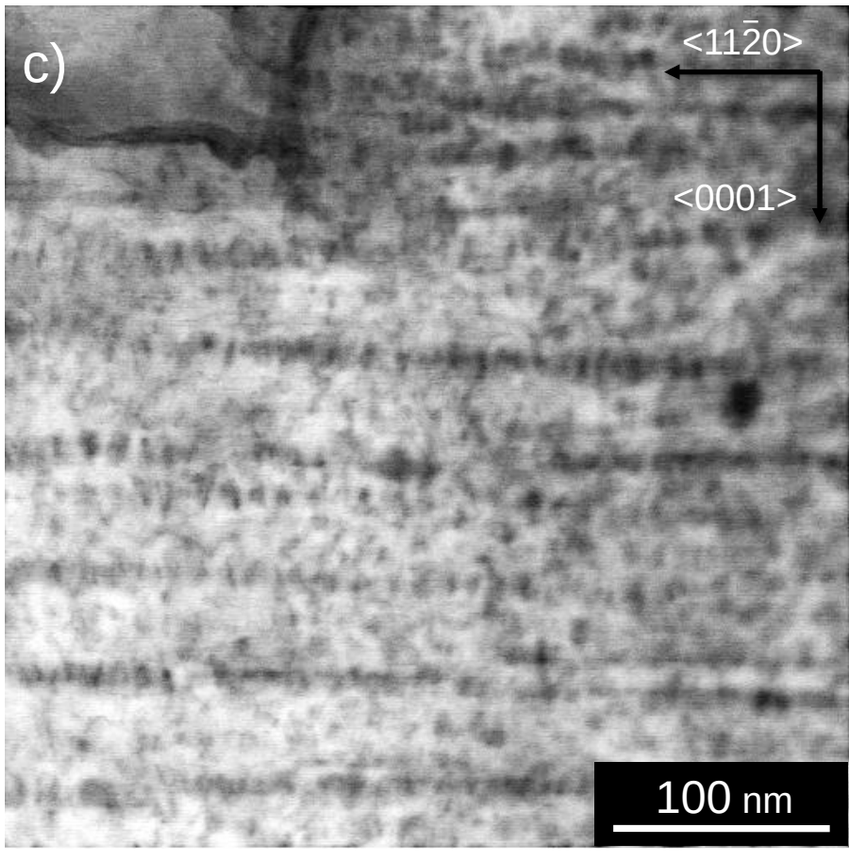
2.3 dpa



4.7 dpa



7.0 dpa

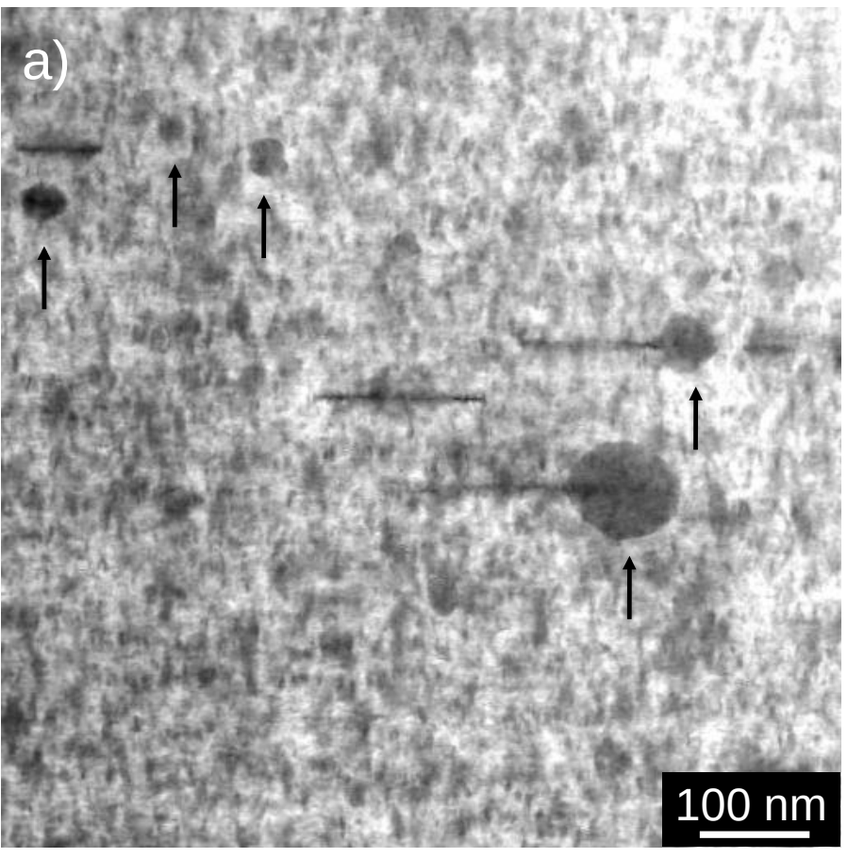


a-loops become smaller and align parallel to the basal plane with spacing in <0001> ~50 nm

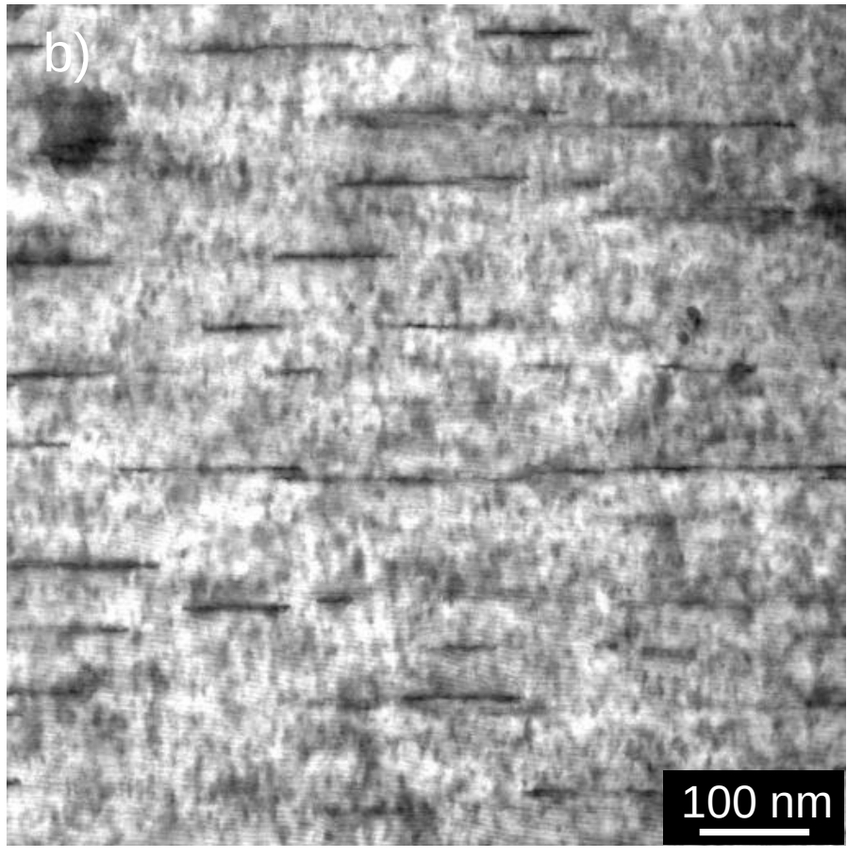
Proton-irradiated Zr-2

Evolution of c-loops

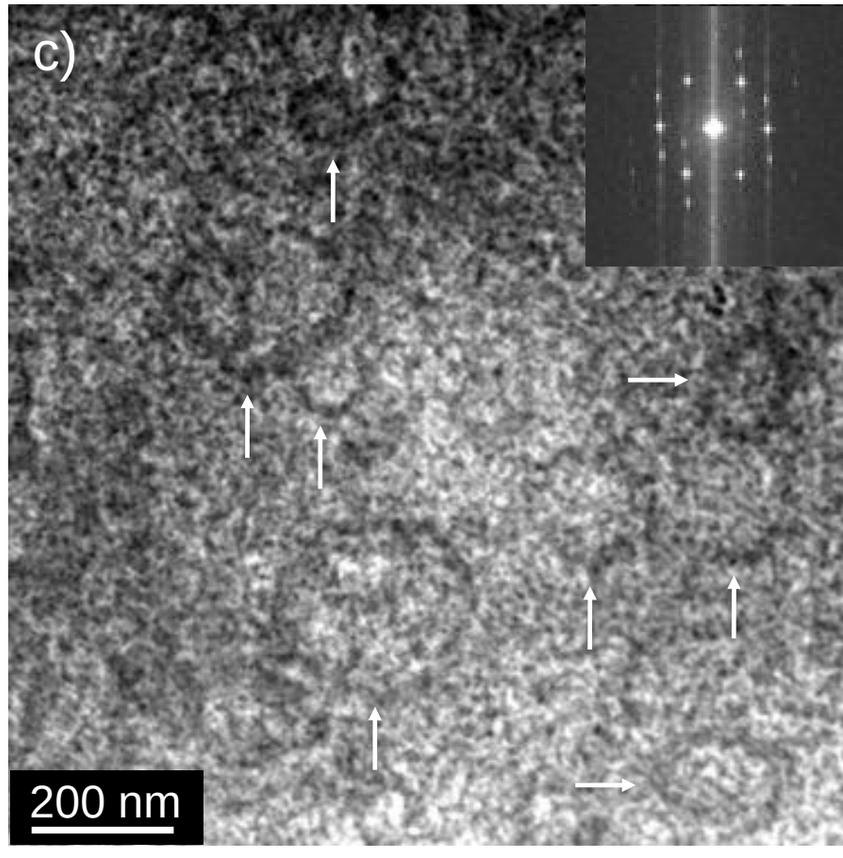
4.7 dpa



7.0 dpa



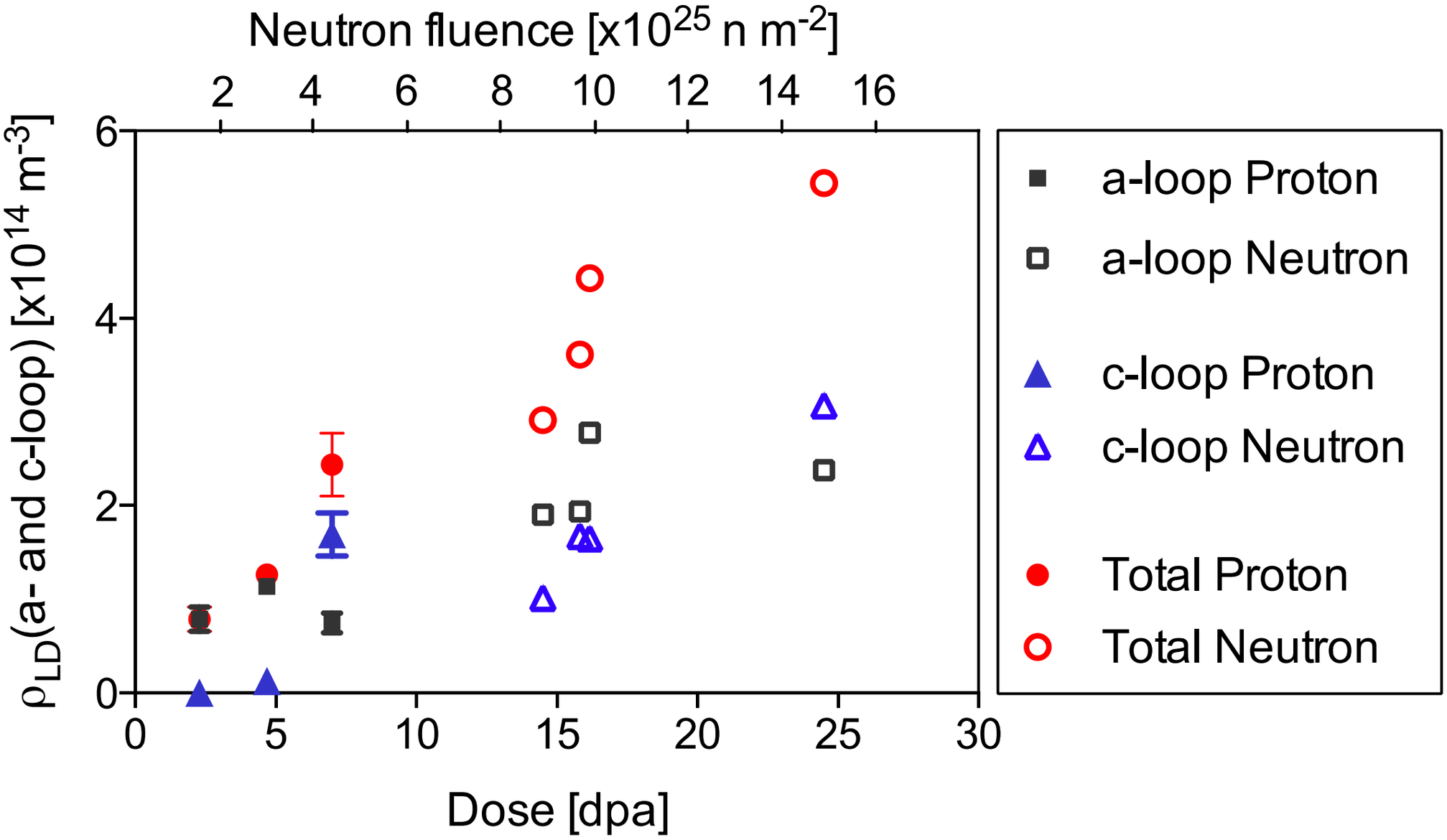
7.0 dpa



- c-loops align parallel to the basal plane with spacing in <0001> ~50 nm
- c-loops are observed from the <0001> direction as circular with a large diameter (~160 nm)

Comparing proton and neutron irradiation

Dislocation line density = loop number density * loop line length



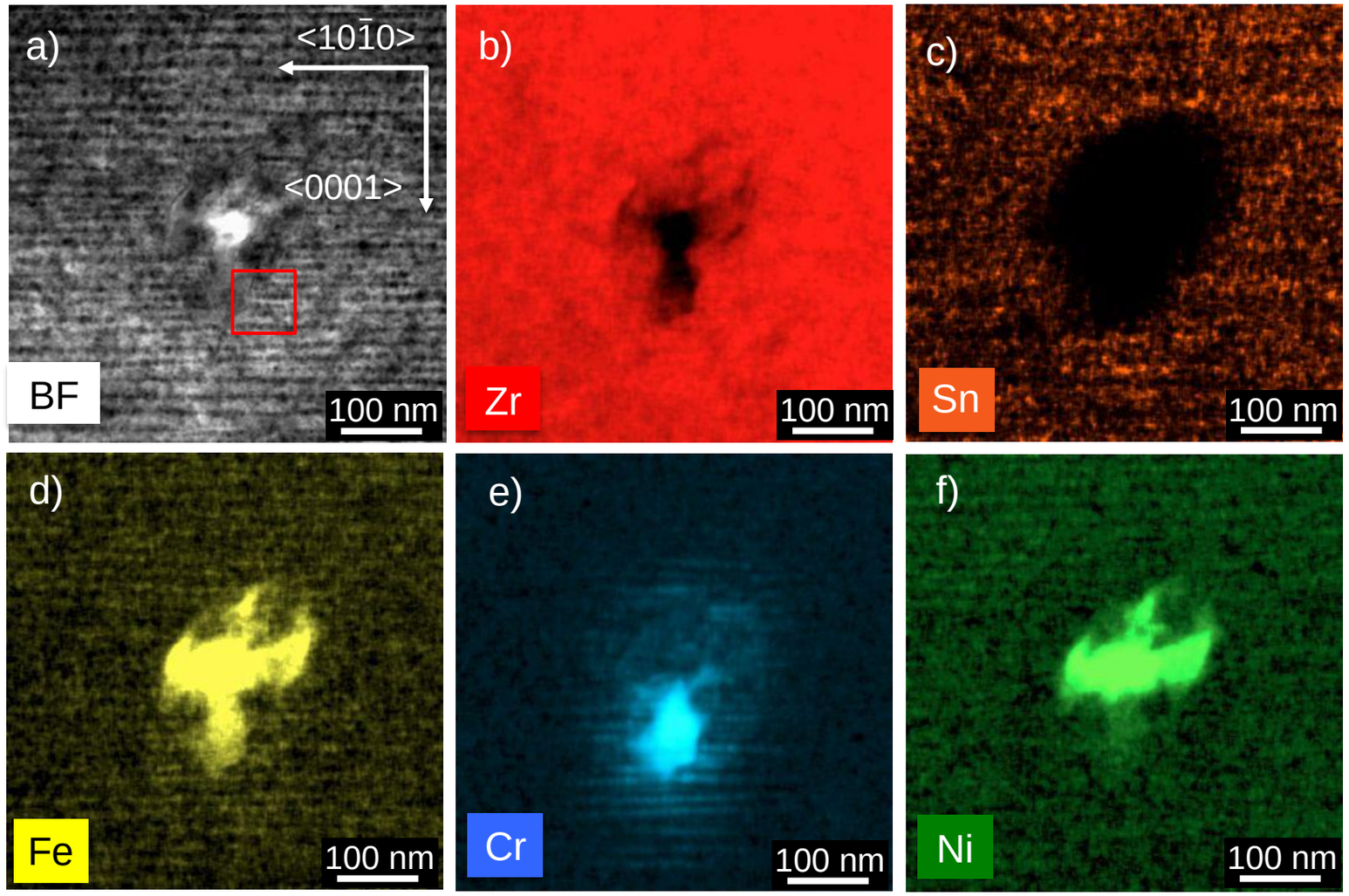
Reasonable trends when comparing proton and neutron irradiation

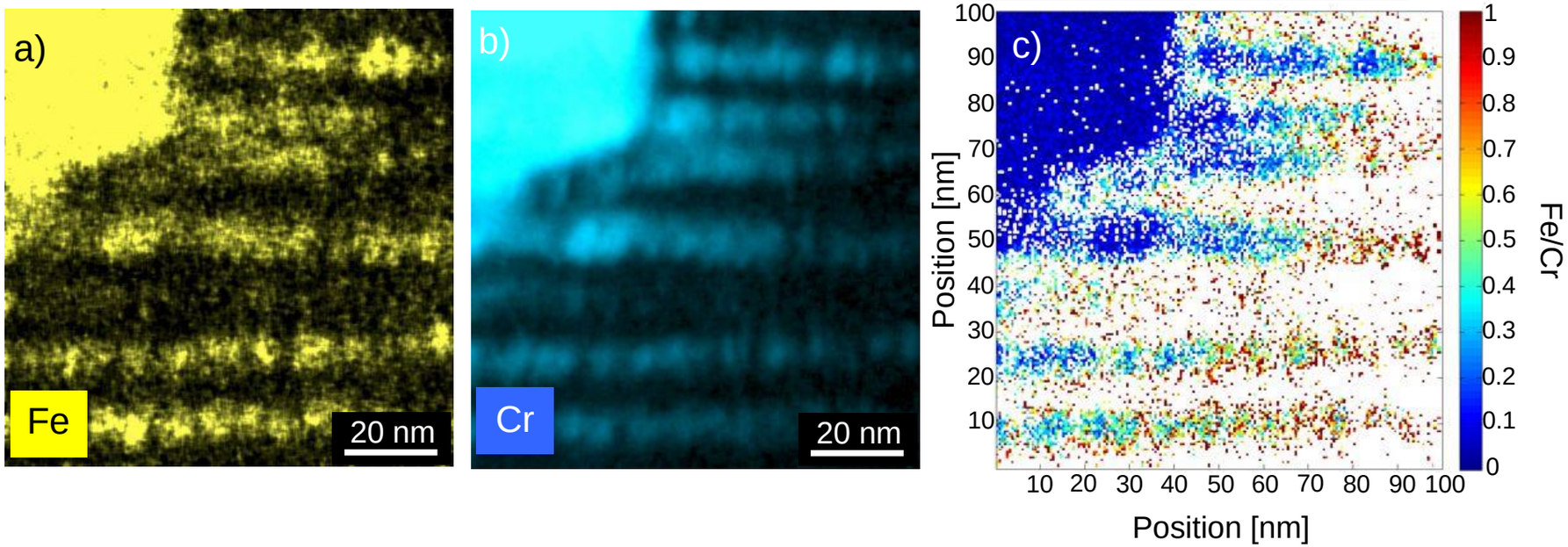
Zircaloy-2 neutron irradiated

14.7 x10^{25} n m^{-2} ~24.5 dpa

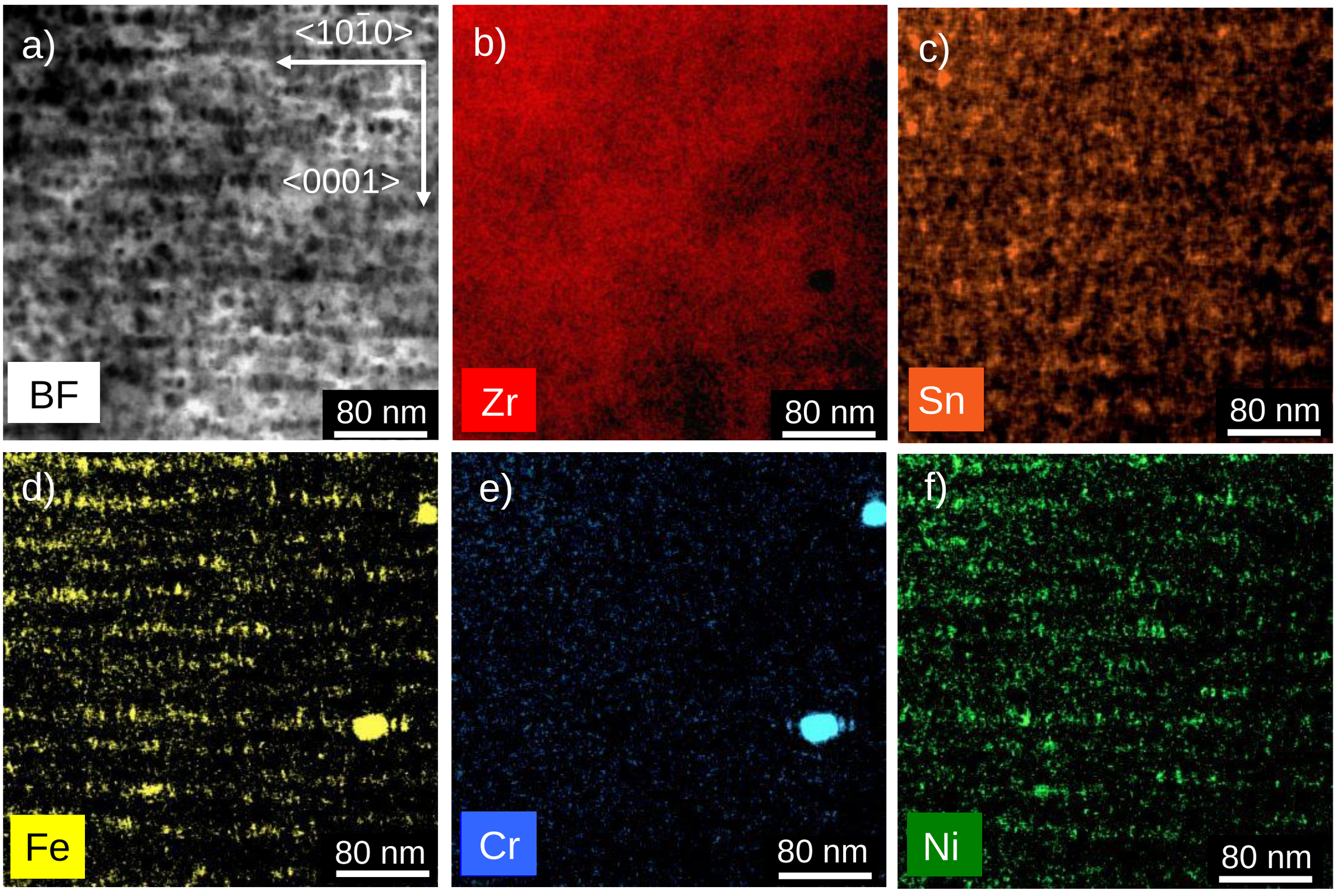
zone axis:

$< 11\bar{2}0 >$ BF STEM

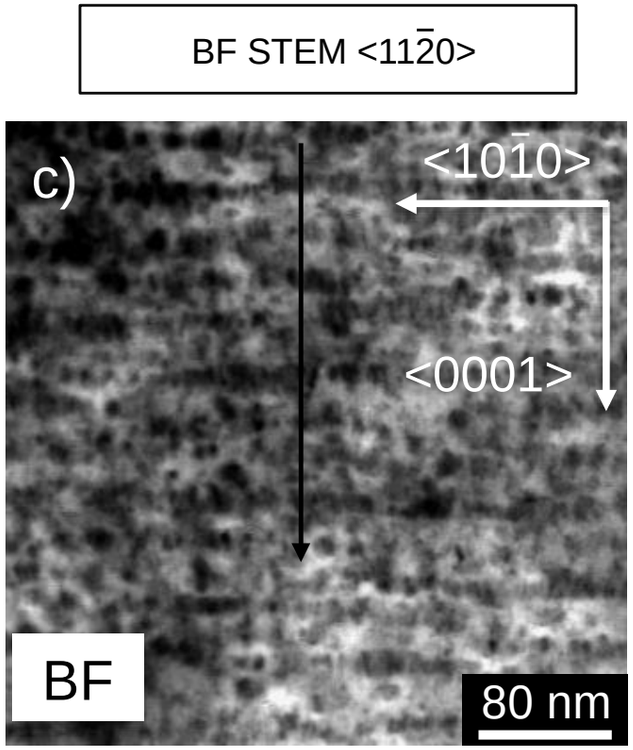


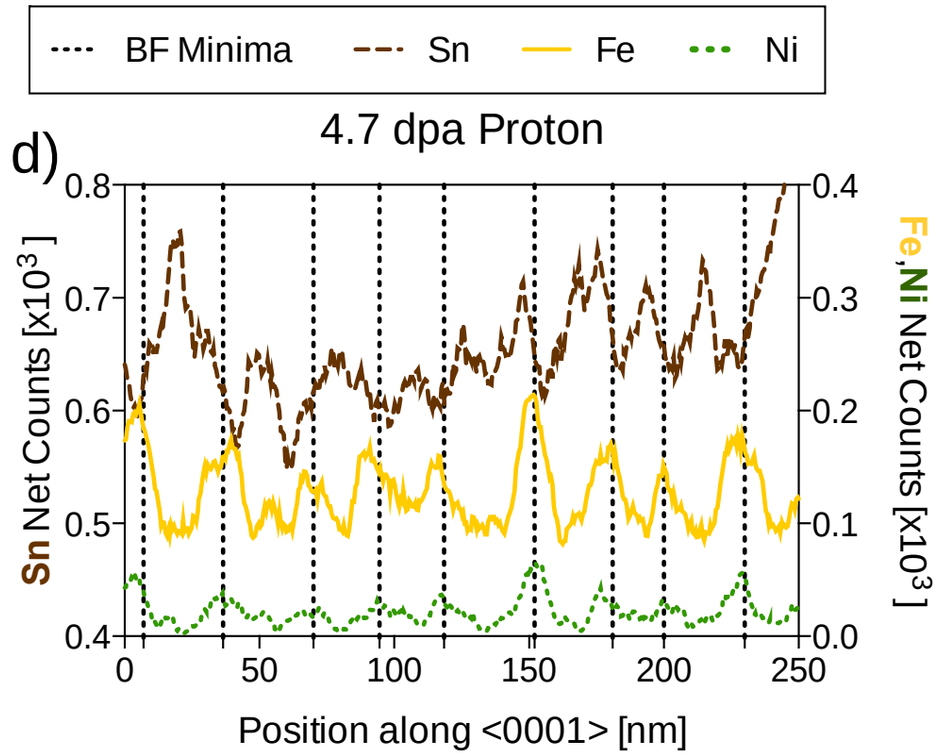


Zircaloy-2 proton irradiated

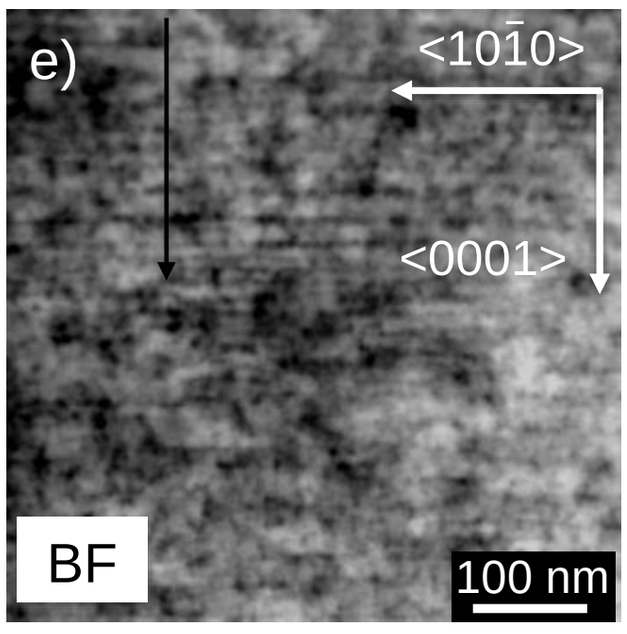


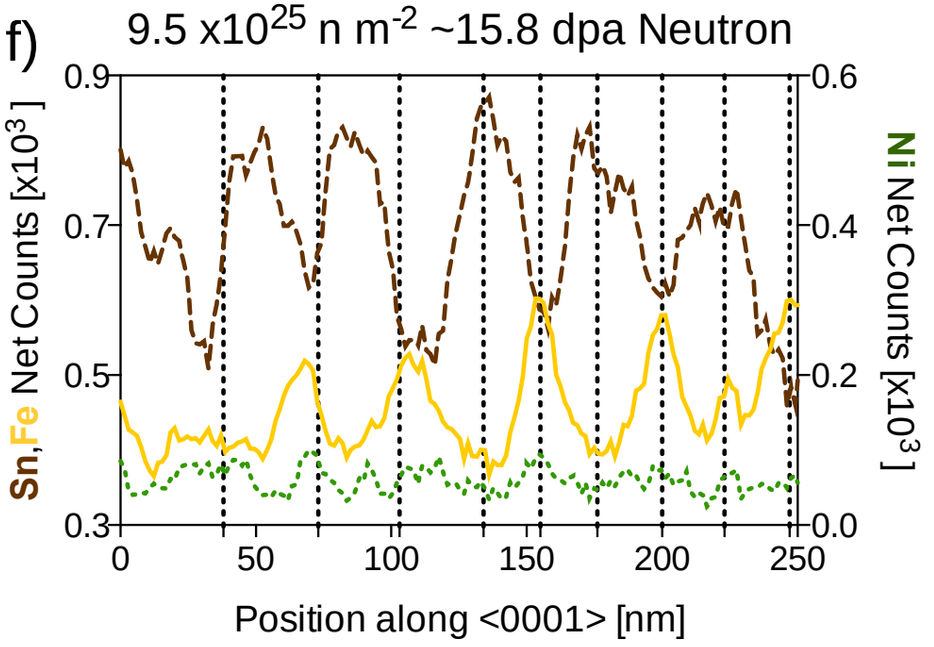
Microchemical Correlation





Proton





Neutron

Fe, Ni segregates to loops, Sn sits between loops

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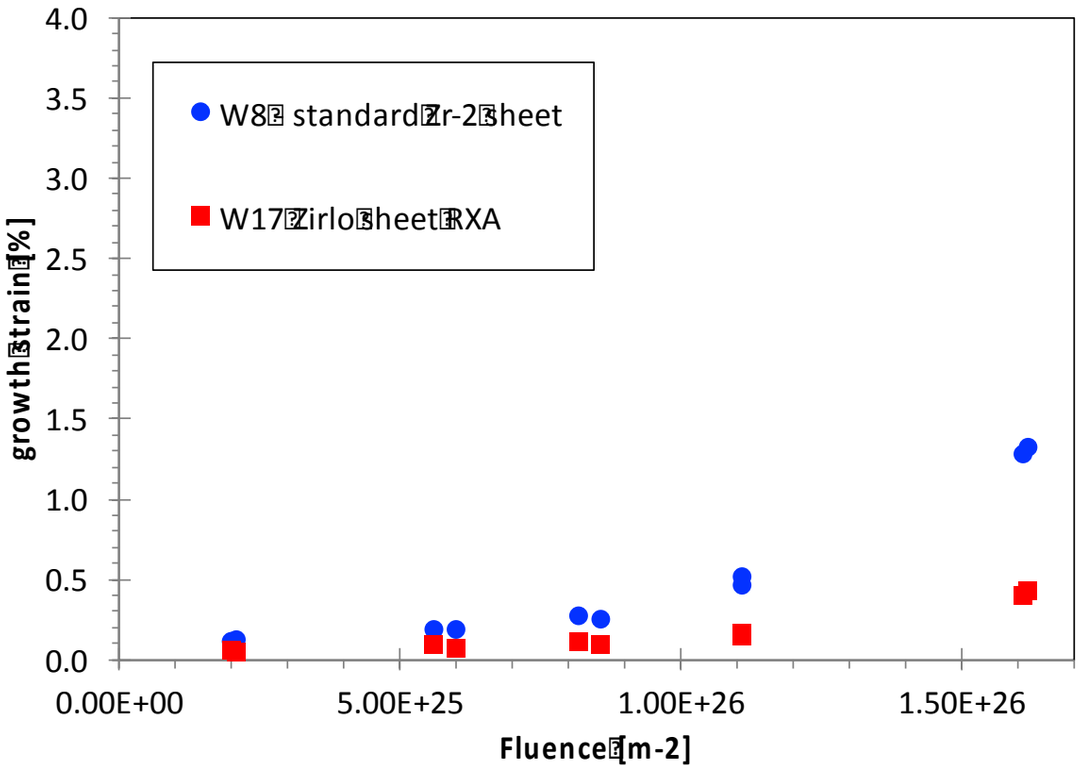
Question: What is the role of Nb during irradiation?

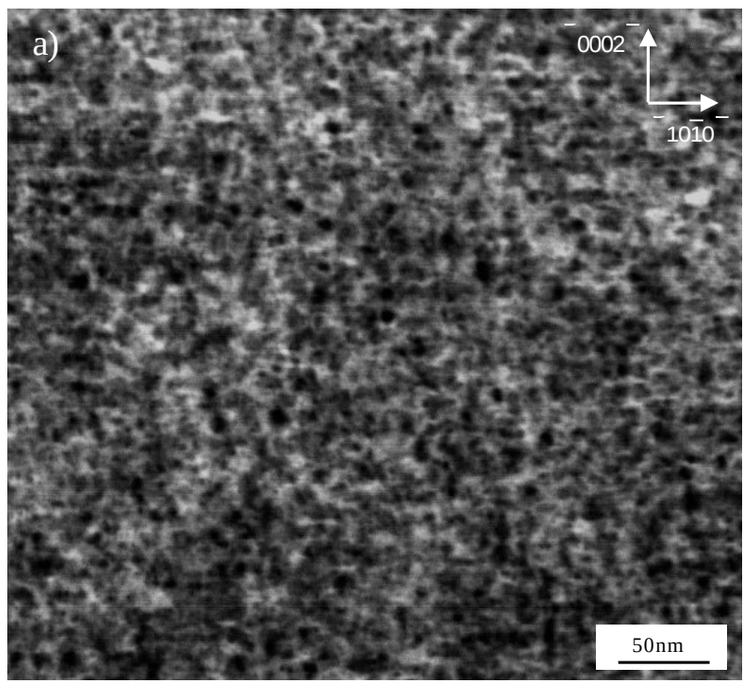
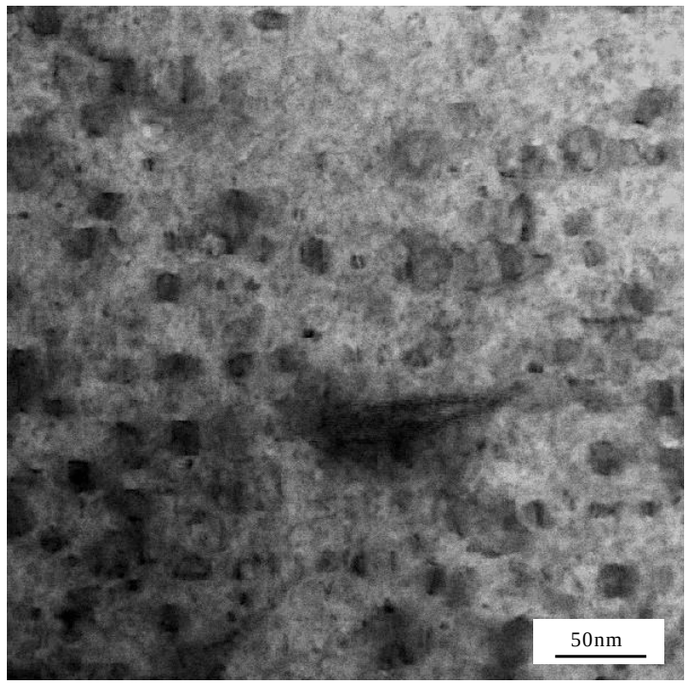
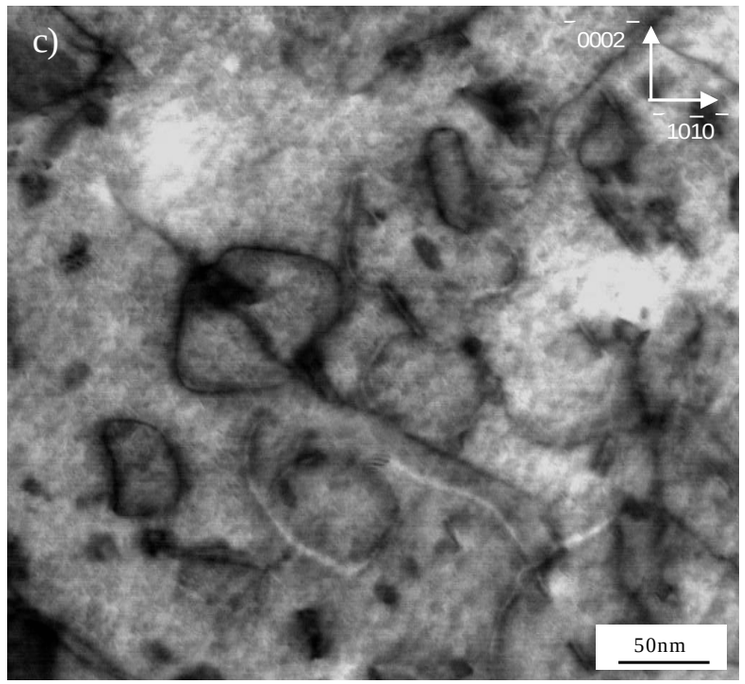
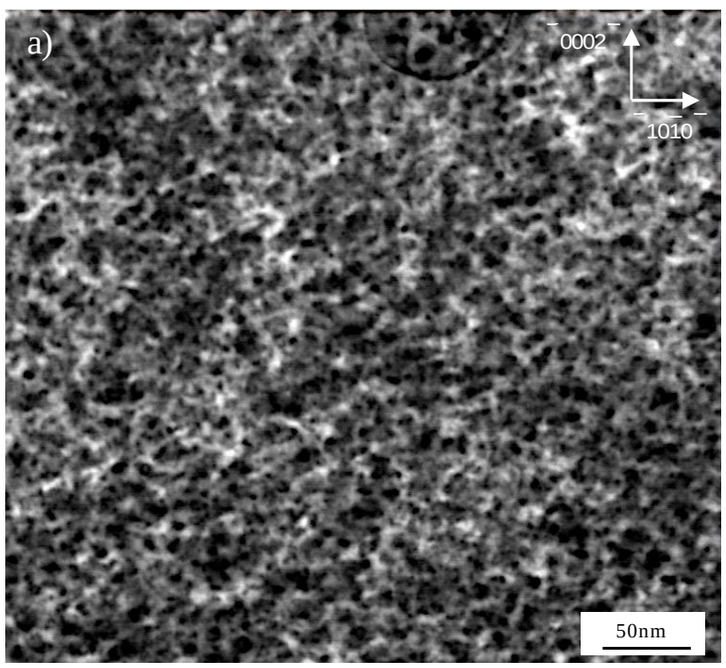
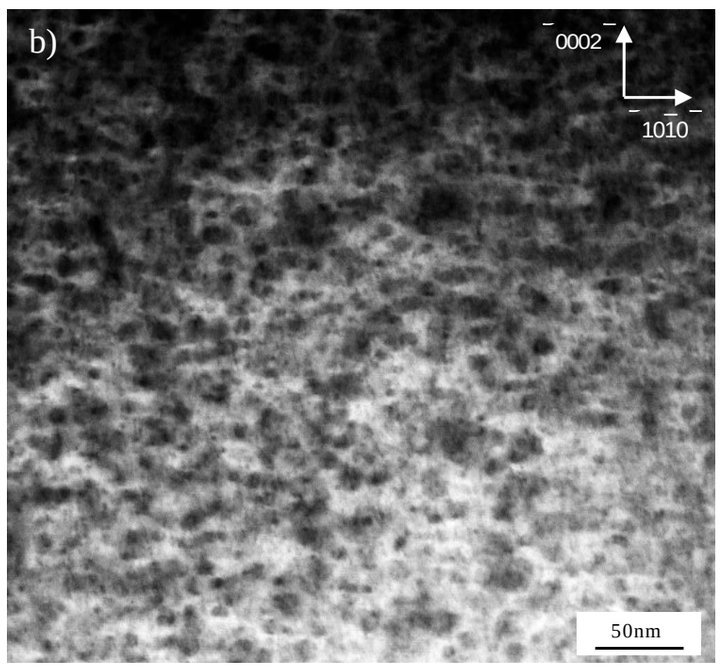
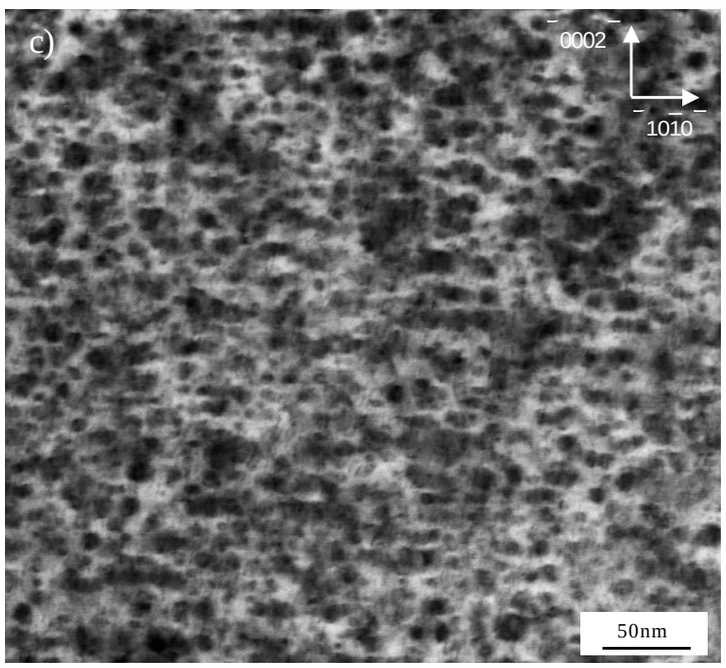
Effect of Nb

ZIRLO vs Zircaloy-2



Proton irradiation at different T

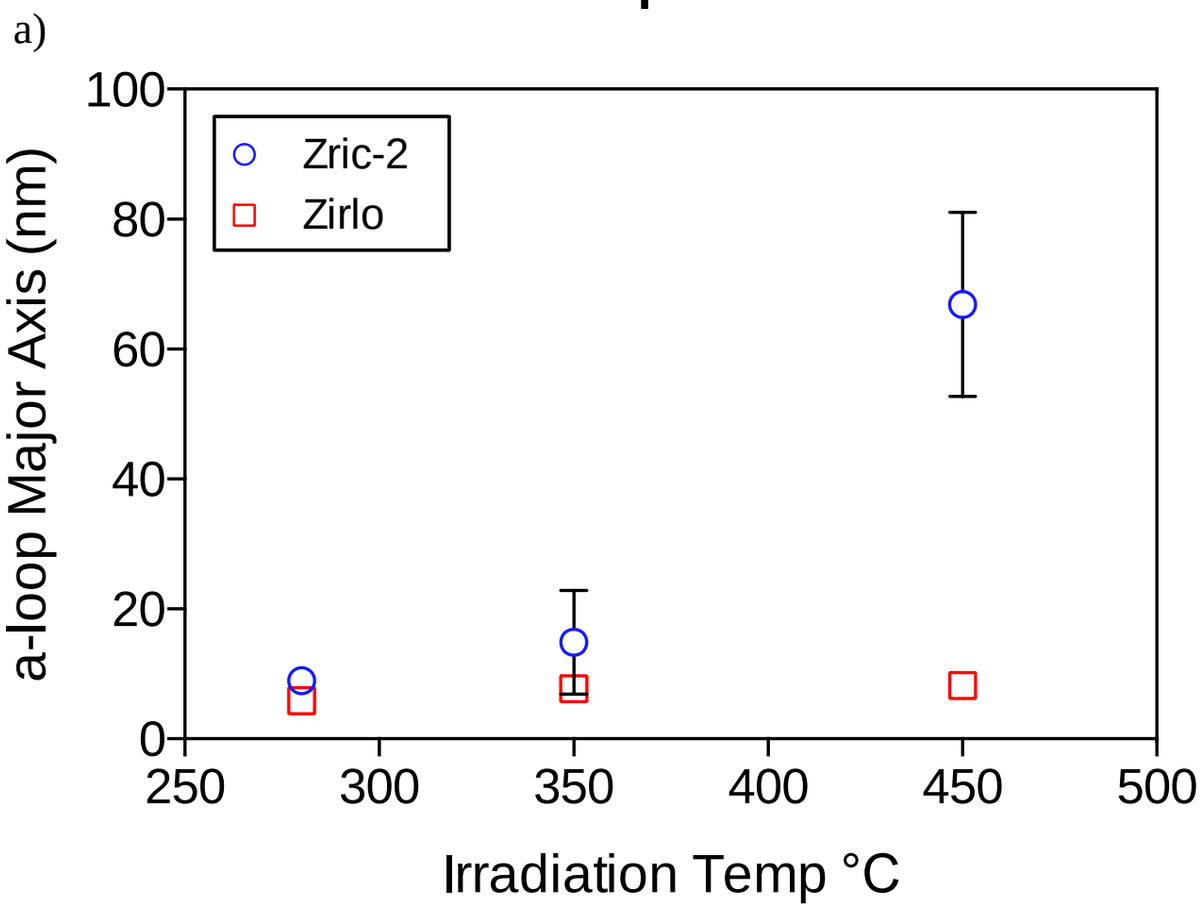
Zircaloy-2 (no Nb) and ZIRLO (contains Nb) irradiated to 2.3 dpa

	280ºC	350ºC	450ºC
Zircaloy-2			
ZIRLO			

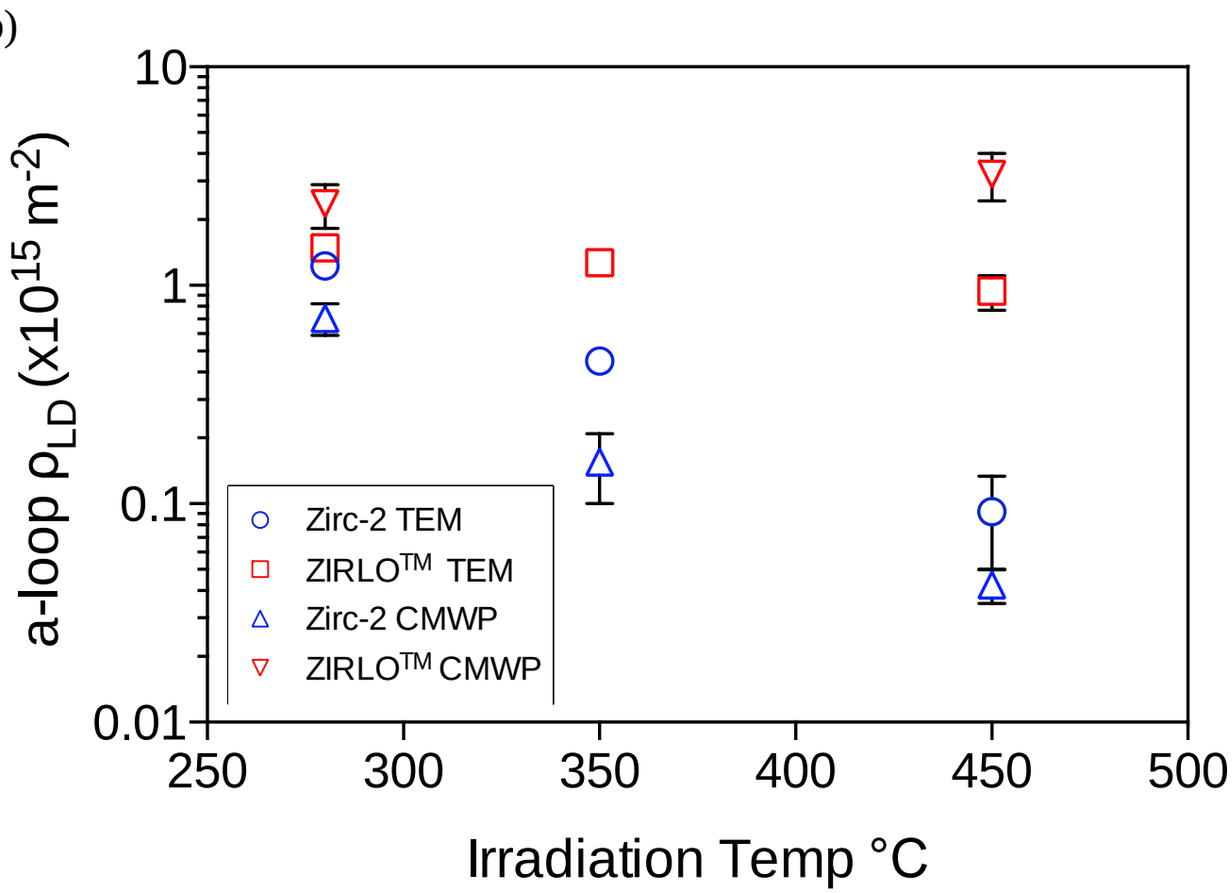
Proton irradiation at different T

Zircaloy-2 (no Nb) and ZIRLO (contains Nb) irradiated to 2.3 dpa

a-loop size



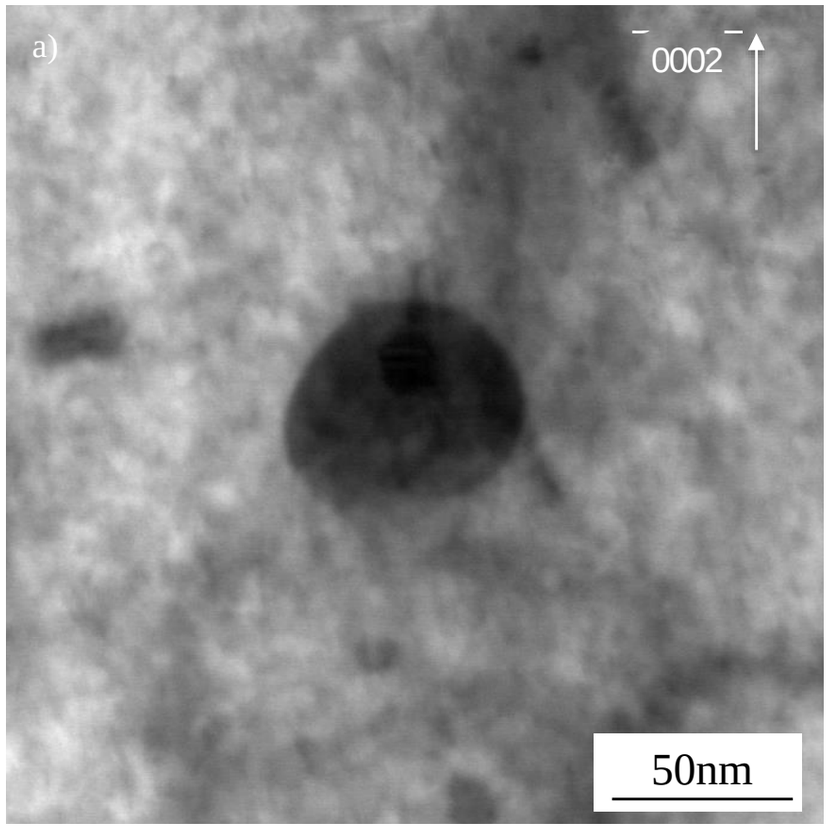
a-loop line density

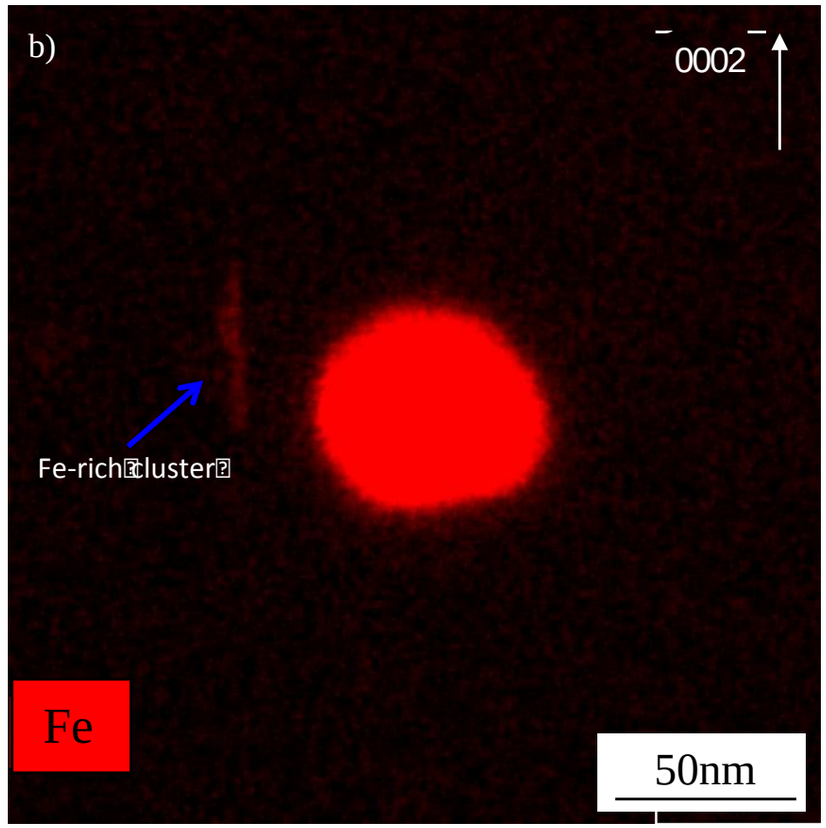


a-loop size and density in Zircaloy-2 very sensitive to irradiation temperature but not in ZIRLO

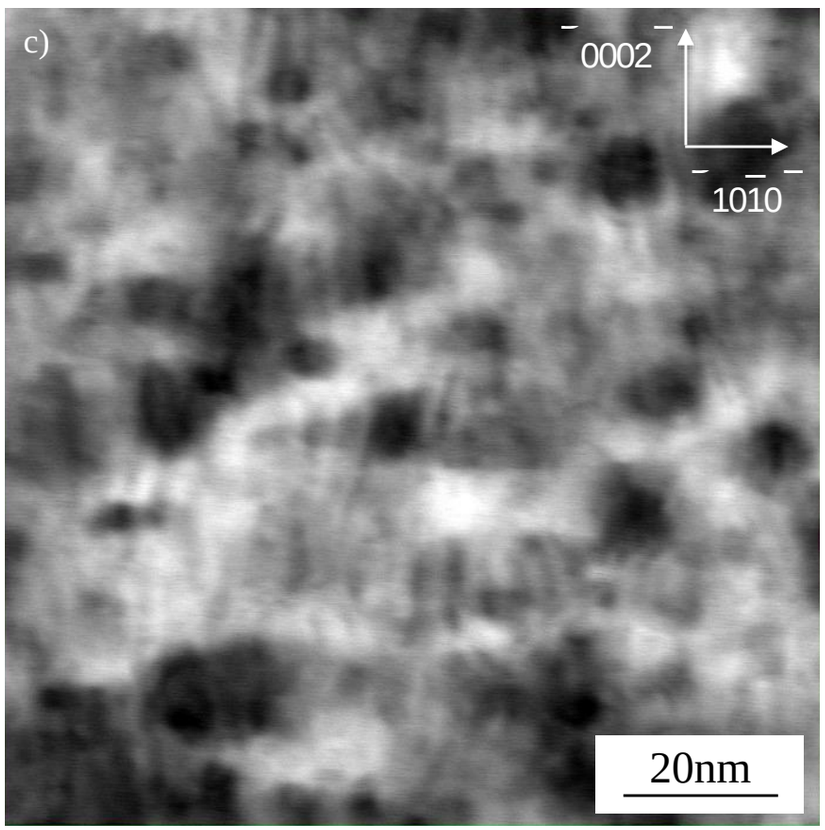
Nano precipitates/clusters

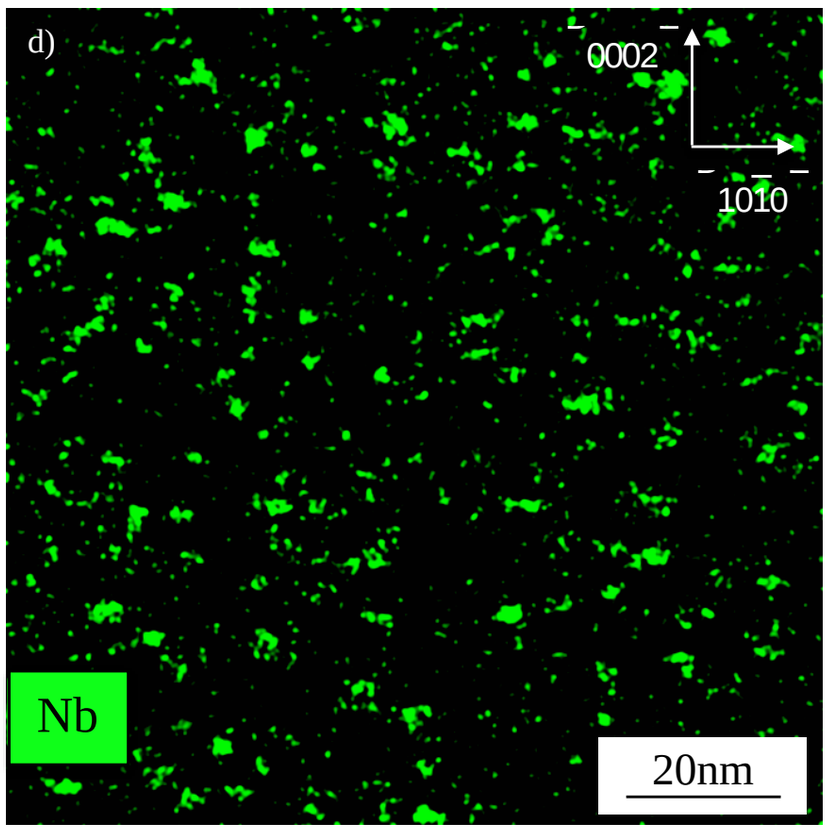
Zircaloy-2, 2.3 dpa





ZIRLO, 2.3 dpa





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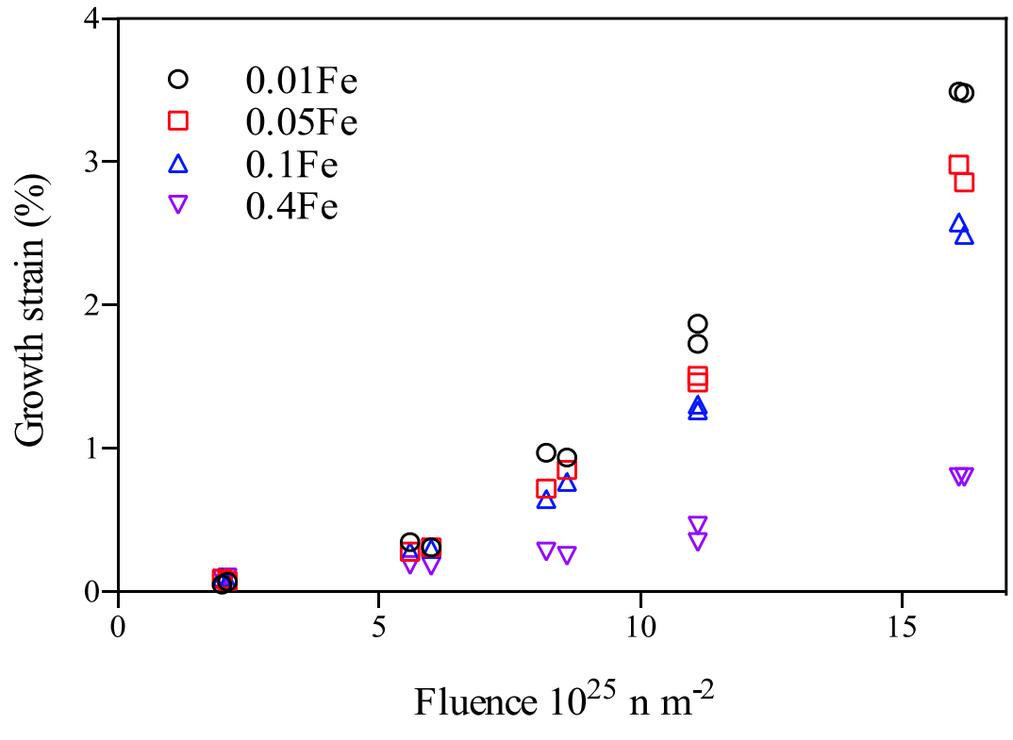
Question: By which way might Fe minimise Irradiation Induced Growth ?

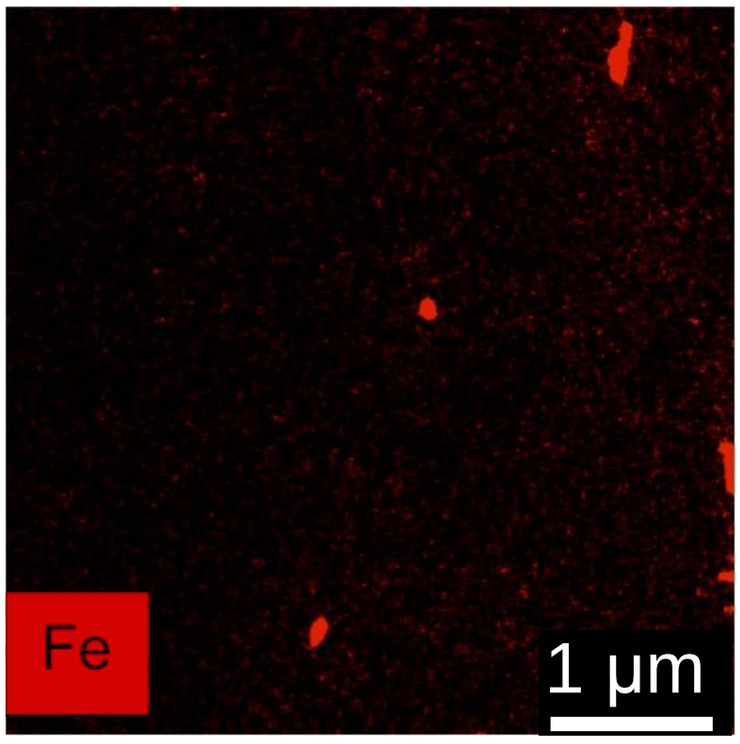
Effect of Fe
Zr-1Sn-XFe



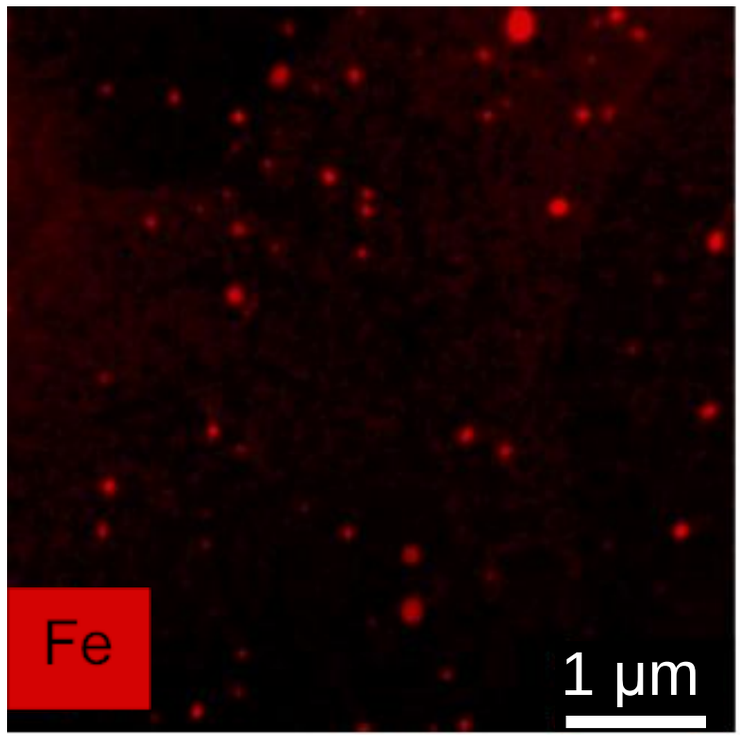
S. Yagnik, R.B. Adamson, G. Koblyansky, J.H. Chen, D. Gilbon, S. Ishimoto, et al., Zirconium in the Nuclear Industry: 18th International Symposium, 2016.

Effect of Fe on loop formation

Comparing Zr-0.1Fe binary alloy and Zircaloy-2



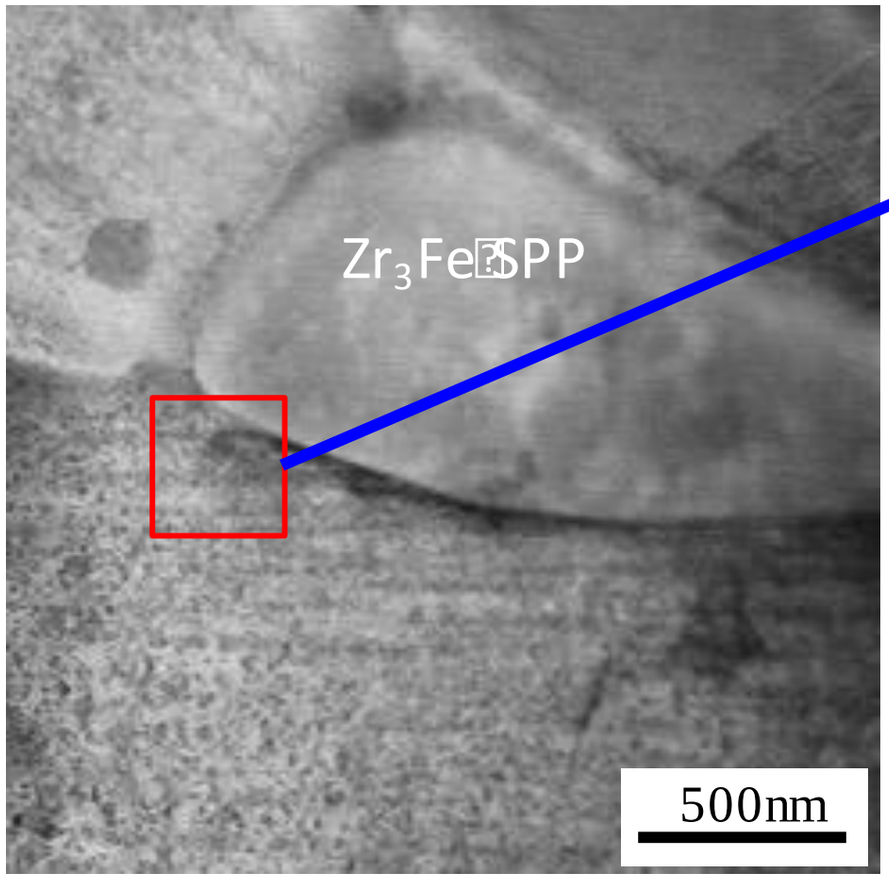
Zr_3Fe SPPs sparse throughout

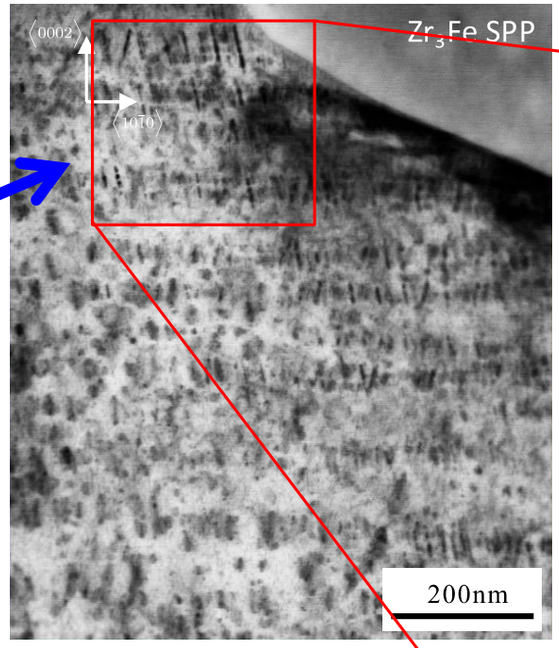


$Zr(Fe,Cr)_2$ SPPs more evenly distributed

Zr-0.1Fe irradiated to 1.5 dpa

ZA = <11$\bar{2}$0>



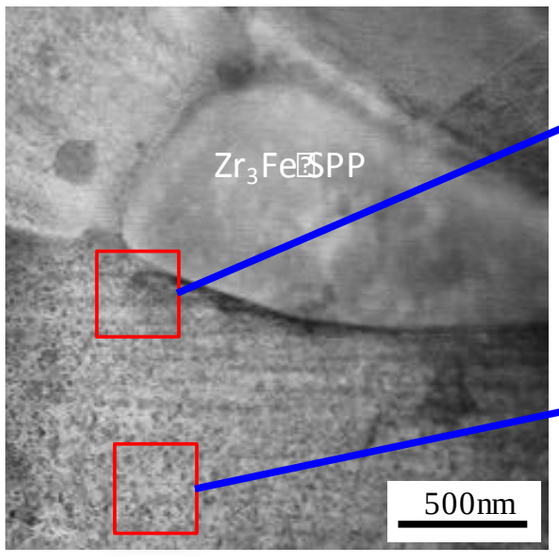


- Initially tilted c+a loops, which develop into tilted precipitates
- Can also bee seen in neutron irradiated material

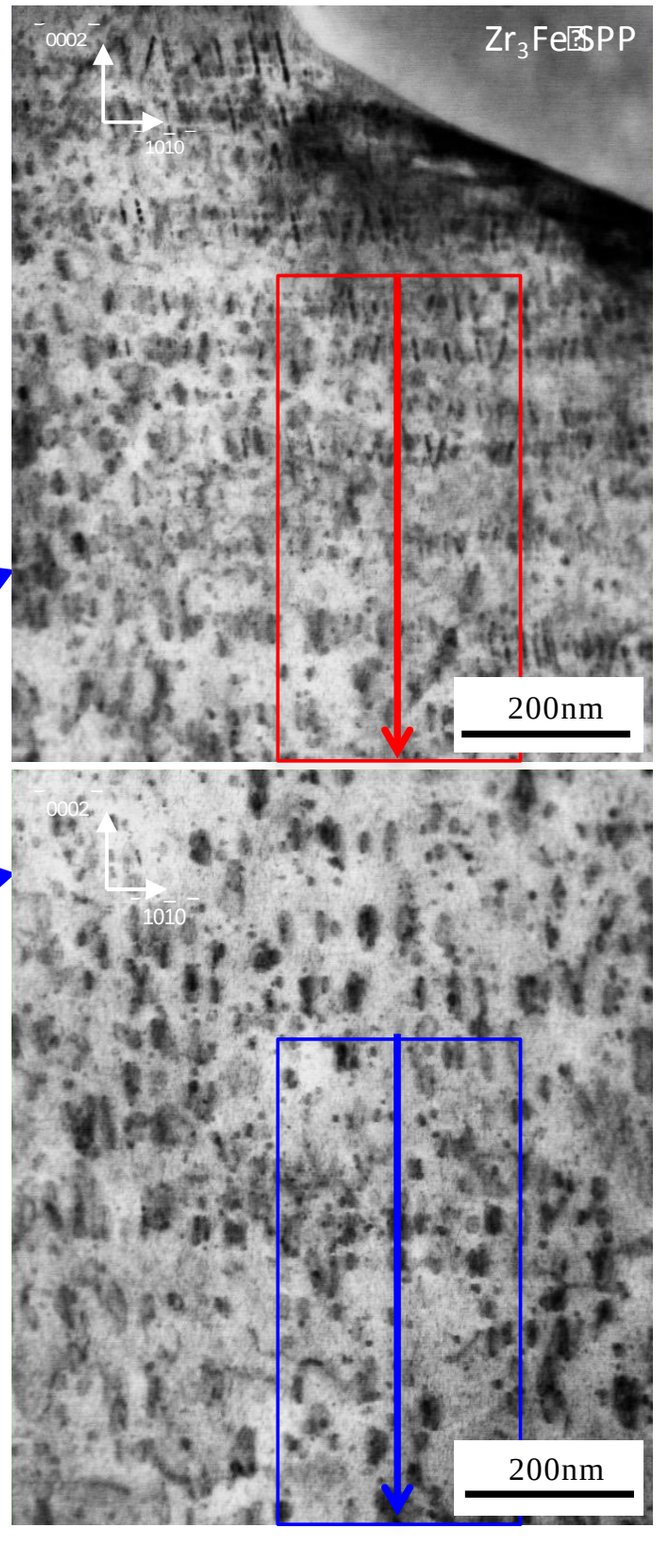
Zr-0.1Fe irradiated to 1.5 dpa

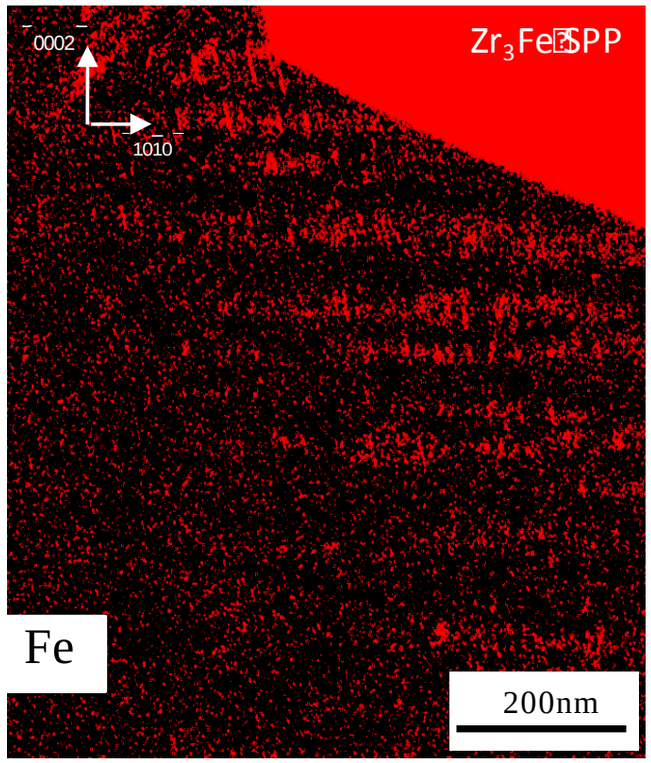
ZA = <11$\bar{2}$0>

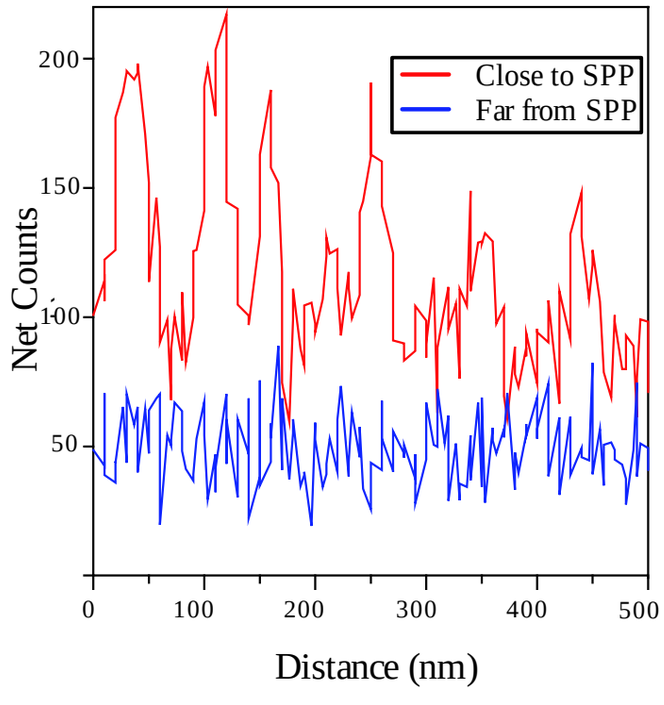
Close to a Zr_3Fe SPP



Away from a Zr_3Fe SPP







Trend does not change for Zr-0.1Fe with increasing dpa level

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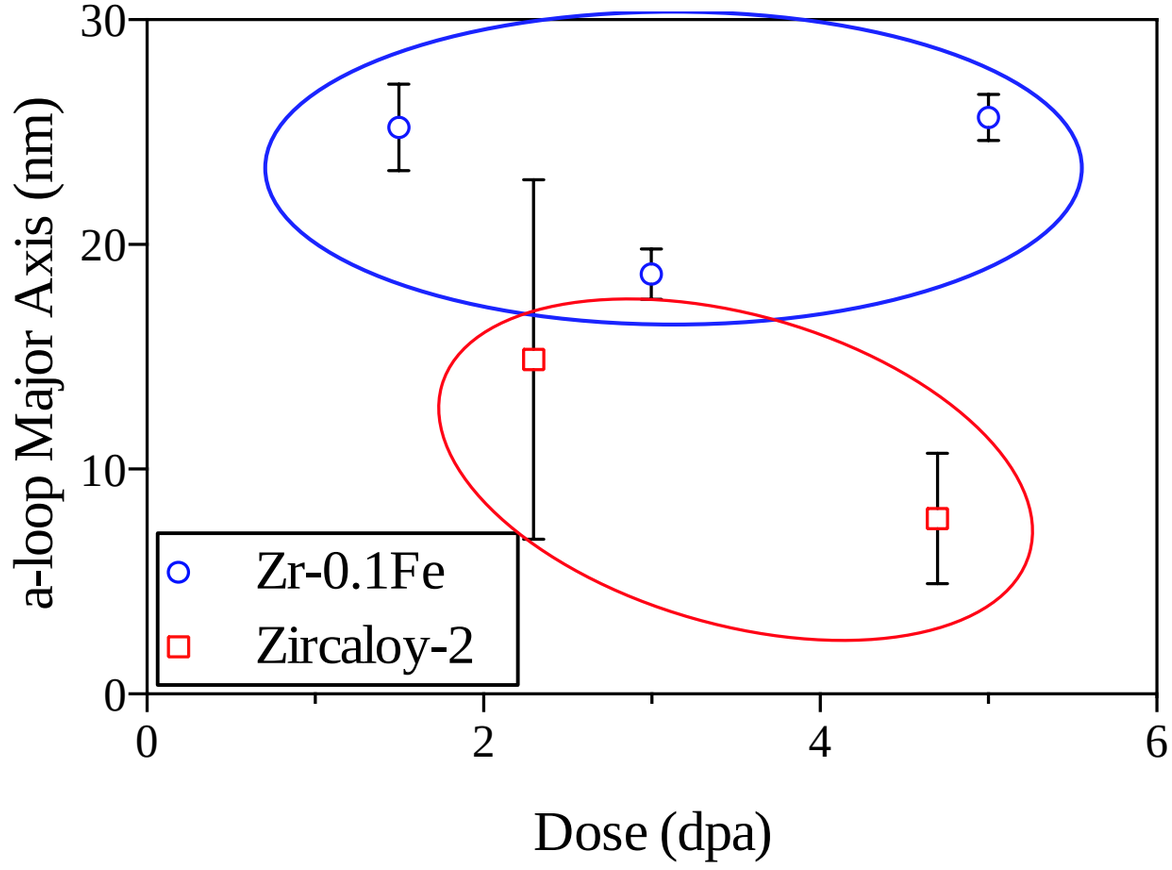
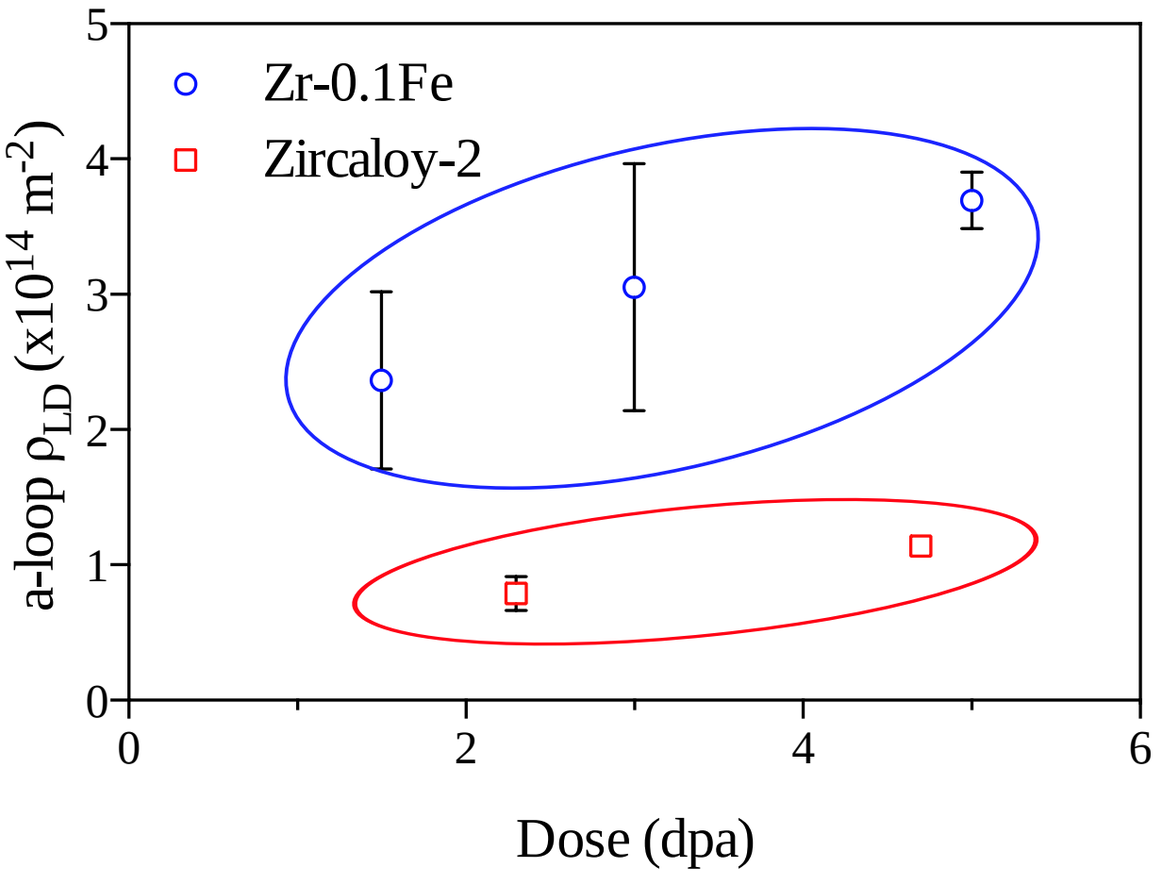
So we have Zircaloy-2 that develops rod-shape nano-precipitates throughout the material and Zr-0.1Fe that develops these features only close to sparsely spaced SPPs

Effect of Fe on a-loop size & density





a-loop size

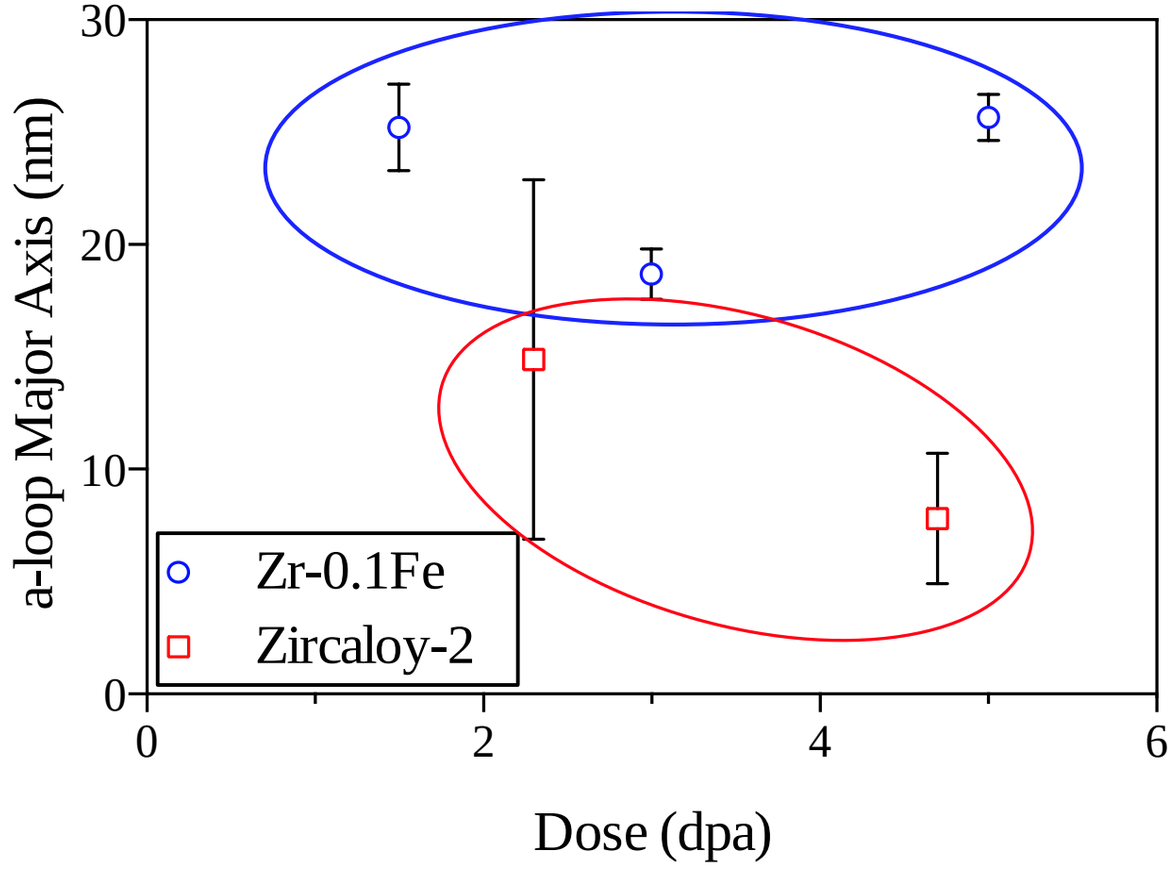
Zr-0.1Fe > Zircaloy-2

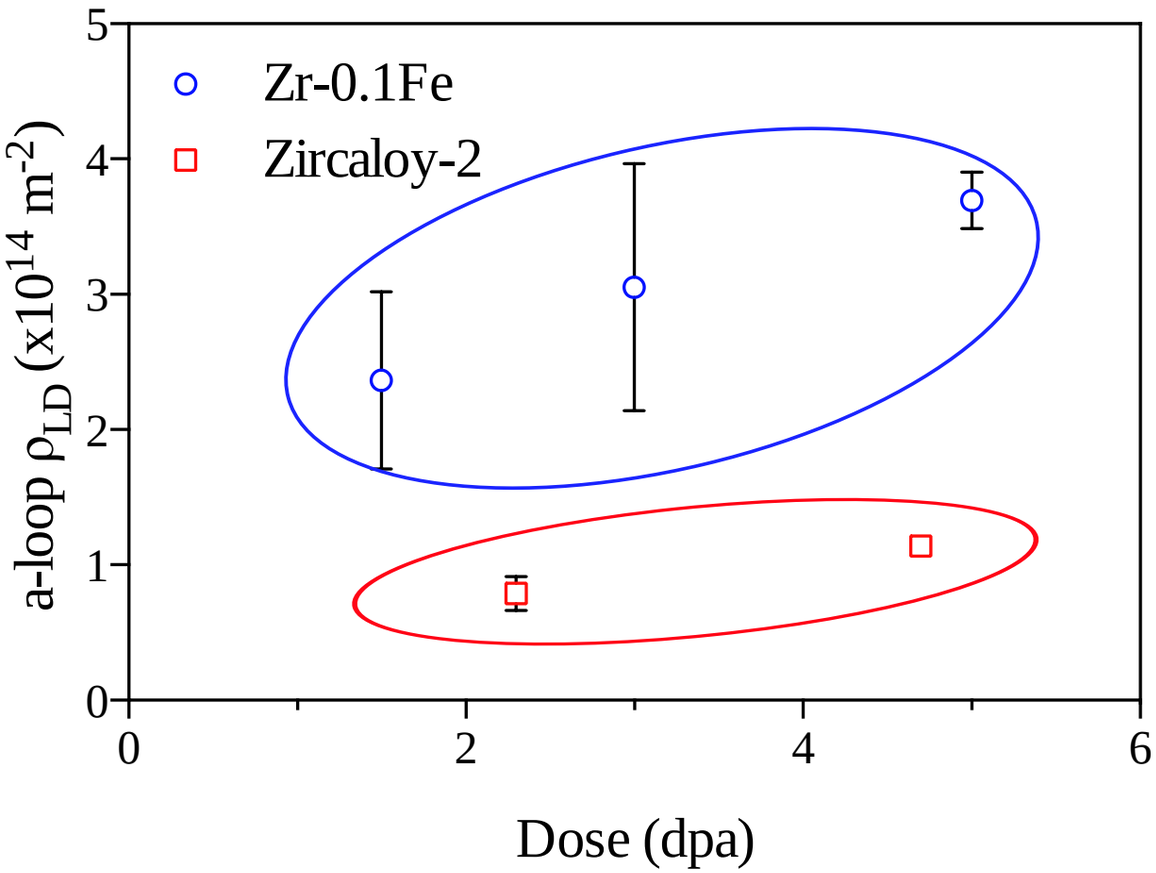
a-loop density

Zr-0.1Fe > Zircaloy-2

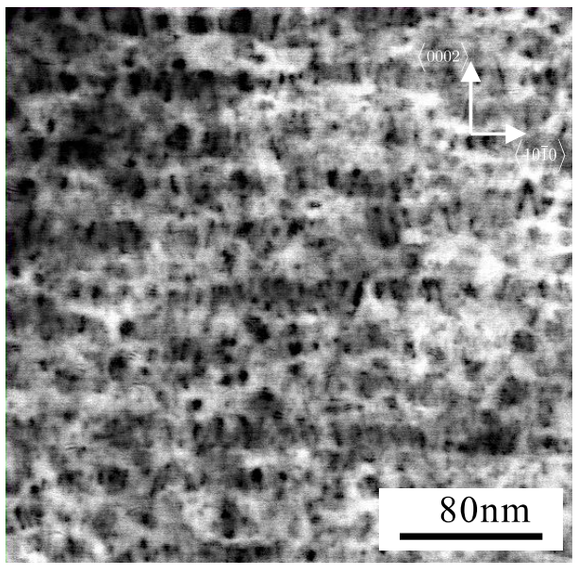
Once c-loops form: size and density also Zr-0.1Fe > Zircaloy-2

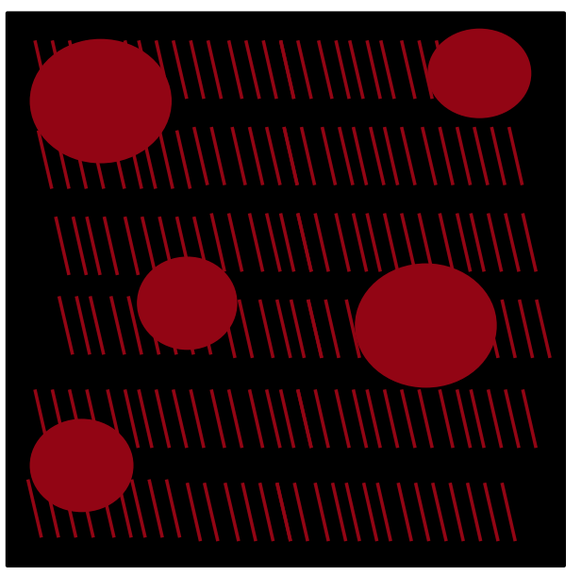
Effect of Fe on a-loop size & density



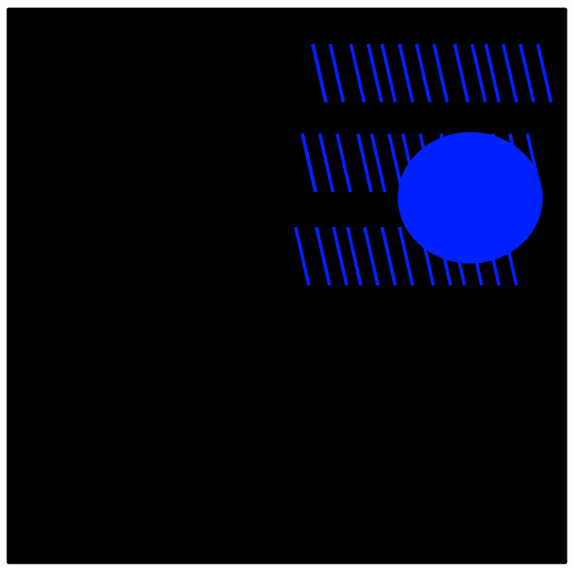


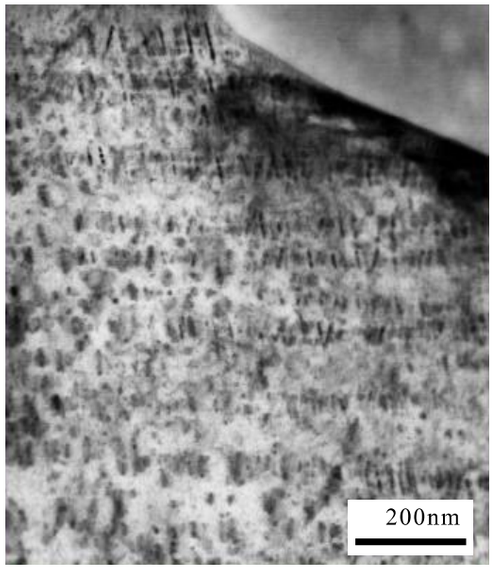
Zirc-2 4.7dpa





Zr-0.1Fe 5.0dpa





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Conclusion

- To date, all experimental data suggest that proton irradiation is an excellent surrogate for neutron irradiation for Zr alloys
- Experimental observations suggest complex chemical behaviors observed in neutron irradiated Zr alloys are replicated with proton-irradiations
- Fe promotes the formation of Fe-rich nano rods during irradiation, which might be point defect sinks
- Nb dramatically reduces growth and mobility of a-loops by forming Nb nano cluster.

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Thank You For Your Attention

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