Effet des impuretés sur l'endommagement induit par irradiation dans le tungstène

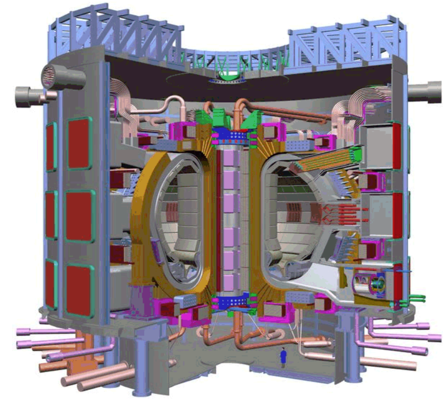
M-F. Barthe[a]

Z. Hu (phD)[a], C. Genevois[a], P. Desgardin[a], B. Decamps[b], R.Schaeublin[c], F. Jomard[d], M Loyer Proste[e]

[a]CEMHTI, CNRS/Orléans University, 3A Rue de la Férollerie, 45071 Orléans, France
[b]IJCLab/CNRS, Paris - Saclay University, France
[c]Metal Physics and Technology Department of Materials, ETH Zürich, HCI G515, 8093 Zürich Switzerland
[d]GEMac, Univ. Versailles, 45 avenue des Etats Unis, 78035 Versailles cedex, France
[e]CEA-DEN-DANS/DMN/SRMP, Saclay, 91191 Gif-sur-Yvette cedex, France

WPMAT

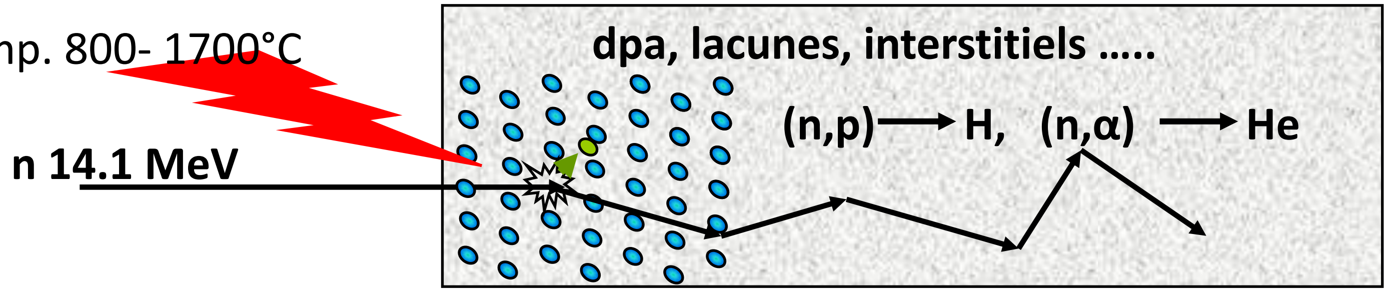
- ✓ ITER, DEMO : W divertor, première paroi?
- ✓ Fusion réaction $^{2}H + ^{3}H \rightarrow ^{4}He(3,5\ MeV) + n\ (14.1\ MeV)$



ITER Tokamak

dpa : displacements per atom

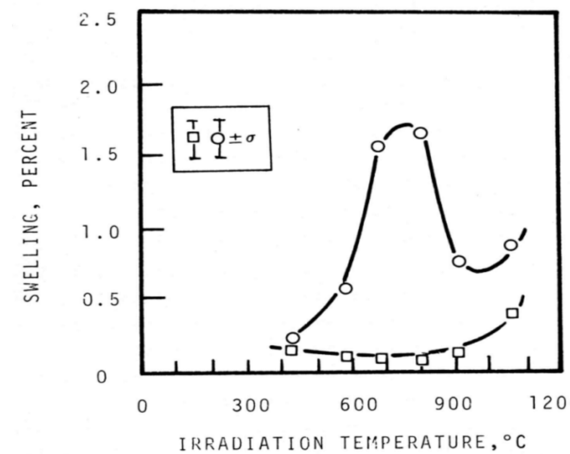
Temp. 800- 1700°C



Modification de la microstructure et de la composition chimique

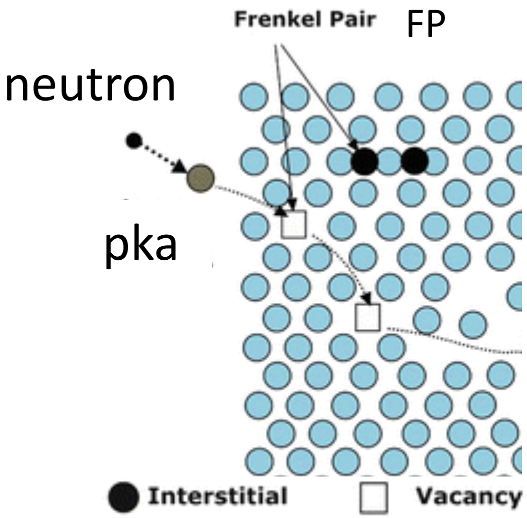
⟹ Evolution des propriétés thermiques, électriques et mécaniques des matériaux

- Gonflement in W irradiated with neutrons (E>0.1MeV), $5.5 \times 10^{26} n.m^{2}$ (9.5dpa) [1]

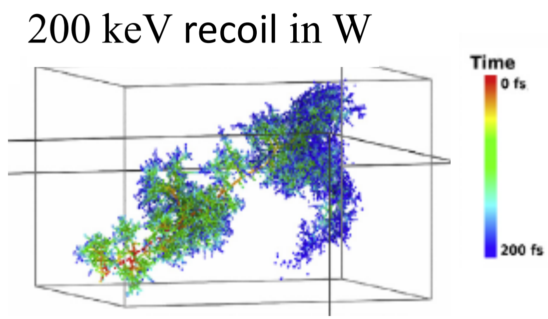


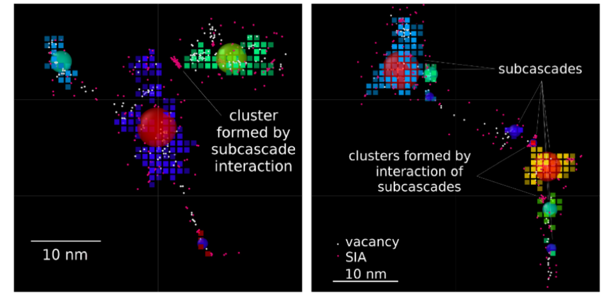
Irradiation induced damage

Irradiation ➔ collision cascades, atomic displacements, defects, damage dose (dpa)



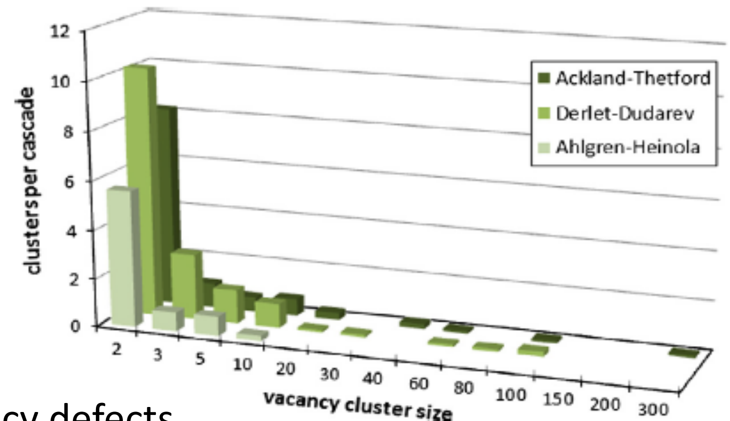
pka (primary knocked atom, recoil)





single vacancy (V)
interstitials (SIA)
and clusters (loops, cavities ….)

MD cascades for 150 keV W in W [2]

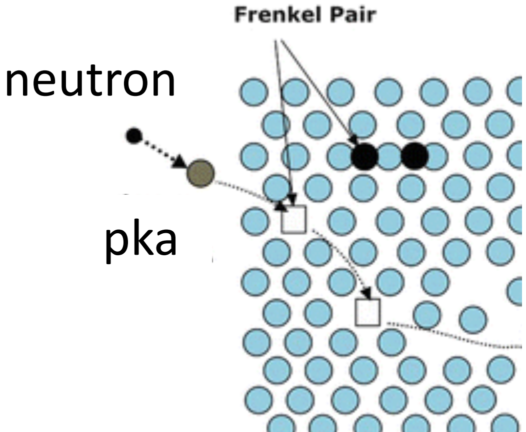


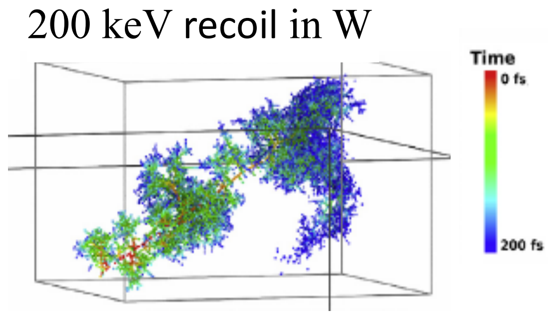
Vacancy defects

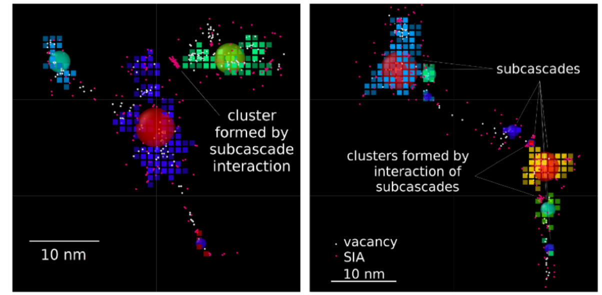
PAS : Positron annihilation spectroscopy
➔ single vacancy to small cavity (< 1 nm)
TEM : Transmission electro Microscopy
➔ cavities from 0.5-1 nm

Irradiation induced damage

Irradiation ➔ collision cascades, atomic displacements, defects, damage dose (dpa)







single vacancy (V)

SIA)

….)

pka

Irradiation with ***W ions (self-irradiation)*** *to mimic neutron irradiation*

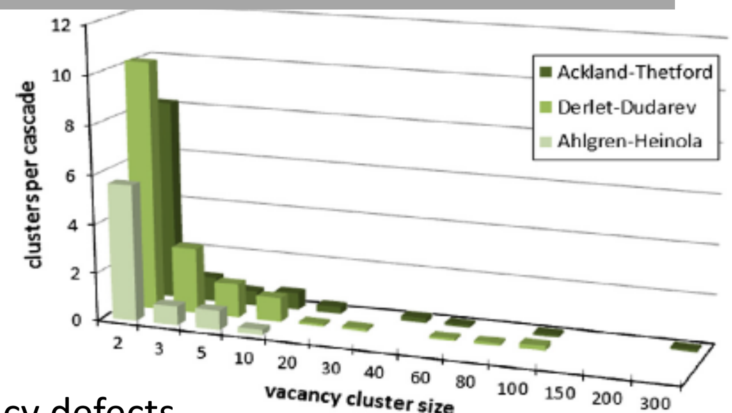
]

PAS : Positron annihilation spectroscopy

➔ single vacancy to small cavity (< 1 nm)

TEM : Transmission electro Microscopy

➔ cavities from 0.5-1 nm

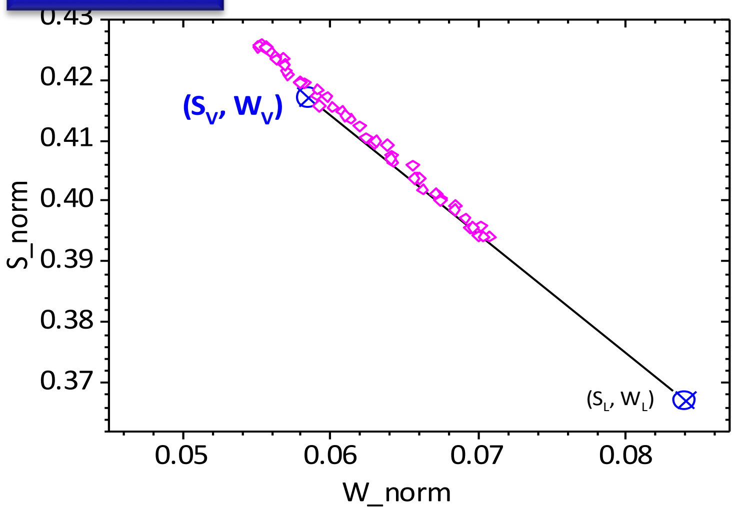


Vacancy defects

Vacancy defects at RT

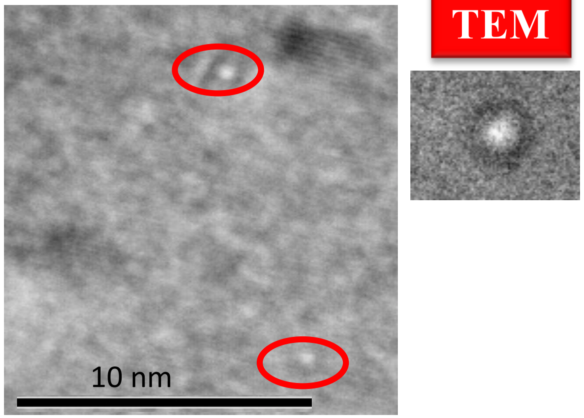
Self-irradiated W ions 0.02 dpa*, RT

PAS-DB



➔ ***Tokamak [1] ~ 48 hours***

TEM

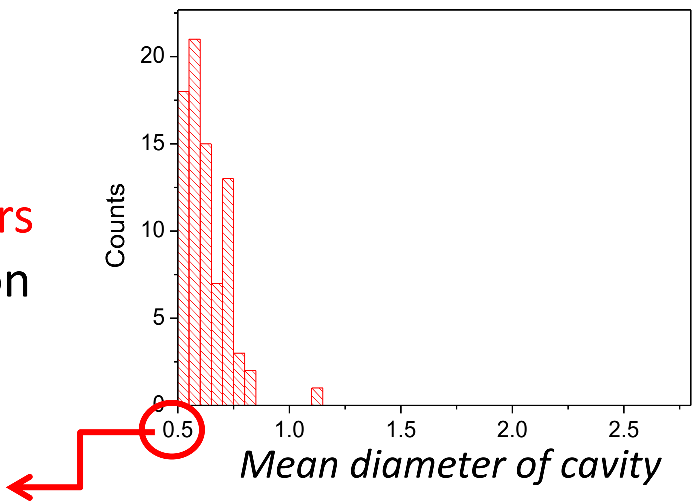


Underfocused

Majority of Single vacancy and small clusters

[V] ~ 0.2 %

- Cavities or Vacancy clusters
- Non symetrical distribution



[1] MR Gilbert JNM 442 (2013) S755

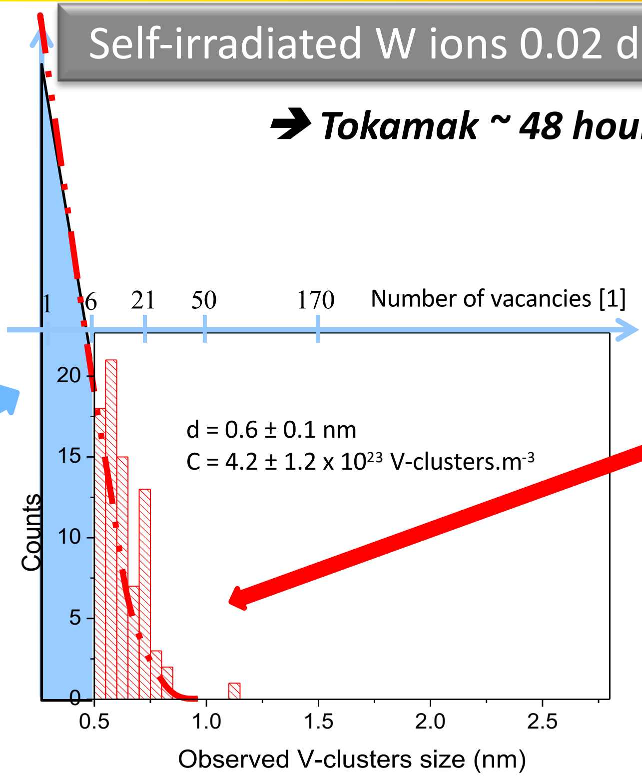
Vacancy defects at RT

PAS-DB

Self-irradiated W ions 0.02 dpa*, RT

➔ *Tokamak ~ 48 hours*

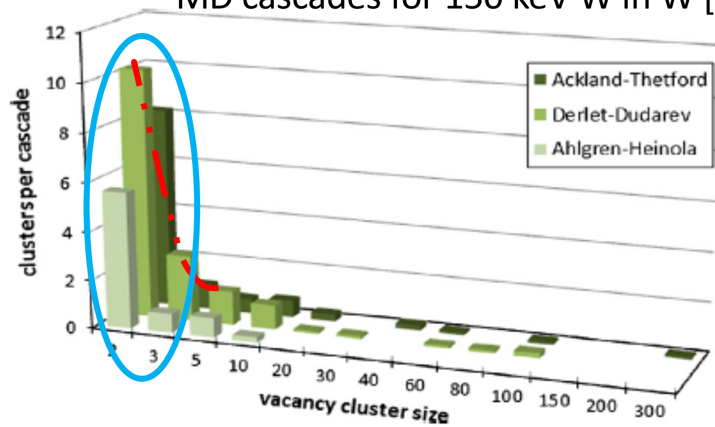
TEM



Majority of Single vacancy and small clusters

Cavities

MD cascades for 150 keV W in W [2]



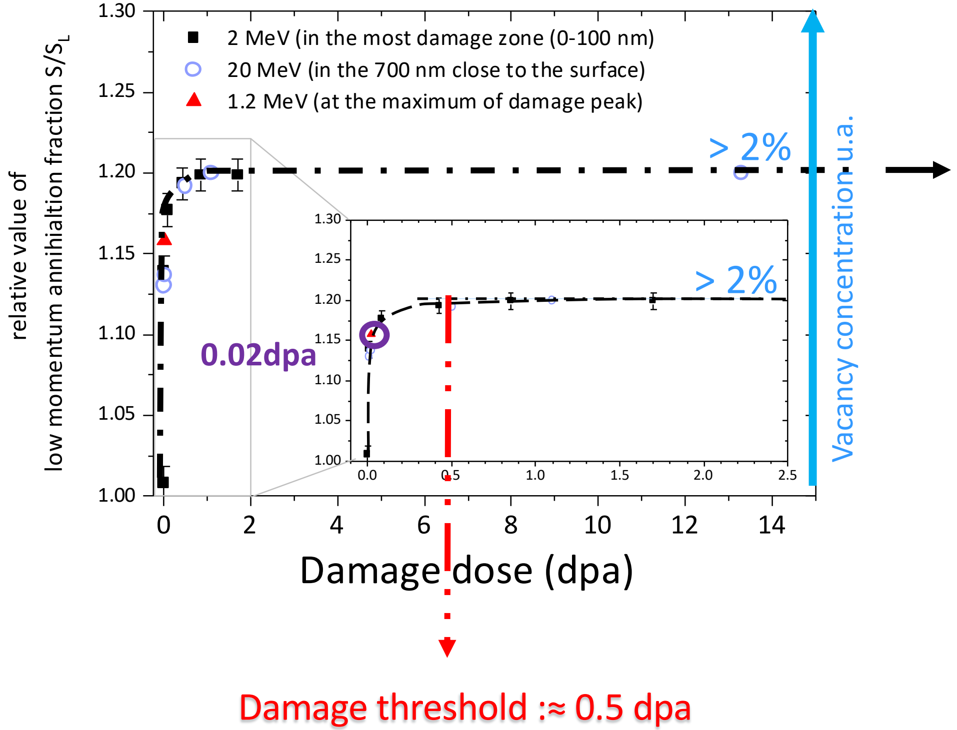
- Vacancy clusters are formed in the cascades
- $E_{mig}(V)$=1.66 eV [3,4] ➔ no clustering expected at RT

[1] $N=(d/(2xr_W))^3$
[2]A. Sand et al JNM 455 (2014) 207–211

When damage dose (dpa) increases

PAS-DB

Experiments :
W ions irradiations at RT

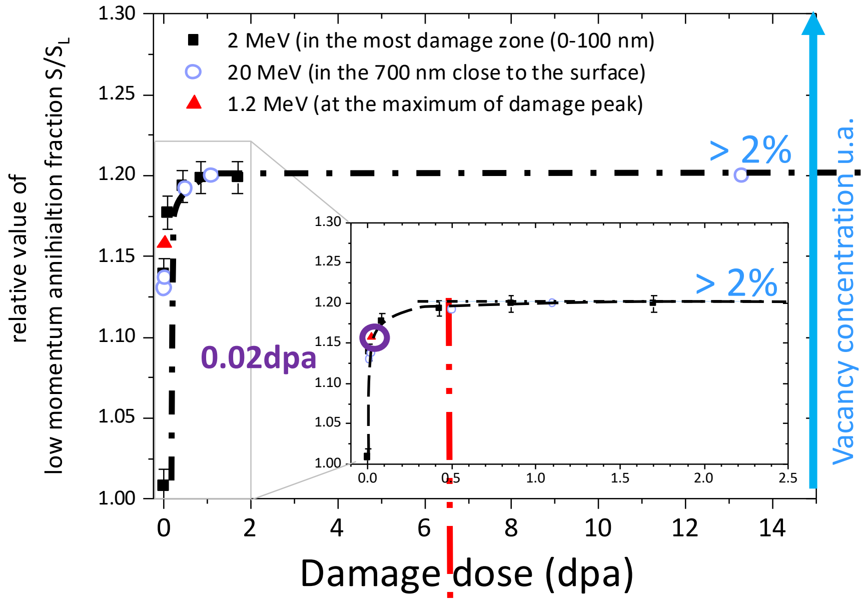


→ **Saturation of PAS signal or of the damage distribution**

[1] A. Hollingsworth, MF Barthe et al JNM 558 (2022) 153373
[2] P.M. Derlet and S.L. Dudarev, Physical Review Materials **4**, 023605 (2020).

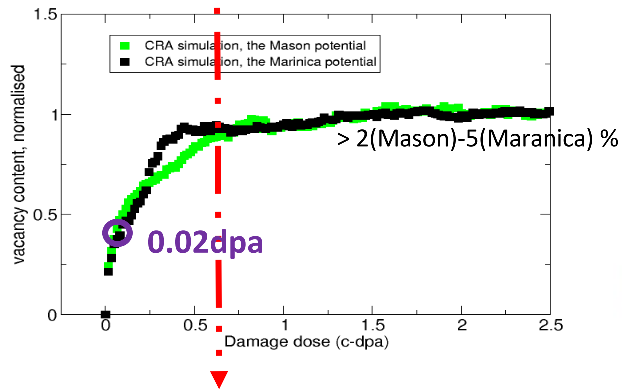
When damage dose (dpa) increases

Experiments :
W ions irradiations at RT



→ **Saturation of PAS signal or of the damage distribution**

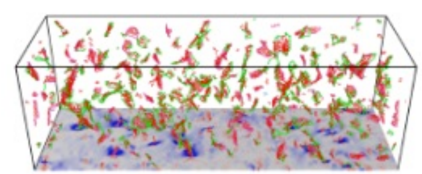
Simulations using the FP **creation-relaxation algorithm (CRA)** :2 interatomic potentials [2]



Damage threshold :≈ 0.5 dpa

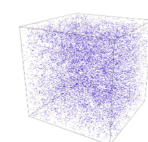
➔ Saturation of the damage distribution

Frenkel pairs accumulation

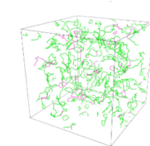


0.05 dpa

V



dislocations



Box size :20.2x20.2x63.2 nm^3
Vacancy (blue)
Interstitial (red)
clusters with > 3 point defects are shown
1/2<111> (green), <100> (pink) dislocation lines

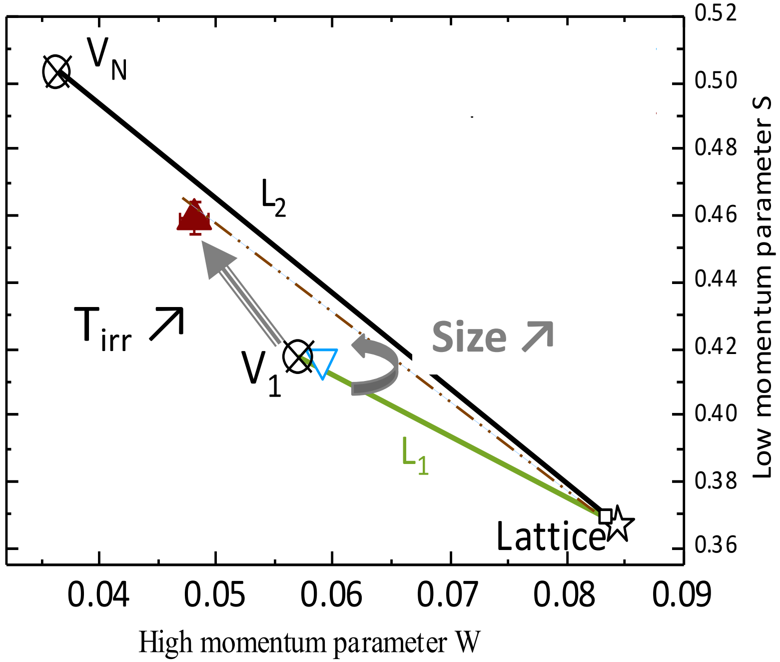
➔ Tokamak ~ 50 days

[1] A. Hollingsworth, MF Barthe et al JNM 558 (2022) 153373
[2] P.M. Derlet and S.L. Dudarev, Physical Review Materials **4**, 023605 (2020).

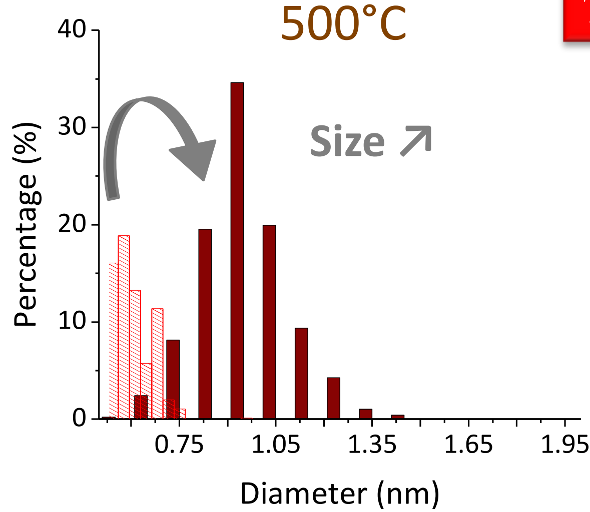
Irradiation-induced damage at high temperature

PAS-DB

Self-irradiated W ions 0.02 dpa*, 500°C



TEM



Damage level (dpa)	0.02
Irradiation Temp. (°C)	500
Mean diameter (nm)	**0.97 ± 0.19**
Density (10^{24} m^{-3})	**1.73 ± 0.71**
Swelling (%)	**0.09 ± 0.02**

V cluster size ↗ drastically when irradiation temperature ↗

high mobility of single and tri-vacancy at 500°C [1-4]

$$E_m^{V_1} = 1.66eV$$ [1]

→ *agglomeration of V between them or on Vn directly generated in collision cascades*

[1] Becquart C.S et al. Nuclear Instruments and Methods in Physics Research Section B 2007.
[2] A. Debelle, M.F. Barthe, T. Sauvage,, Journal of Nuclear Materials. 376 (2008) 216–221. https://doi.org/10.1016/j.jnucmat.2008.03.002.
[3] K. Heinola, F. Djurabekova, T. Ahlgren, On the stability and mobility of di-vacancies in tungsten, Nucl. Fusion. 58 (2018) 026004. https://doi.org/10.1088/1741-4326/aa99ee
[3] D. Mason, D. Nguyen-Manh, C. Becquart, An empirical potential for simulating vacancy clusters in tungsten, Journal of Physics: Condensed Matter. 29 (2017). https://doi.org/10.1088/1361-648X/aa9776

ITER Material Specification for W

Table 1 - Chemical composition of W plates

Element	Composition max, wt. %	Permissible variation in Check analysis, wt. %
C	0.01	±0.002
O	0.01	+10% relative
N	0.01	+0.0005
Fe	0.01	+0.001
Ni	0.01	+0.001
Si	0.01	+0.001

Effect of purity

ITER Material Specification for W

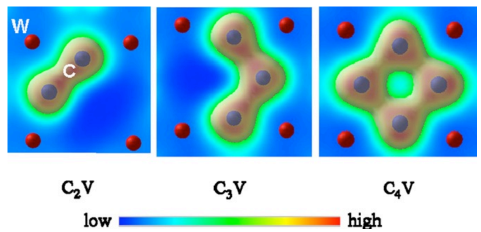
Light elements (LEs)
1100- 1500 at. ppm

Table 1 - Chemical composition of W plates

Element	Composition max, wt. %	Permissible variation in Check analysis, wt. %
C	0.01	±0.002
O	0.01	+10% relative
N	0.01	+0.0005
Fe	0.01	+0.001
Ni	0.01	+0.001
Si	0.01	+0.001

Vacancy interactions with LEs
DFT calculations

X	E_m^X (eV)	$E_b^{V-X_1}$ (eV)	$E_{diss}^{V-X_1}$ (eV)
H	0.21 (TIS-TIS) [24]	1.24 [25]	1.45
C	1.46 (TIS-OIS) [20]	1.93 [20]	3.39
N	0.73 (TIS-OIS) [26]	2.48 [21]	3.21
O	0.17 (TIS-TIS) [22]	3.05 [22]	3.22

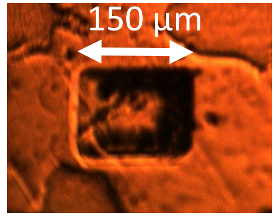


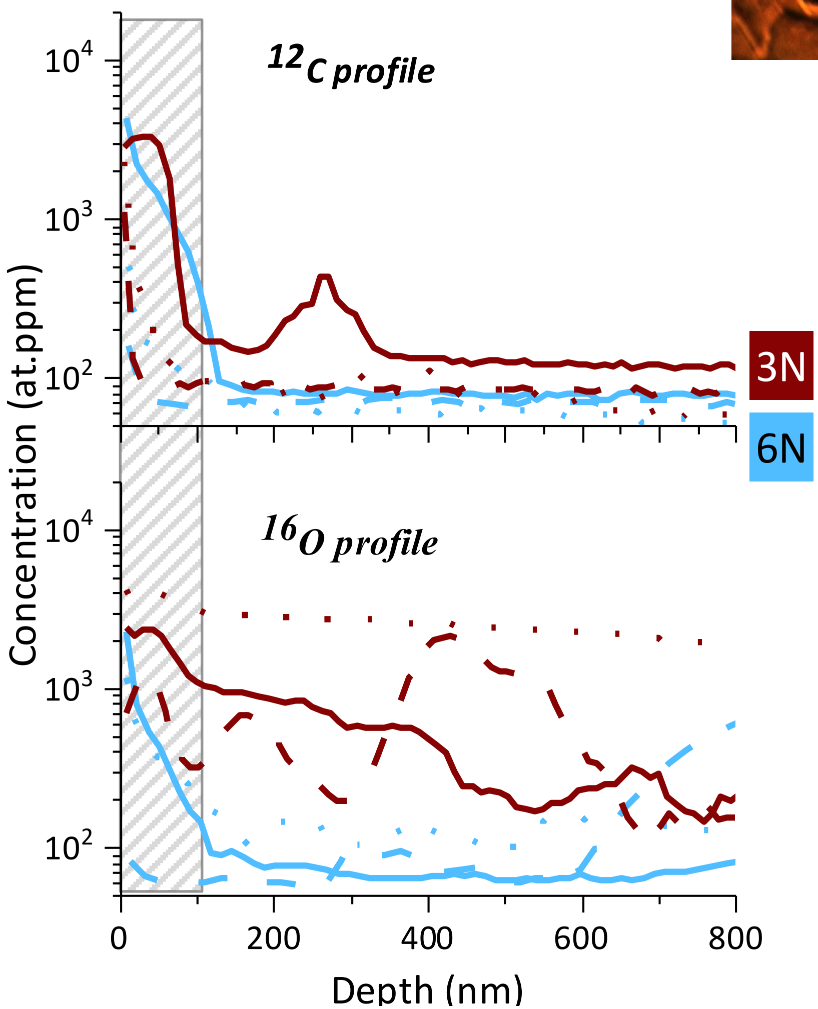
Light element impurities (H,C,N,O..) **defects**(single vacancy, vacancy-complexes and SIA).

[1] N. Castin, et al J. Nucl. Mater. 527 (2019); 1 .Hirai, T. et al.. Nuclear Materials and Energy 9, 616–622 (2016). 2 Greenwood.al 212–215, 635–639 (1994). 3 *Computational Materials Science* **184**, 109932 (2020). 3 Liu. Computational Materials Science 50, 3213–3217 (2011). 4 You, Y.-W. et al.. RSC Adv. 5, 23261–23270 (2015). 5 Alkhamees, A. et al.. Journal of Nuclear Materials 393, 508–512 (2009). 6 A. Vehanen et al Phys Rev B 25 (1982) 762

Effect of purity

SIMS : 3 craters





For 2 purities 3N 99.95% and 6N 99.9999%

- Surface pollution (0- ~100nm)
- Heterogeneous distribution specially for oxygen in 3N

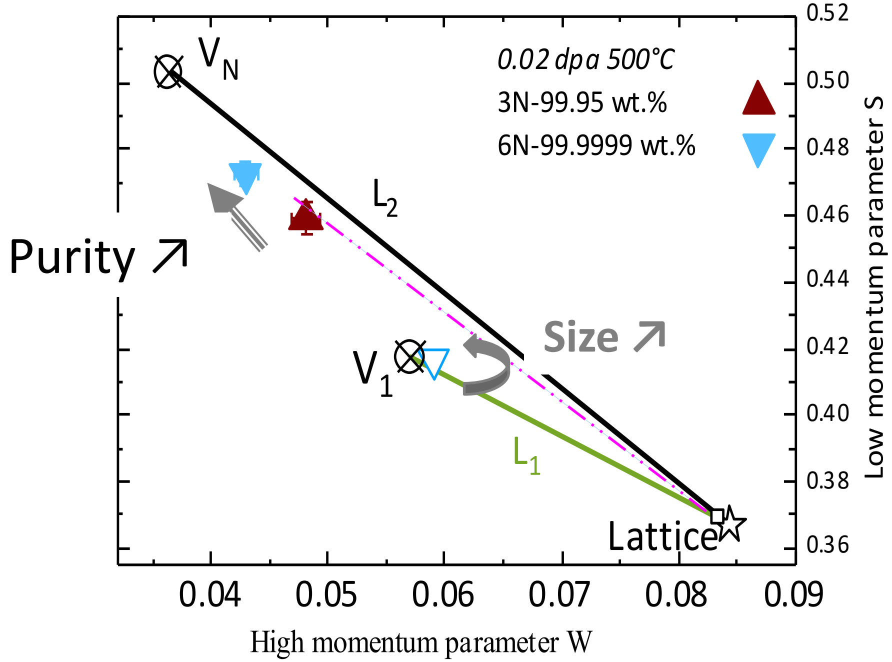
	Concentration (at. ppm)		
	3N 99.95%		6N 99.9999%
H	910 [a]		Ud
C	460 [a]	**~ 90**	**~ < 65**
N	130 [a]		Ud
O	345 [a]	**1200** [105 : 3000]	**~ 100**

a/ provided concentration by supplier corresponding to detection limits

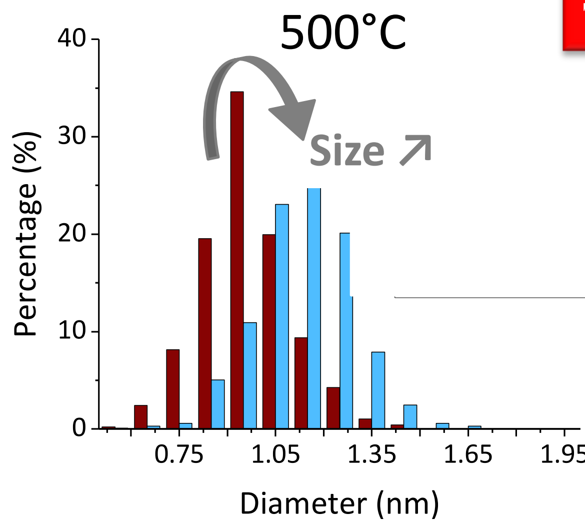
Effect of purity

PAS-DB

Self-irradiated W ions 0.02 dpa*, 500°C



TEM



Damage level (dpa)	0.02	
Irradiation Temp. (°C)	500	
Purity	**3N**	**6N**
Mean diameter(nm)	**0.97 ± 0.19**	**1.13 ± 0.21**
Density (10^{24} m^{-3})	**1.73 ± 0.71**	**1.56 ± 0.52**
Swelling (%)	**0.09 ± 0.02**	**0.12 ± 0.04**

Increase of swelling when purity ↗

- More large vacancy clusters when purity ↗

➔ **WHY ?**

Effect of purity

- TEM confirms PAS results

Formation of smaller clusters in HP: why?

Vacancy mobile at 500 °C → agglomeration to form larger vacancy clusters

but in 3N, this agglomeration is prevented : what could limit it?

	Concentration (at. ppm)			E_m^X	$E_b^{V_1-X_1}$	$E_{diss}^{V_1-X_1}$
	3N		6N	(eV)	(eV)	(eV)
H	910 [a]	~ 90 [b]	Ud	0.21 (TIS-TIS) [1]	1.24 [1]	1.45
C	460 [a]		~ < 65 [b]	1.46 (TIS-OIS) [2]	1.93 [2]	3.39
N	130 [a]		Ud	0.73 (TIS-OIS) [3]	2.48 [3]	3.21
O	345 [a]	**1200** [105 : 3000] [b]	~ 100[b]	0.17 (TIS-TIS) [4]	3.05 [4]	3.22

a/ provided concentration by supplier corresponding to detection limits, ***b/*** measured value by SIMS

→ formation of V-complexes (V_m-C_n, V_m-O_n) in 3N sample?

In particular V_mO_n

1 Yang, L. et al.Computational Materials Science 184, 109932 (2020). 2 Liu, Y.-L et al. Computational Materials Science 50, 3213–3217 (2011). 3 You, Y.-W. et al.. RSC Adv. 5, 23261–23270 (2015).
4 Alkhamees, A. et al. Journal of Nuclear Materials 437, 6–10 (2013). 357-63

Summary

TEM and PAS complementarity : PAS (Single vacancy → small cavities), TEM (cavities > 0.5-1 nm)

Self-irradiation to mimic neutron irradiation

SIMS to analyse C and C in W

Irradiation at RT :

- majority of single vacancies and small clusters
- saturation at 0.5 dpa (~ 50 days in Tokamak)

For irradiation temperature = 500°C

- Vacancy clusters size increases allowing diffusion of vacancies and their agglomeration
- More clusters in pure samples → larger swelling

→ Vacancy-Oxygen complexes limits V diffusion and agglomeration

Perspectives :

Saturation of damage at high temperature ??

microstructure using PAS TEM and DRX?

Thanks:

- CEMHTI : Z. Hu (phD), C. Genevois, P. Desgardin, J. Joseph
- GEMAC : F. Jomard
- JANNuS Orsay : B. Décamps, C. Baumier, Accelerator Team
- JANNuS Saclay : M. Loyer-Prost, Accelerator Team
- ETHZ : R. Schaublin

Thank you for your attention