DE LA RECHERCHE À L'INDUSTRIE

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IMPACT OF HELIUM IRRADIATION ON TRITIUM RETENTION IN TUNGSTEN

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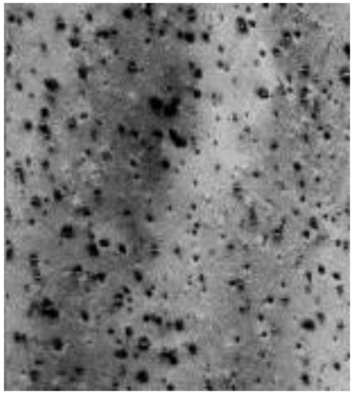
INTRODUCTION AND CONTEXT

TUNGSTEN AS A PLASMA-FACING MATERIAL

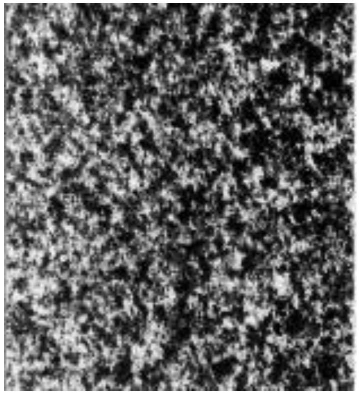
- W: used in JET, WEST, **ITER**
  - → intensive fluxes of He and H isotopes at high temperatures
- Impact of He irradiation at the surface
  - dislocation loops
  - bubbles
  - W-fuzz

} <530ºC, $E < E_{disp.\ min}$ (538 eV)

W irradiated with 6.5×10^{19} $H.m^{-2}$



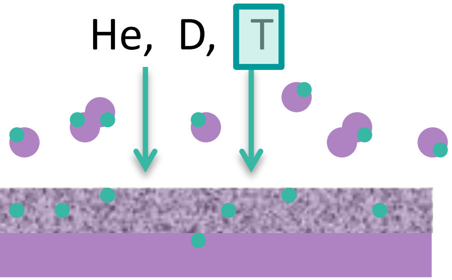
W irradiated with 6.5×10^{19} $He.m^{-2}$



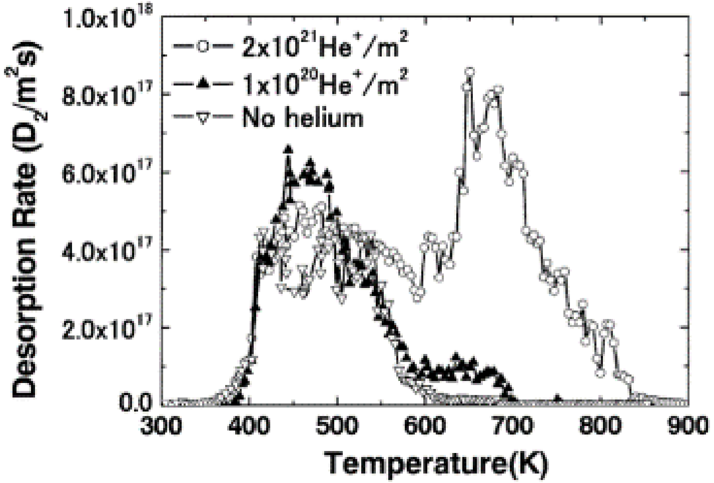
R. Sakamoto

➡ ***He has a strong impact on the material.***

➡ ***These modifications can affect the trapping of hydrogen (T).***



H. Iwakiri et al., JNM **2002**, 307-311, *135-138*.



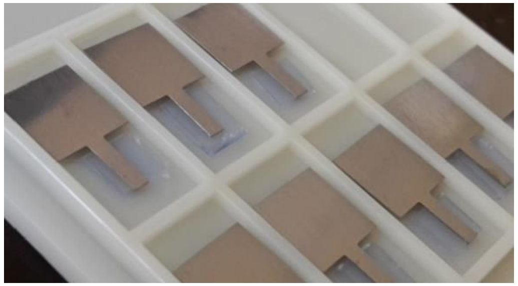
SUMMARY

- He irradiation: experimental setups
  - In situ conditions: LHD exposures
  - Parametric study: PSI-2 exposures
- Micro and nano structure change of tungsten under helium irradiation
  - TEM observation and results
  - Larger scale characterization: TEM – GISAXS coupling
  - Defects created in the material: PAS
- Impact of He irradiation on T inventory:
  - Tritium gas loading on massive W samples
  - Parametric study
- Conclusions and perspectives

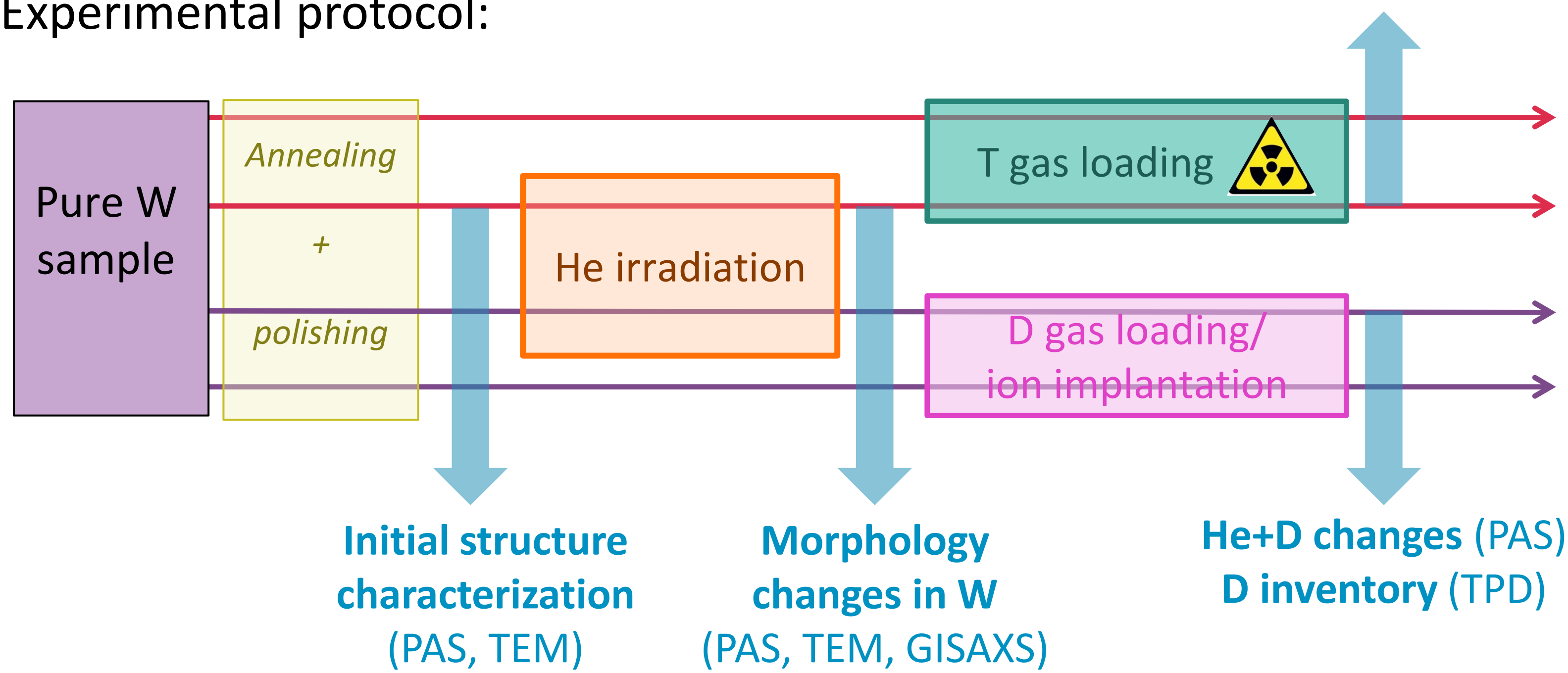
EXPERIMENTAL PROTOCOL AND IRRADIATION SETUPS

MATERIAL PREPARATION AND EXPERIMENTAL PROTOCOL

- Goal: **ideal state of the W structure before He irradiation**
  - → *Easier evaluation and understanding of mechanisms at stake*
  - → *Comparative study with "as received" W (with defects caused by the manufacturing processes)*
    - Tohoku Kinzoku pure W
    - 1500° C annealing for 2h
    - Mechanical polishing

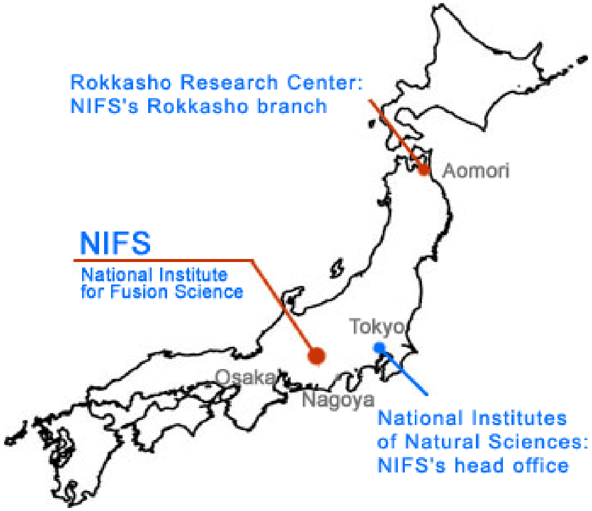


- Experimental protocol:



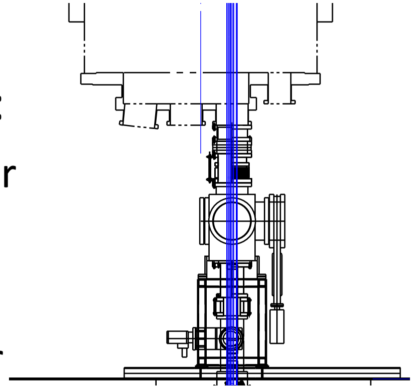
IN SITU EXPOSURE: THE LARGE HELICAL DEVICE (LHD)

- LHD and the retractable material probe:
  - LHD: Large Helical Device, a superconducting stellarator with heliotron configuration
    - A pair of continuous winding helical coils and 3 pairs of poloidal coils
    - R=3.9 m, a_{eff}=0.63, V_p~30m^3, B_T~3 T
  - Net-current free plasma with various heating methods (NBI, ECH and ICH)



➡ Observe real plasma-wall interaction conditions and complement them with laboratory exposures

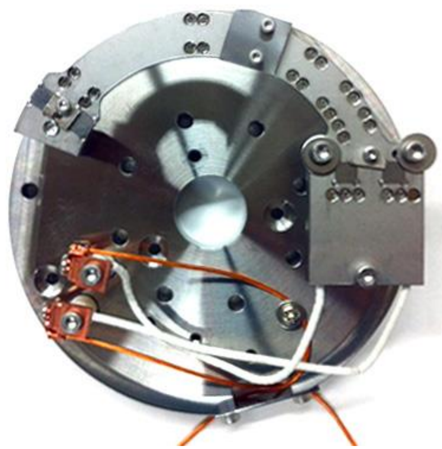
- Two retractable material probes installed for PWI studies:
  - Plasma exposure under various conditions: SOL plasma, divertor leg, CX particles
  - Electric feedthrough for controlling temperature
  - Motion feedthrough for controlling exposure condition (shutter and insertion)

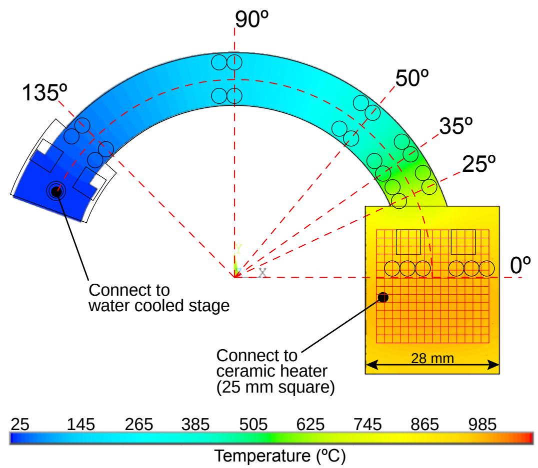


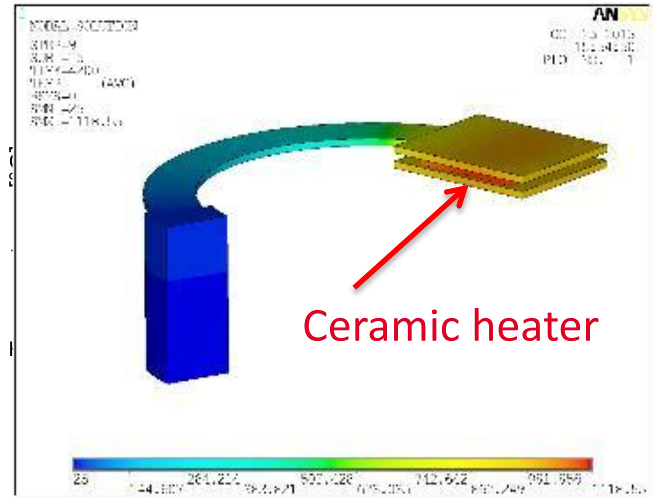
IN SITU HIGH TEMPERATURE EXPOSURES: EXPERIMENTAL SETUP

- Holder 1: *17th LHD experimental campaign*

  Multi temperature exposure using temperature gradient between a heater and heat sink, **65 - 600 ° C**





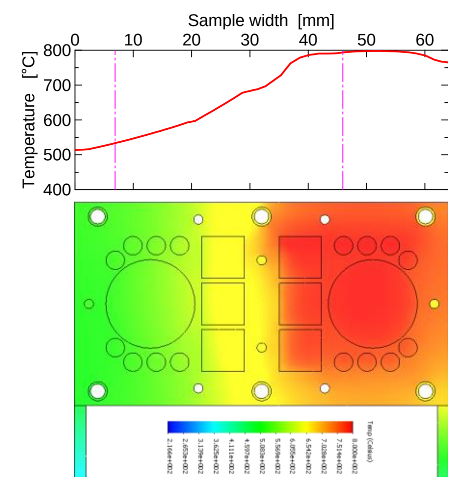


- Holder 2:

  18th LHD experimental campaign

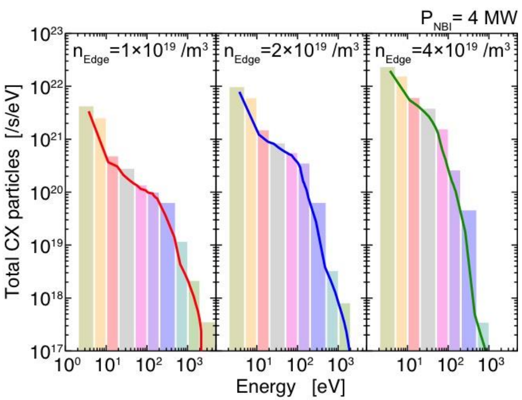
  Higher temperature exposure with reduced thermal loss structure, **500 - 800 ° C**

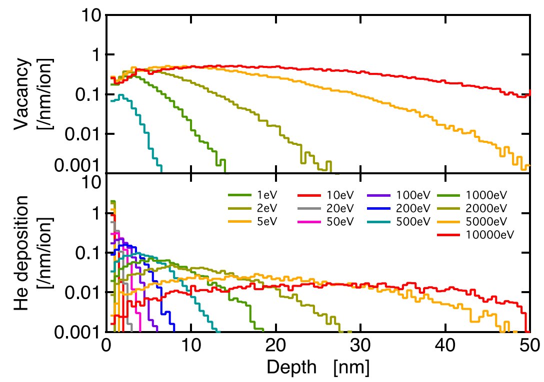
  - Pre-thinned samples for TEM + large samples for Temperature Programmed Desorption (TPD)

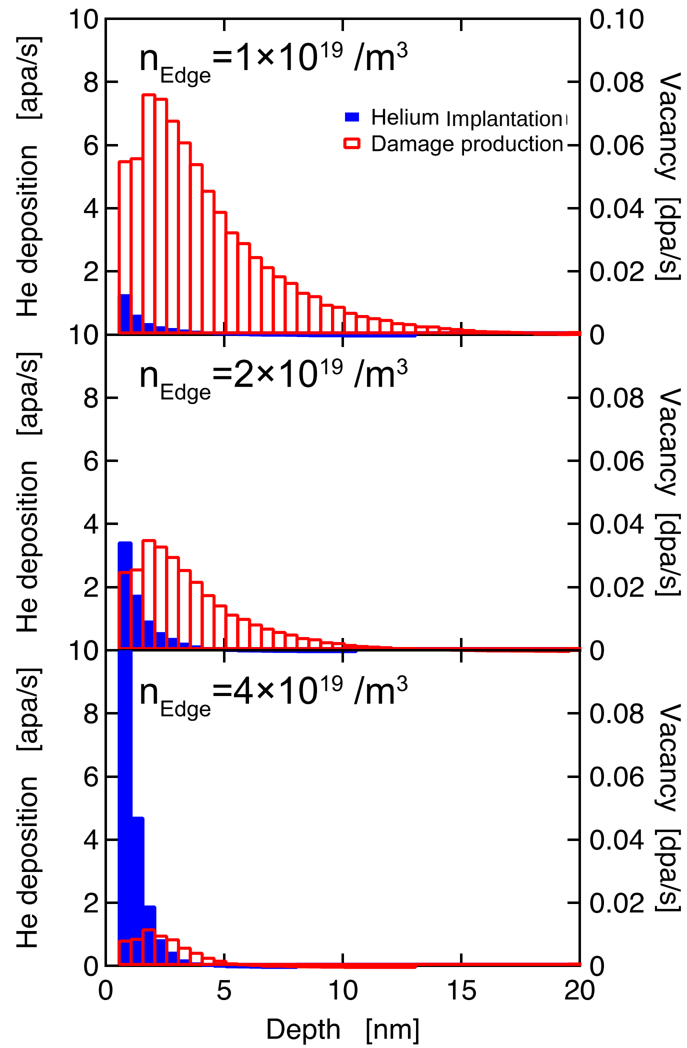


HELIUM IRRADIATION CONDITIONS IN LHD

- Goal: expose to CX particles inserting the retractable material probe at the 1st wall position
  - ➡ Sequences of 100 shots of NBI heated plasma discharges with helium gas puff
- Estimation of radiation damage profiles created at plasma-facing surface





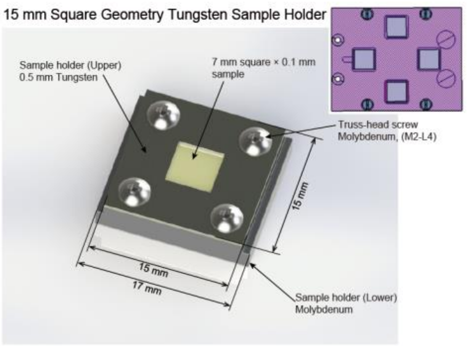


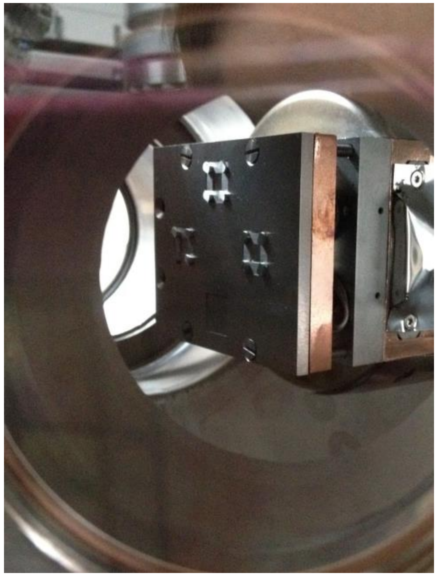
- ➢ 10^{22}-10^{23} He.m^{-2} fluences
- ➢ 10^{21}-10^{22} He.m^{-2}.s^{-1} flux
- ➢ More helium implantation than displacement damage is expected

ACCESSING A LARGER RANGE OF PARAMETERS: PSI-2

- PSI-2 (Jülich)
  - **A plasma-Surface Interaction laboratory device:**

    Stationnary plasma produced in a low pressure high-current arc discharge and directed toward a biased, temperature-controlled sample holder





  - **Conditions:**

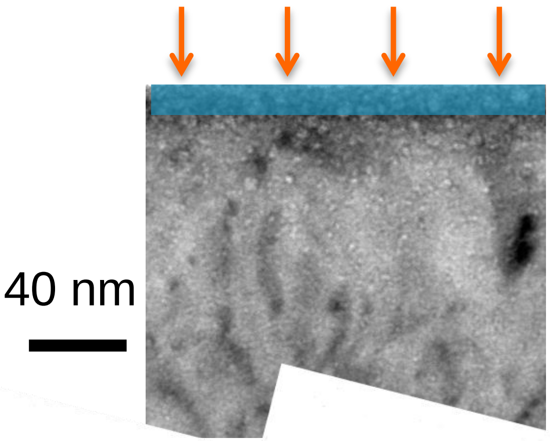
    - 75 eV He plasma, 100 V bias
    - Temperature: 523 to 1073 K
    - Flux : $2.5x10^{20}$ to $2.5x10^{22}$ $s^{-1}.m^{-2}$
    - Fluences : $3x10^{23}$ to $1x10^{26}$ m^{-2}

Sample	W_{ref}	18-AL	20-CL	21-BL	21-AL	20-BL
T (K)	/	473	1023	1053	1073	473
Flux ($He.m^{-2}.s^{-1}$)	/	$2.2x10^{22}$	$2x10^{22}$	$2.3x10^{22}$	$2.9x10^{20}$	$2.2x10^{22}$
Fluence ($He.m^{-2}$)	/	$1x10^{26}$	$3x10^{25}$	$3x10^{23}$	$3x10^{23}$	$3x10^{23}$

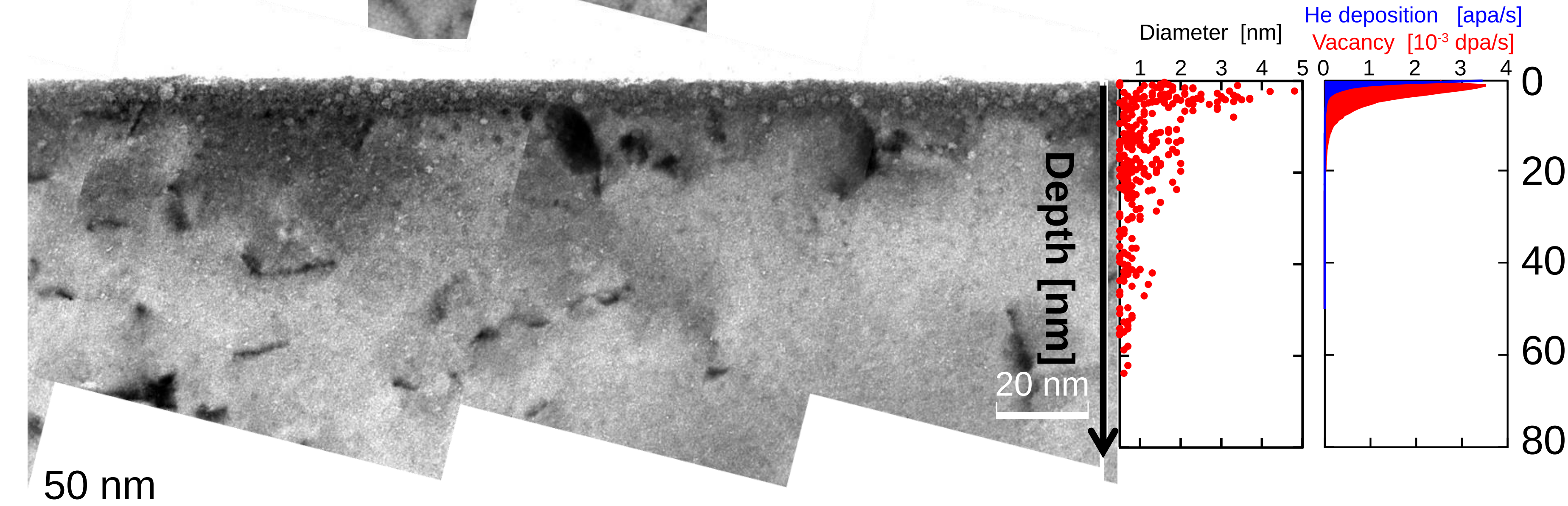
STRUCTURE CHANGES IN W CAUSED BY HE EXPOSURE

LHD exposed samples

TEM+FIB



Heavily damaged layer formed at the surface (dislocation loops + bubbles)
➔ gets thicker as He fluence increases



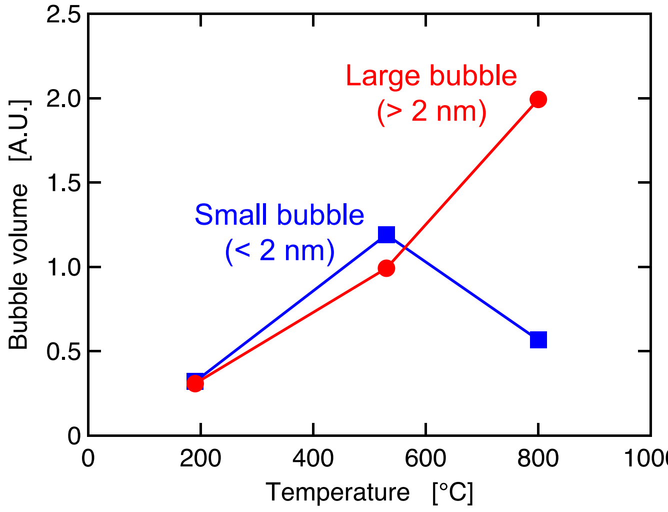
800 ° C LHD18_FR1

DAMAGE DISTRIBUTION

LHD exposed samples

TEM+FIB

- Bubble formation
  - **Distributes up to 70 nm depth at any temperature range.**
    - Bubble depth distribution is insensitive to vacancy mobility.
      - supported by theoretical simulations by A.M. Ito and R. Kobayashi.

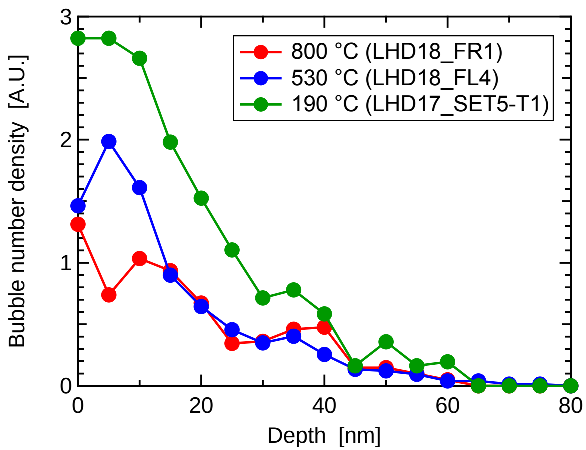


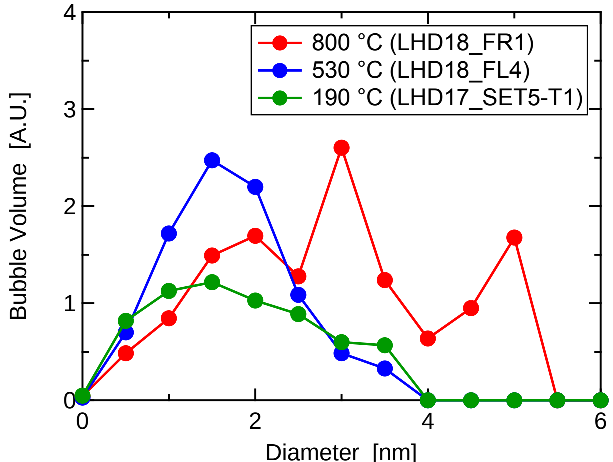
  - Irradiation temperature ↗

➡ Size ↗ and density ↘

- Vacancy: 1,7 eV
- He: 0.06 eV

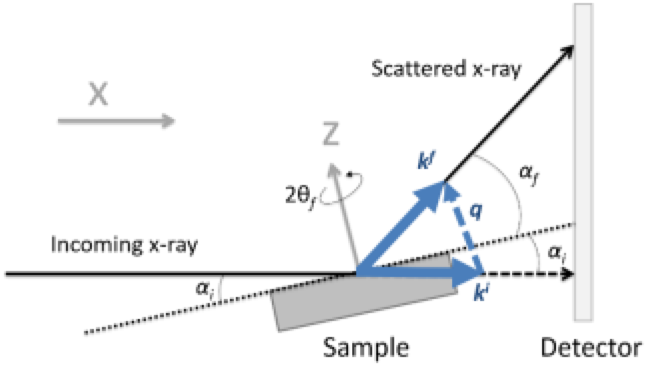
➢ He migration at any temperature range but vacancy migration above 500ºC.

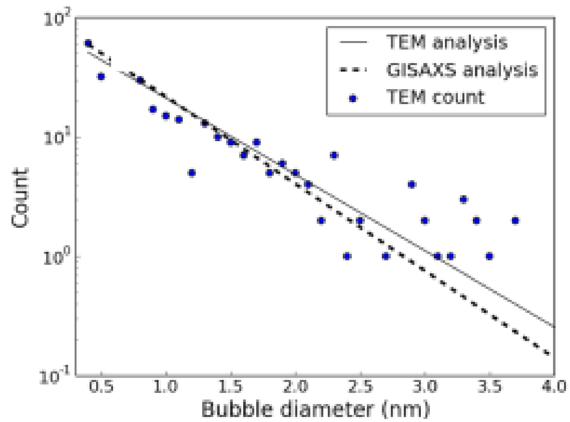


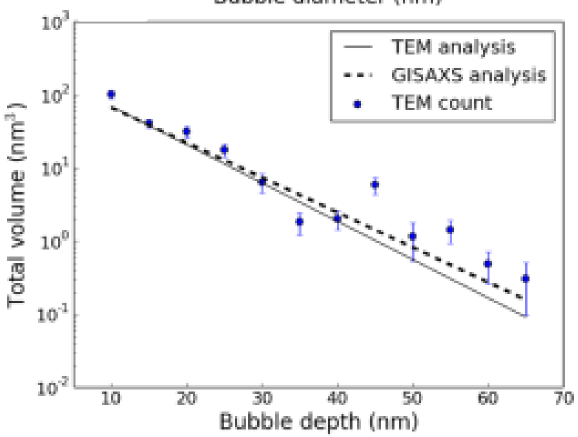


LHD exposed samples

GISAXS







- Grazing-incidence Small Angle Scattering:
  - non destructive technique using a photons probe to study nanostructure materials, combining the length scales of small-angle scattering and surface sensitivity of grazing incidence diffraction.
  - ➢ ideal complement for TEM: determines average particle properties on a larger scale.

➡ Excellent agreement of the results on bubble characterization:

- Nano-bubbles range from spheroidal to ellipsoidal, displaying exponential diameter distributions with mean diameters μ:

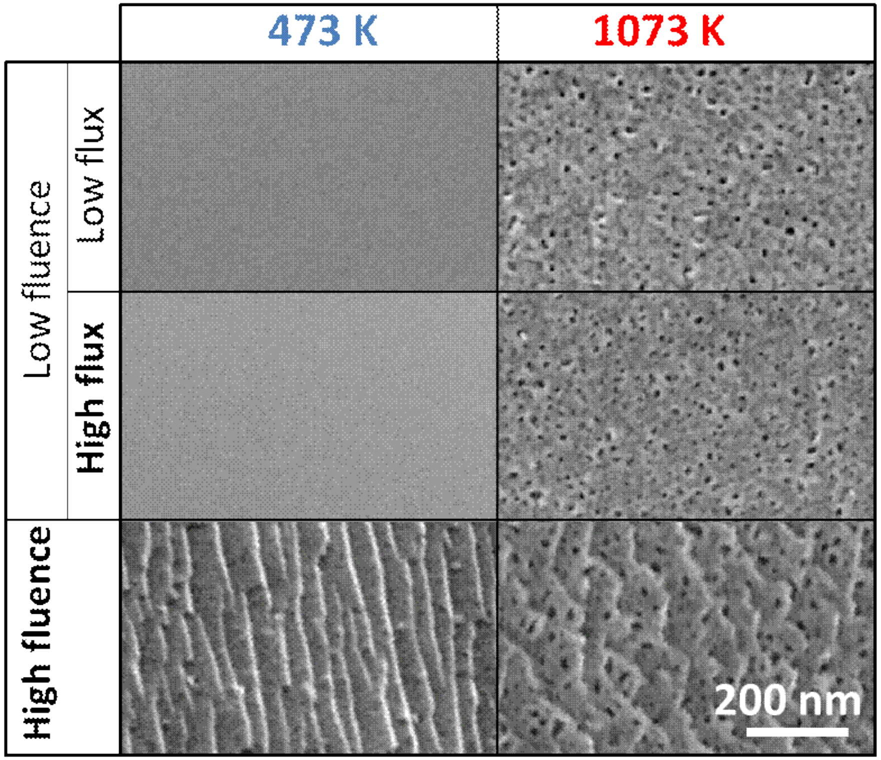
  $\mu_{TEM}=0.596 \pm 0.001$ nm

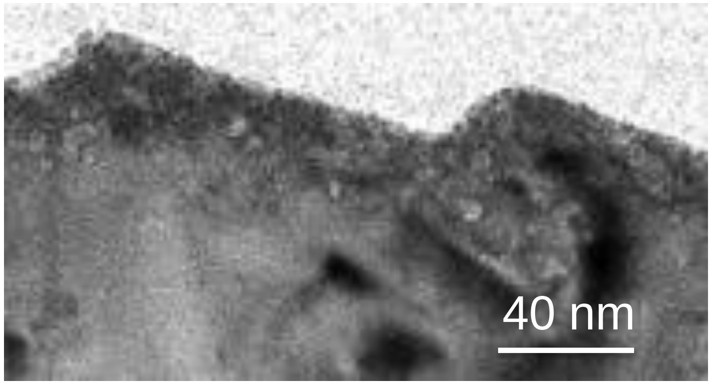
  $\mu_{GISAXS}=0.68 \pm 0.04$ nm

- Depth distributions were also computed, with calculated exponential depth distributions with mean depths r:

  $r_{TEM}=8.4 \pm 0.5$ nm

  $r_{GISAXS}=9.1 \pm 0.04$ nm





PSI-2 exposed samples

TEM+FIB

- **Surface state:**
  - No flux dependence of the damage structure
  - Significant change of the surface morphology: holes, fuzz

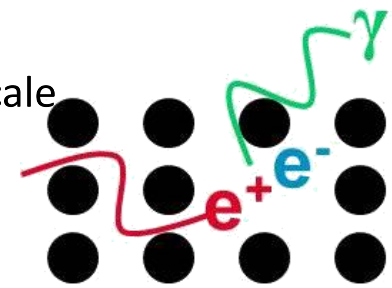
  -> Overall roughness increase

    - ➢ **E_{He}<$E_{sputtering\ threshold}$ yet material erosion?**
  - **Undulating surface structure for high fluence**

- **Cross-sectional study:**
  - Large flattened bubbles (>10 nm) at 1073 K
  - Surface undulation aligning with <100> direction

- Positron Annihilation Spectroscopy (PAS)
  - Non-destructive technique to characterize defects at the atomic scale located close to the surface

    ➔ **Detect presence and free volume of defects**

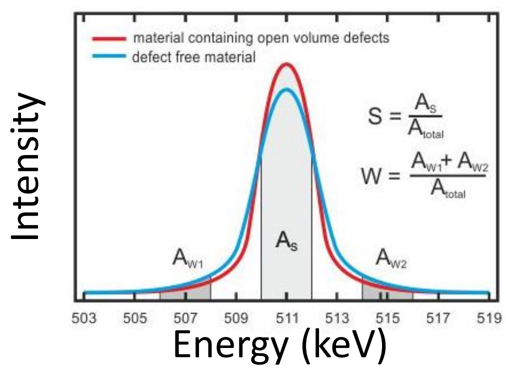


  - Traps for positrons (e+): low electronic density areas

    monovacancy (V) vacancy cluster (nV)

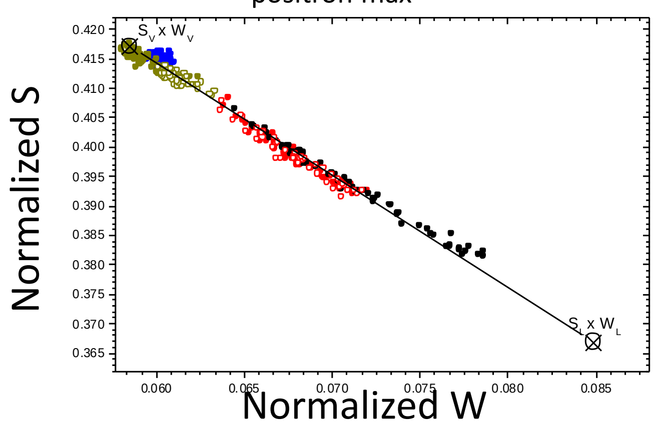
    He bubbles in W nV-mHe complexes

    - timelife of the positron ➔ presence of defects
    - positron energy ➔ scanned depth
    - type of annihilation ➔ electronic state



  - CEMHTI:

    ➔ $E_{positron\ max}$ = 541 keV, i.e. ~70 µm depth scanning in W



Nilaco W, as received

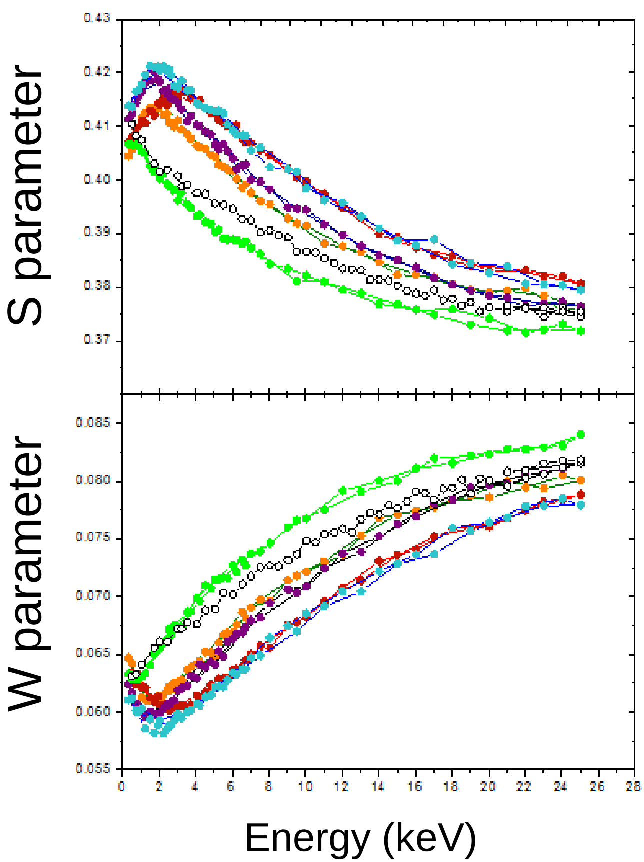
Nilaco W, 1273 K 3h annealing, MP

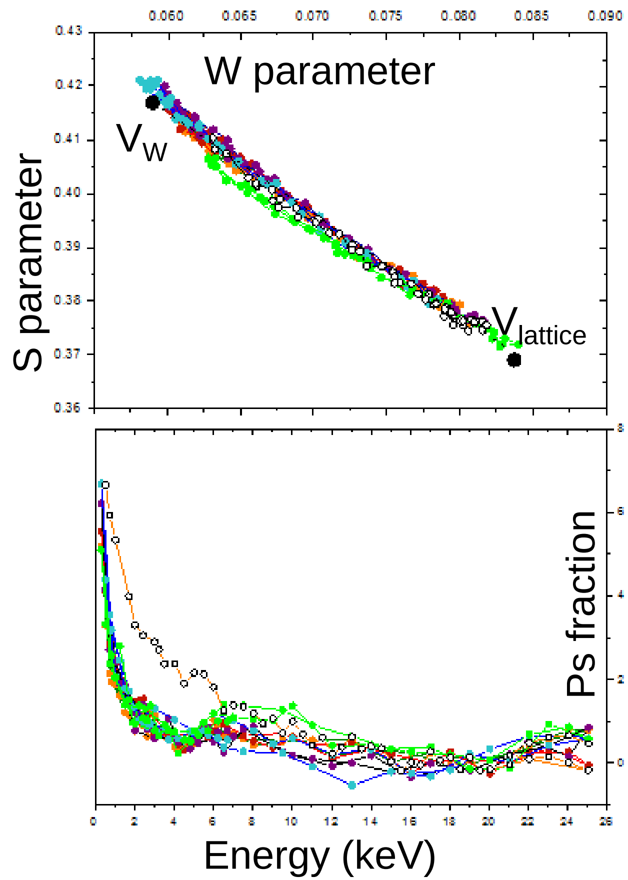
Allied Materials W, 1573 K 2h annealing MP

WPhy05, 1873 K 1h vacuum annealing

➔ Great variation between materials: major role of W preparation process and pre characterization

DEFECTS CREATED IN THE W STRUCTURE: PAS





Sample	W_{ref}	18-AL	20-CL	21-BL	21-AL	20-BL
T (K)	/	473	1023	1053	1073	473
Flux ($He.m^{-2}.s^{-1}$)	/	$2.2x10^{22}$	$2x10^{22}$	$2.3x10^{22}$	$2.9x10^{20}$	$2.2x10^{22}$
Fluence ($He.m^{-2}$)	/	$1x10^{26}$	$3x10^{25}$	$3x10^{23}$	$3x10^{23}$	$3x10^{23}$

PSI-2 exposed samples

PAS

- **W_{ref}: no defect**
- Free volume traps for positrons detected under 2 keV (0-10 nm depth)
  - **Traps in the heavily damaged layer**
- Free volume ~ monovacancy
  - **He is present in the bubbles observed**
- **20-BL (low temperature, low fluence): free volumes smaller than monovacancy**
  - **nV-mHe with m>n**
    - **low tempeterature: no vacancy cluster?**
    - **low exposure time: little He migration?**

IMPACT ON TRITIUM RETENTION IN W

TRITIUM GAS LOADING

- Tritium handling at Saclay Tritium Lab
  - Technique: Gas overpressure at high temperature
    - **no damage** or defect created in the material structure
    - Tritium study:
      - + High sensitivity
      - + Mixed H/D/T exposures

- W experiments:
  - Gas loading procedure:
    - Oxyde reduction
    - Tritium gas loading:

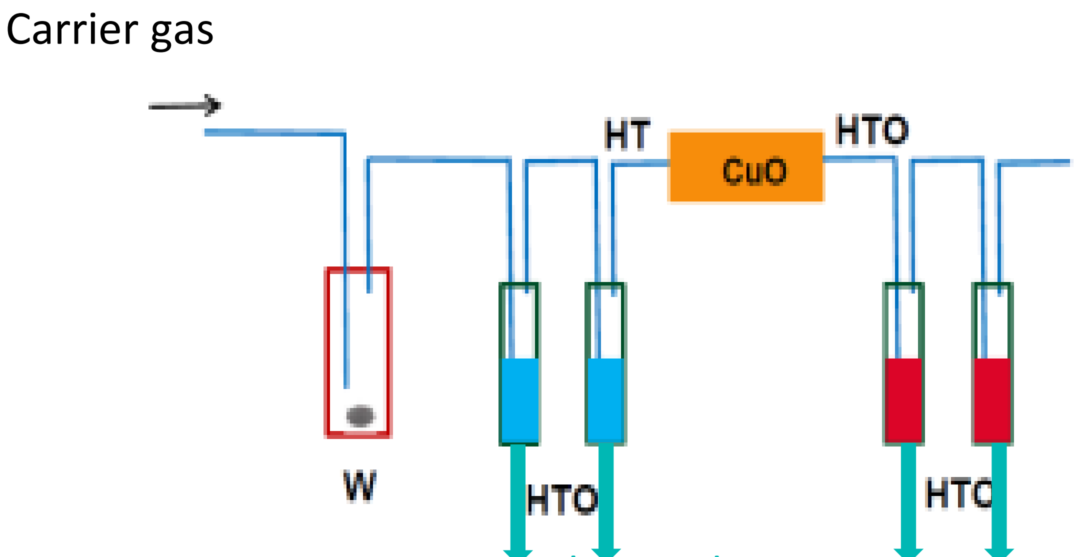
      2h at 450ºC under 1 bar $^3H_2$
  - T room temperature desorption
  - T high temperature desorption (800° C)
  - Sample dissolution for in-bulk T counting

} Liquid Scintillation Counting (LSC)

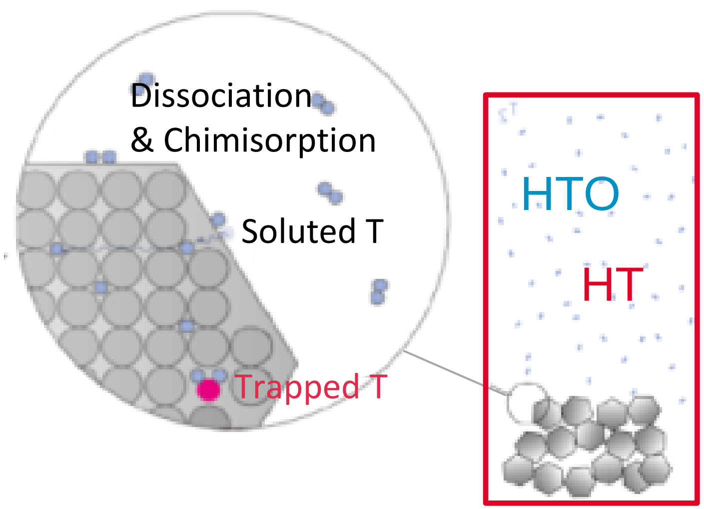
Active material:
D loading bench to couple results with surface analyses

- **Global T inventory in the sample**

TRITIUM DESORPTION ANALYSIS



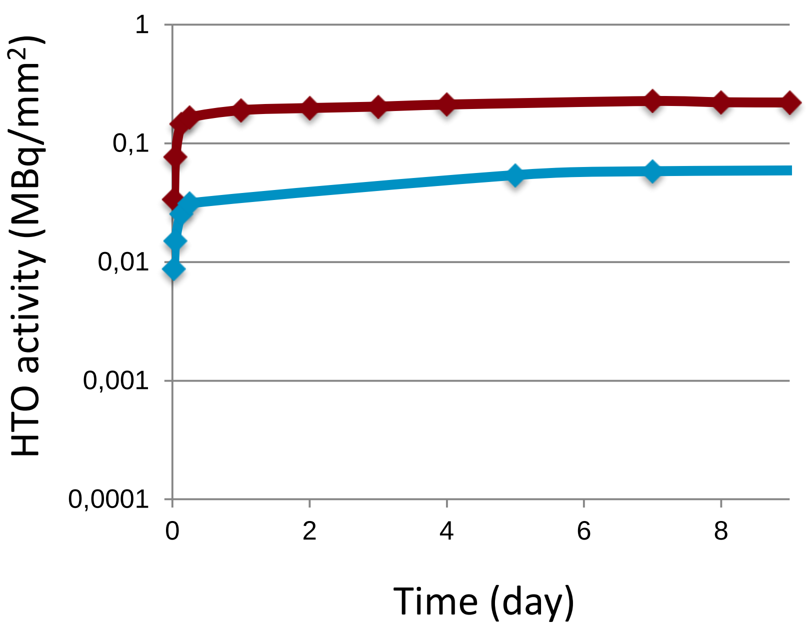
Sampling and LSC at t=0, 30 min, 1h, 3h, 1 day…
+ 250, 500, 800ºC
+ Bulk sample dissolution and LSC

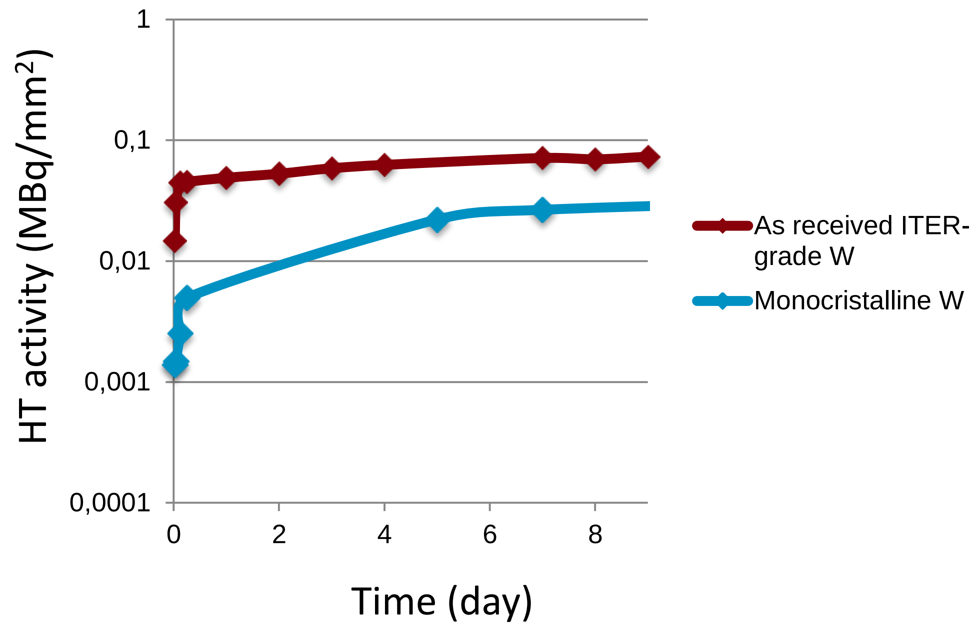


➡ **T global inventory with additional information on traps for H in W:**

- HT/HTO desorption: trap nature
- Time/temperature desorption: trapping energy
- T/D/H mix loading: isotopic preferential trapping?

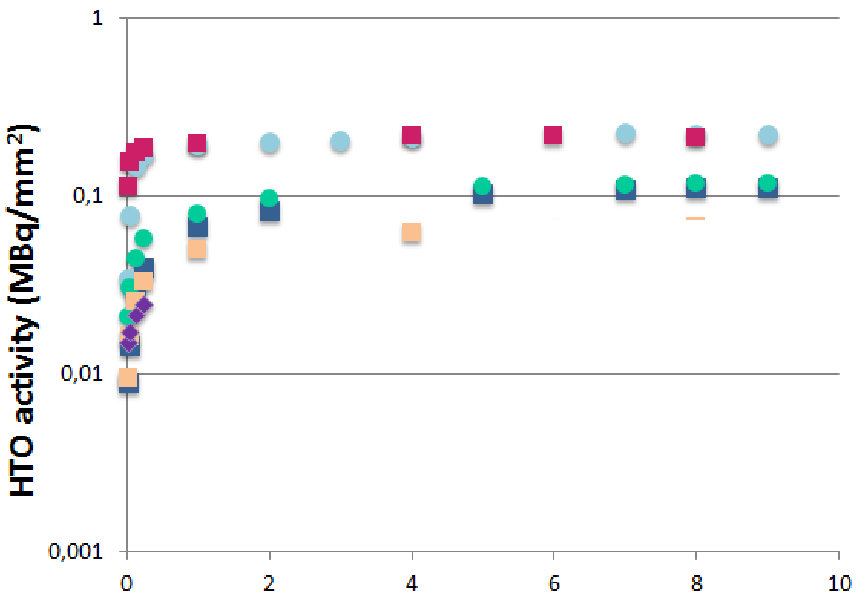
TRITIUM RETENTION IN PRISTINE SAMPLES

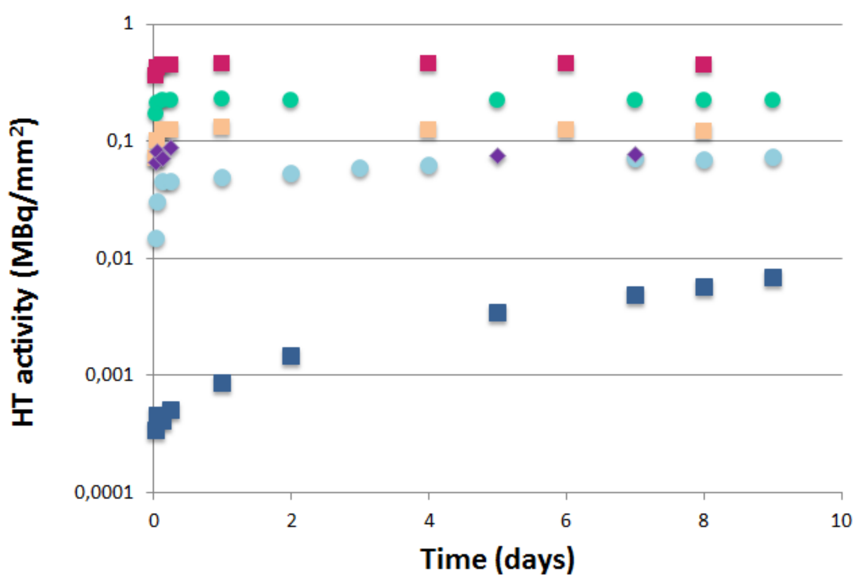




- Pristine W: T inventory is mainly linked with HTO desorption
- Grain boundaries and existing defects in ITER grade W are additional traps for T retention compared to monocristalline W
  - Initial W structure plays a major role in retention studies
- More pristine W structures need to be studied

IRRADIATION IN PSI-2: IMPACT ON T LOADING





- As-received W
- Annealed W: pristine
- Annealed W: 800ºC – 3×10^{20} $s^{-1}.m^{-2}$ - 3×10^{23} m^{-2}
- Annealed W: 800ºC – 2×10^{22} $s^{-1}.m^{-2}$ - 3×10^{25} m^{-2}
- Annealed W: 800ºC – 2×10^{22} $s^{-1}.m^{-2}$ - 3×10^{23} m^{-2}
- Annealed W: 300ºC – 2×10^{22} $s^{-1}.m^{-2}$ - 1×10^{26} m^{-2}

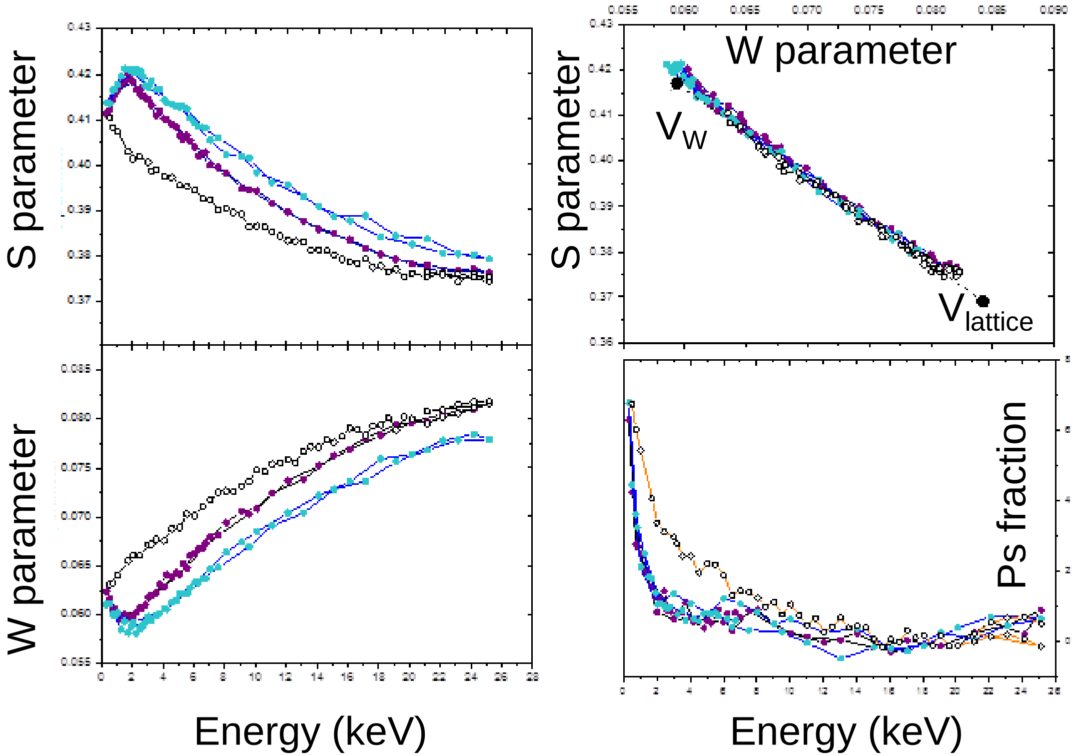
Tº *Flux* *Fluence*

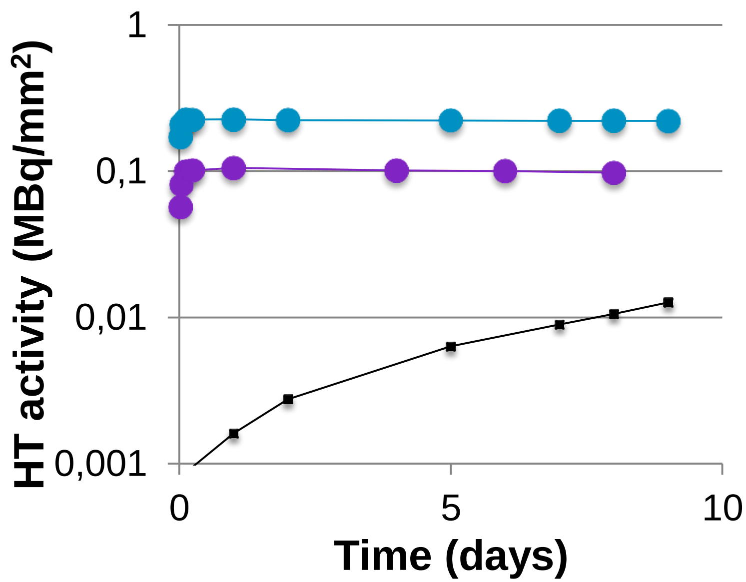
- **Annealing decreases traps for T in W:**
  - **Pre existing traps in industrial W**
- **All He irradiation increase T trapping sites in the material**
- **T inventory increase linked with HT desorption: drastic increase compared to pristine W**
  - **Additional and specific trapping sites for T due to the He bubbles?**

PARAMETRIC STUDY: IMPACT OF HE FLUX

21-AL
1073 K
$3x10^{20}$ He.m^{-2}.s^{-1}
$3x10^{23}$ He.m^{-2}

21-BL
1053 K
$2x10^{22}$ He.m^{-2}.s^{-1}
$3x10^{23}$ He.m^{-2}

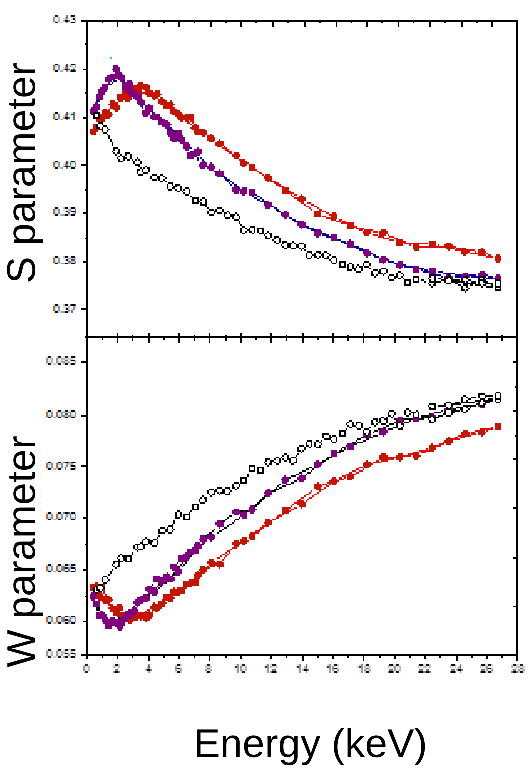


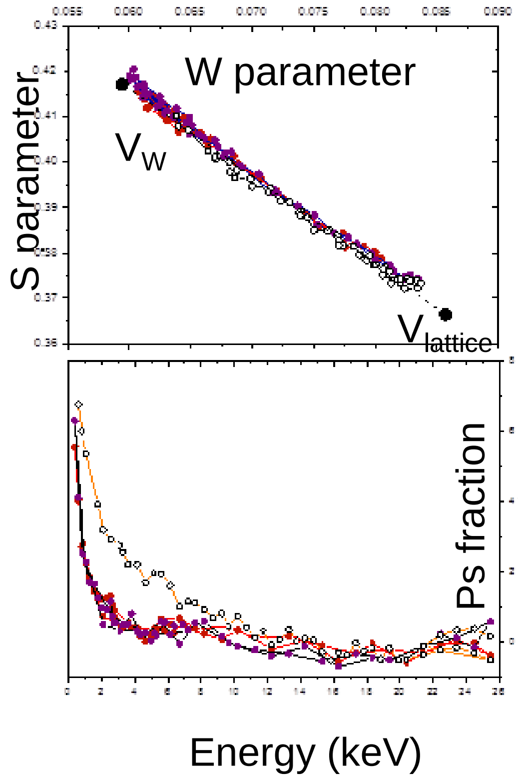


- He flux↗➔Thinner surface layer impacted
- He flux ↗ ➔T trapping sites ↘

Less traps when He flux↗, i.e. exposure time ↘: diffusional process?

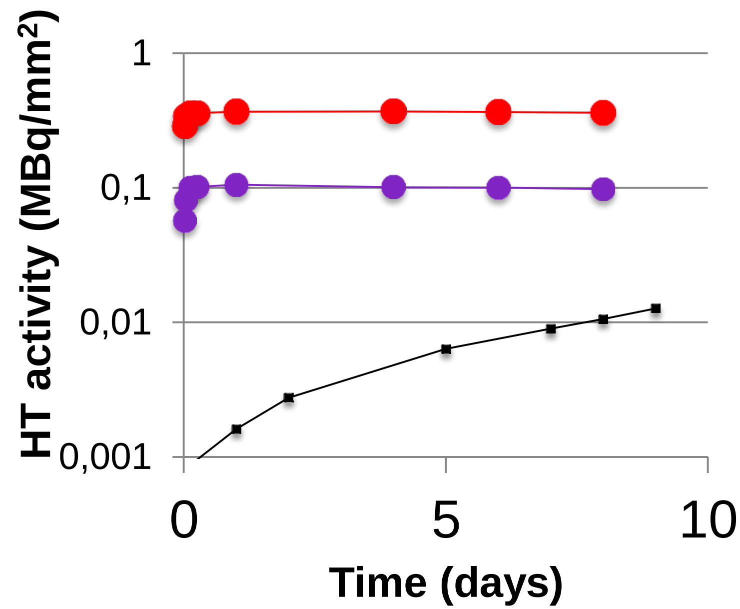
PARAMETRIC STUDY: IMPACT OF HE FLUENCE





20-CL
1023 K
$2x10^{22}$ He.m^{-2}.s^{-1}
$3x10^{25}$ He.m^{-2}

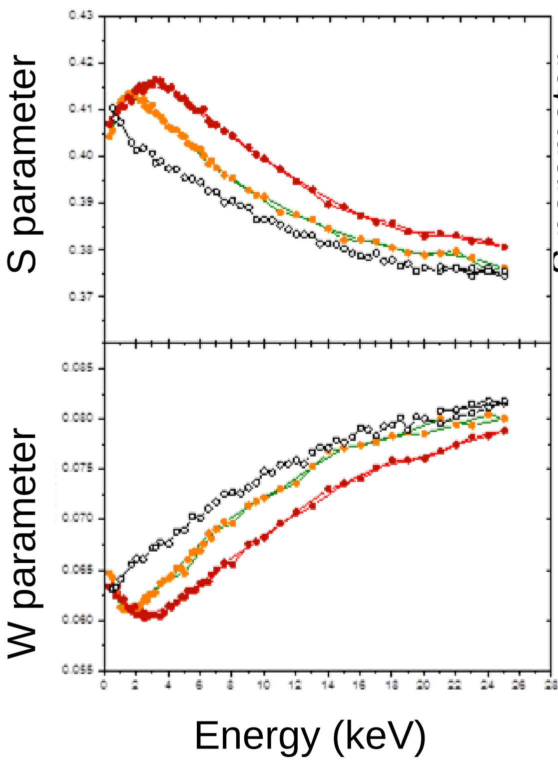
21-BL
1053 K
$2x10^{22}$ He.m^{-2}.s^{-1}
$3x10^{23}$ He.m^{-2}

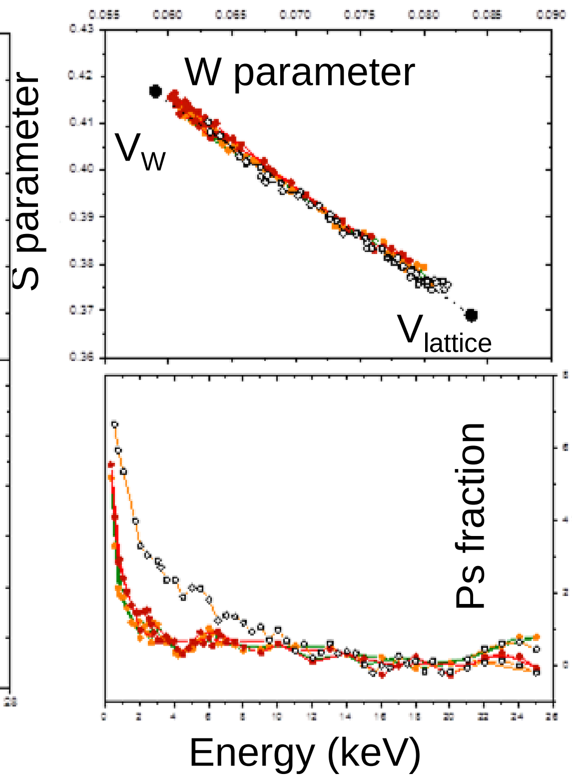


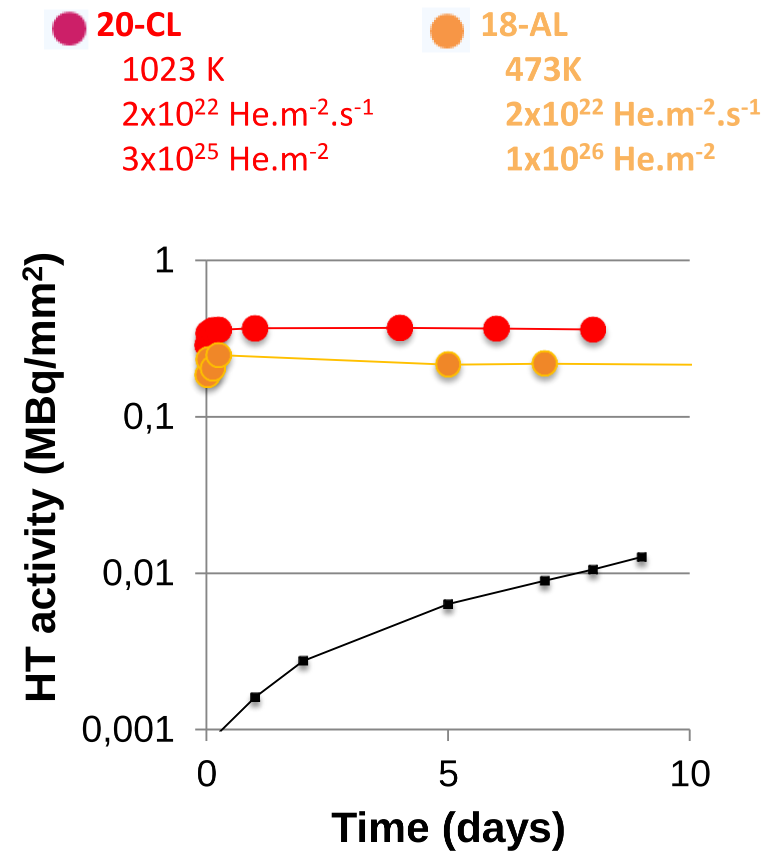
- He fluence ↗ ➔ Free volumes of defects ↗ and defects located deeper
- He fluence ↗ ➔ T trapping sites ↗

➡ ***Traps and defects increase with the incident He fluence***

PARAMETRIC STUDY: IMPACT OF TEMPERATURE







- T° ↗ ➔ Deeper annihilation peak
- T° ↗ above 773 K ➔ Free volumes get larger than monovacancy reference: m/n<1
- T° ↗ ➔ T trapping sites ↗

Temperature is a crucial parameter: vacancy mobility > 773 K ?

CONCLUSION AND PERSPECTIVES

CONCLUSIONS

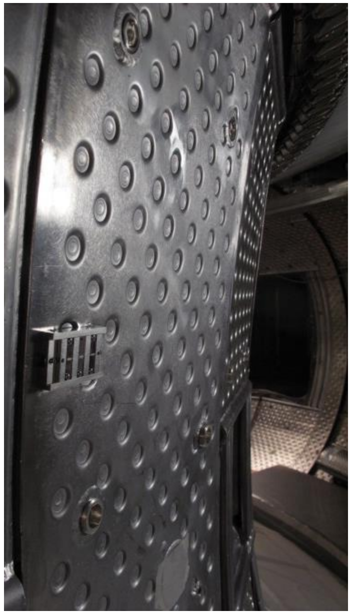
- Significance of **in situ exposures** experiment at **high temperatures**
  - *Complementary to laboratory studies*

- Cross-sectional TEM observation:
  - Heavily damaged layer formed at the surface, whose thickness increases with temperature.
  - Bubbles are distributed deep in W well beyond He implantation range (<15 nm) at any temperature range.
    - Helium accumulation supports bubble nucleation, without vacancy.
  - Coalescence of bubbles (larger and sparser) as temperature increases.
    - Vacancy enhances the growth of bubbles.
  - *Nano-bubbles characteristics in agreement with GISACS experiments*

- Hydrogen retention in W
  - Increase of D/T retention in defect-rich materials:
    - non annealed W
    - He-irradiated W
  - **Increase of T trapping when He fluence increases**
  - **Increase of T trapping when He flux increases ➡ Diffusion-like process**
  - W coated materials: both surface and bulk materials impact the retention.

PERSPECTIVES

- Extend samples panel:
  - Focus on surface impact: native oxides, rugosity (PhD)
  - W « WEST-like »
  - Deposited materials: W-W, Si-W (Polimi), CFC-W (JET)



- Extend irradiation ranges:
  - He ion irradiation in CAMITER (PIIM, AMU)
  - New campaigns at PSI-2 (FZJ)
    - Extend irradiation range: fluence, impurities, He+D plasmas…
  - **WEST: exposure to CX He during C4 (2018)**

- Extend the hydrogen retention study:
  - D ion irradiation and TDP: CAMITER
  - D gas loading surface analyses (Raman, EDX…)
  - T/D/H gas loading: penning jauge under way at Saclay Tritium Lab

Thank you for your attention

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