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1st International Conference on Fusion Energy Materials Science

Comparison of deuterium retention for ion-irradiated and neutron-irradiated tungsten

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Objective

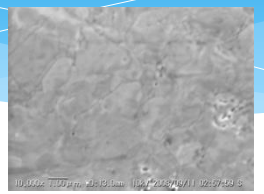
Background

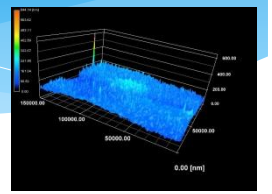
- *Plasma facing components research for fusion applications has recently focused on tungsten materials due to low sputtering yield and low tritium retention. One of the key issues under performance evaluation of plasma facing materials is tritium retention and recycling under fusion relevant conditions.*
- *Recently, high energy ion irradiation experiment was performed on tungsten to demonstrate neutron irradiation effect on tritium retention.*

Objective of this research

- The objective of this research is to understand the tritium behavior in neutron-irradiated materials for fusion first walls (trapping of tritium by defects formed by neutron irradiation) under Task2-1 (Irradiation-Tritium synergism) of Japan-US collaboration program (TITAN).
- Comparison of tritium behavior in ion irradiated materials will help the understanding of neutron irradiation effects on tritium retention behavior.
- Simulation research works using TMAP and Monte Carlo code are also performed to understand the neutron irradiation effect on tritium behavior.

Research Scheme





Sample preparation

Allied Material Co. Ltd (stress relived)
Mechanical polish and cleaned by ultra-sonic bath
(Univ. Toyama)

Heavy ion implantation

Prepare 0.025-3 dpa samples

2.4 MeV Cu^{2+} (Kyushu Univ.)

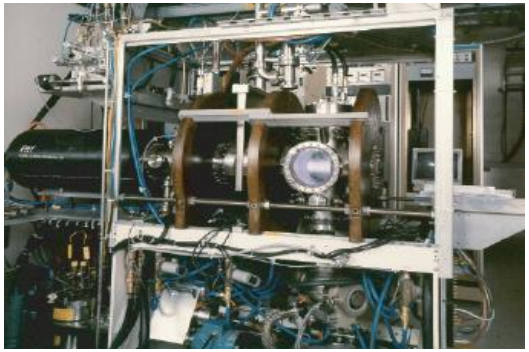
2.8 MeV Fe^{2+} (at Room temperature)
RAPID (Univ. Tokyo)

Surface morphology

TEM (Kyushu Univ.)

D plasma exposure

TPE (INL)

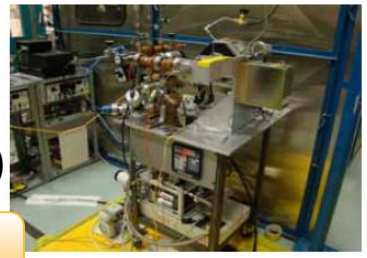


D depth profiling

NRA (Univ. Wisconsin)

D retention

TDS (INL)

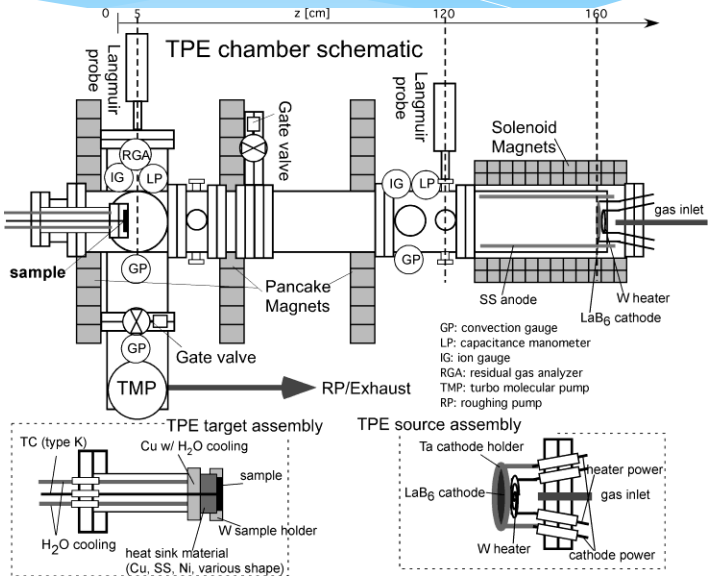


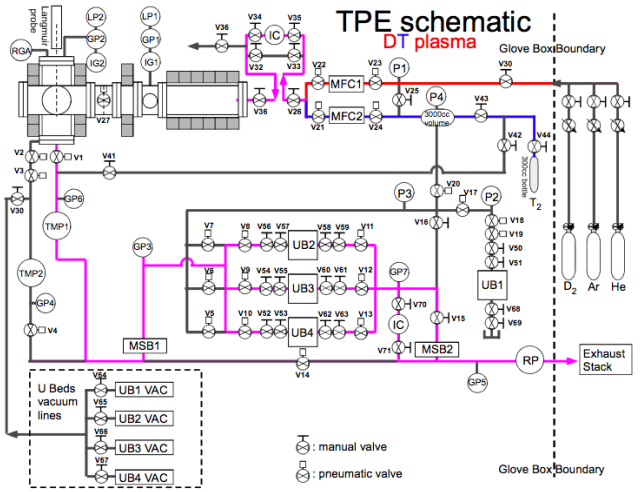
Surface morphology

SEM (Shizuoka Univ.)

Tritium Plasma Experiment (TPE) at INL

- Linear type plasma
  - LaB_6 source and actively water-cooled target
  - Steady state plasma up to high fluence (~10^{26} m^{-2})
  - High flux (~10^{22} $m^{-2}s^{-1}$), surface temp. (300~1000K)
- Tritium use:
  - Tritium inventory in STAR: <15000 Ci (1.5g)
  - (0.1 ~ 3.0 %) T_2/D_2
- Double enclosures for tritium use
  - System piping as primary confinement
  - Glovebox as a ventilation hood (second enclosure)
  - PermaCon box as a third enclosure
  - Ubeds as tritium getter
    - Estimate take of each U bed (^{238}U: 1.27kg)
    - UX_3 (x=H,D,T) ~ 8 mole of X_2
    - 1% T_2/99%D_2: 0.08 mole ~0.5g ~5000 Ci
- Activated material handling in STAR
  - Dose rate: < 100 mrem/hr (1 mSv/hr) at 30cm
  - Radioacitivity: less than HC 3 limit





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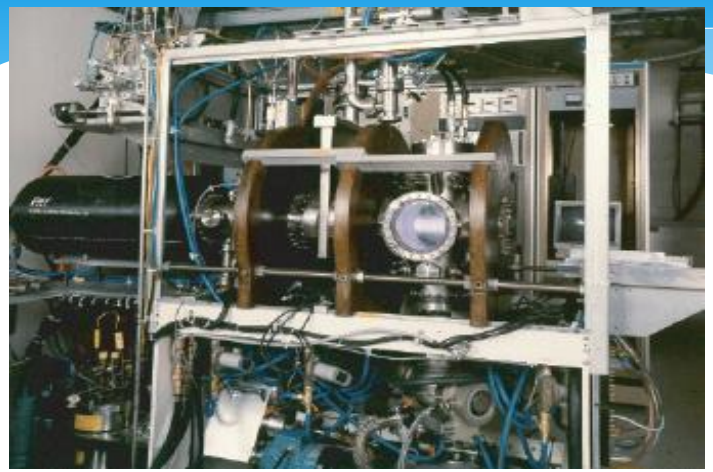
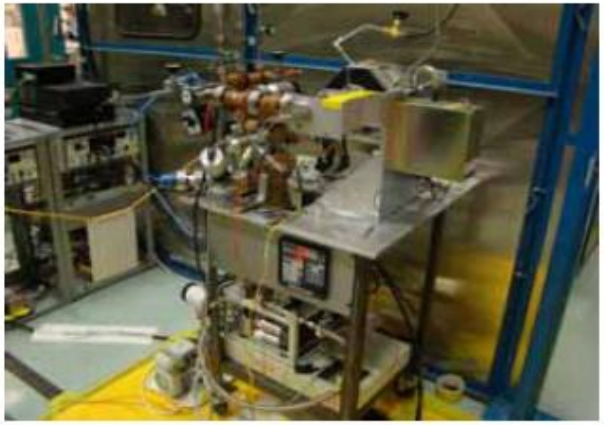
Development of experimental techniques



Sample Holder for Tritium Plasma Experiment,



Thermal Desorption System

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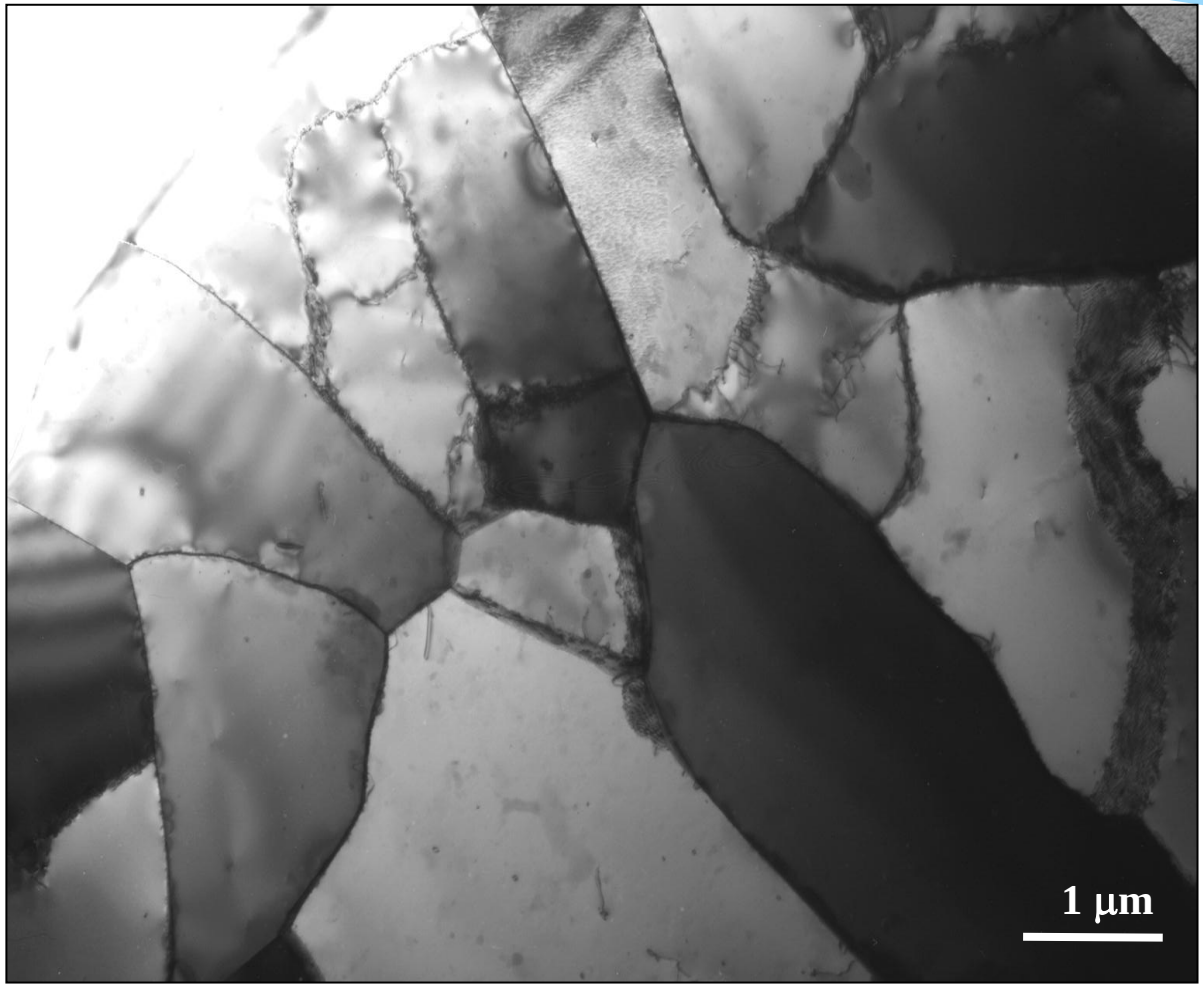
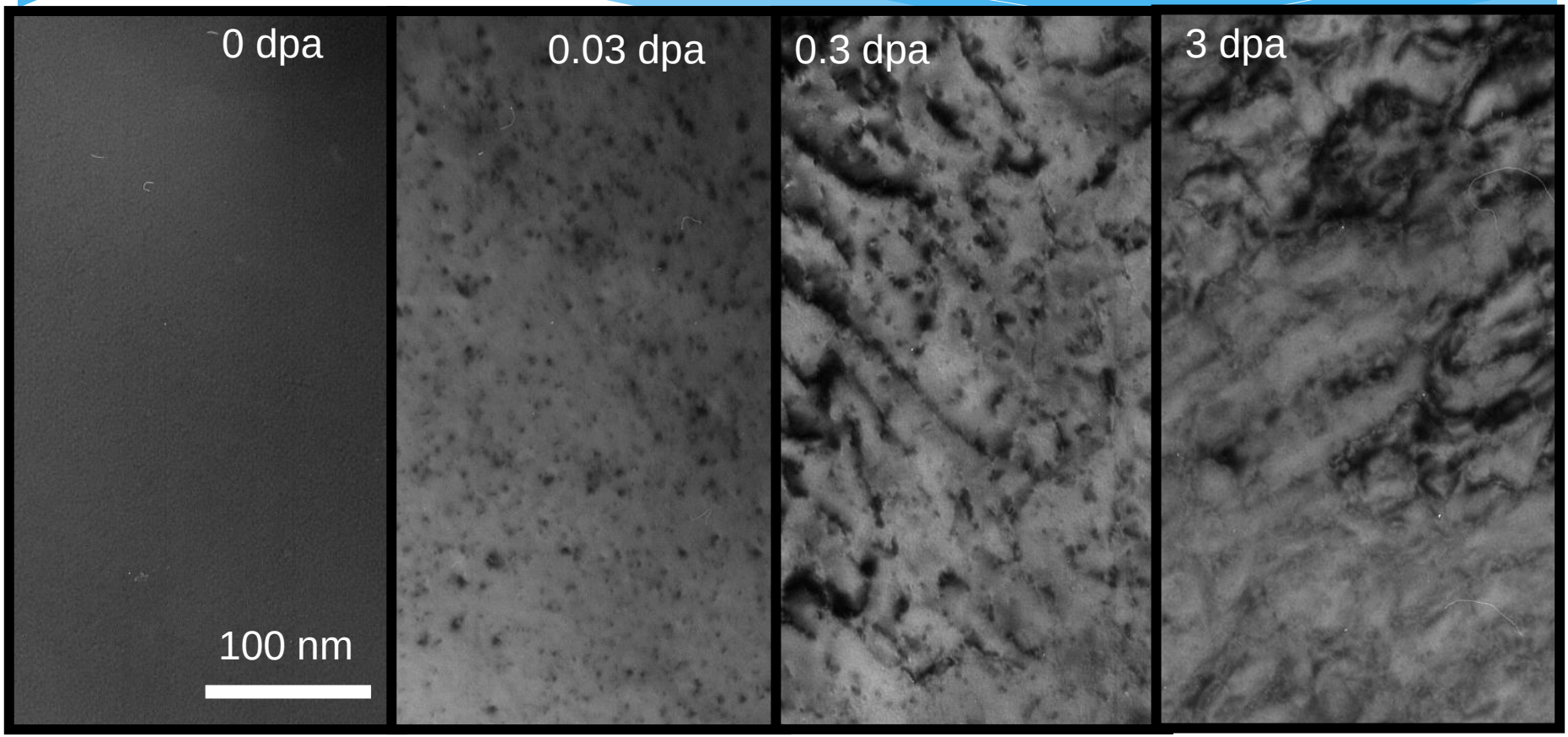
TEM image for tungsten before irradiation



W : stress relived condition
(900 K)

No defects were observed.
Only the grain boundaries were found.

Surface morphology for Cu^{2+} irradiated W observed by TEM

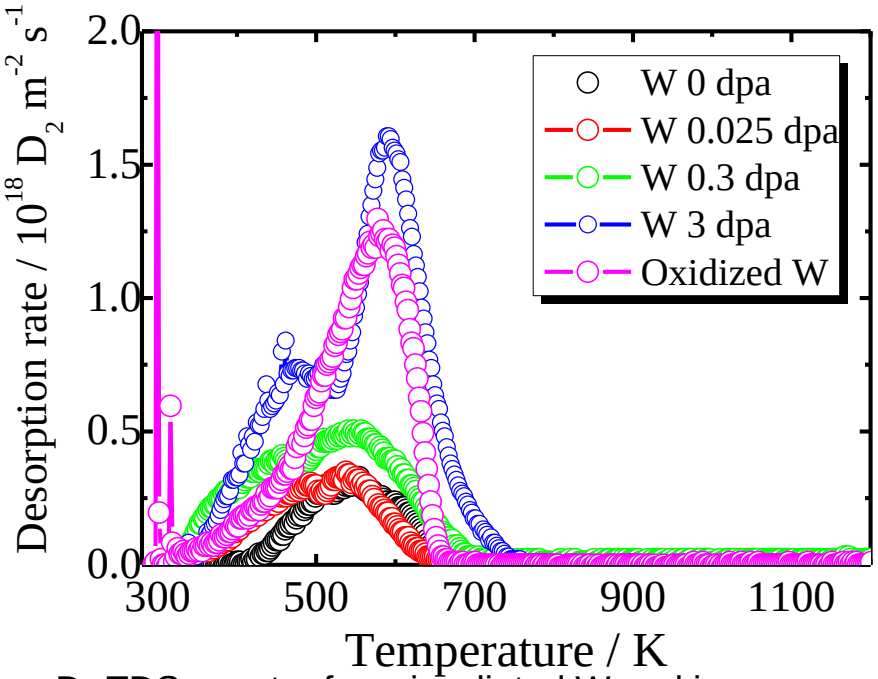


0 dpa : Only small amount of dislocation was observed.
0.03 dpa : A large number of dislocation loops with the size less than 10 nm was dispersedly confirmed.
0.3 - 3 dpa : The density of dislocation loops was enhanced as the number of dpa (irradiation fluence) was increased and the nucleation of dislocation loop was found.

Summary of D plasma exposure conditions at TPE

Sample	Fe imp W 0.025 dpa	Fe imp W 0.3 dpa	Fe imp W 3 dpa	n-irradiated W 0.025 dpa	Un-implanted W
Target current	2.3 A	2.3 A	2.8 A	2.4 A	2.6 A
Flux	9.0×10^{21}	8.2×10^{21}	1.0×10^{22}	5.0×10^{21}	6.5×10^{21}
Exposure Time	6300 s	7200 s	6300 s	7200 s	9600 s
Fluence	5.7×10^{25}	5.9×10^{25}	6.0×10^{25}	3.6×10^{25}	6.2×10^{25}
Electron density	9.6×10^{17}	7.5×10^{17}	1.1×10^{18}	4.0×10^{17}	5.4×10^{17}
Electron temperature	8.2 eV	7.7 eV	5.5 eV	9.0 eV	7.6 eV
Bias energy	-100 eV				
Ion energy	~100 eV				
Sample Temperature	473 K				

TDS spectra for neutron irradiated W and ion implanted W



D_2 TDS spectra for n-irradiated W and ion implanted W

- The peaks of TDS spectrum for ion-implanted samples were concentrated in the lower temperature less thank 700 K.

Un-irradiated sample (0 dpa)

The desorption stage was only observed at 550 K.

Ion-irradiated samples (0.025-3 dpa)

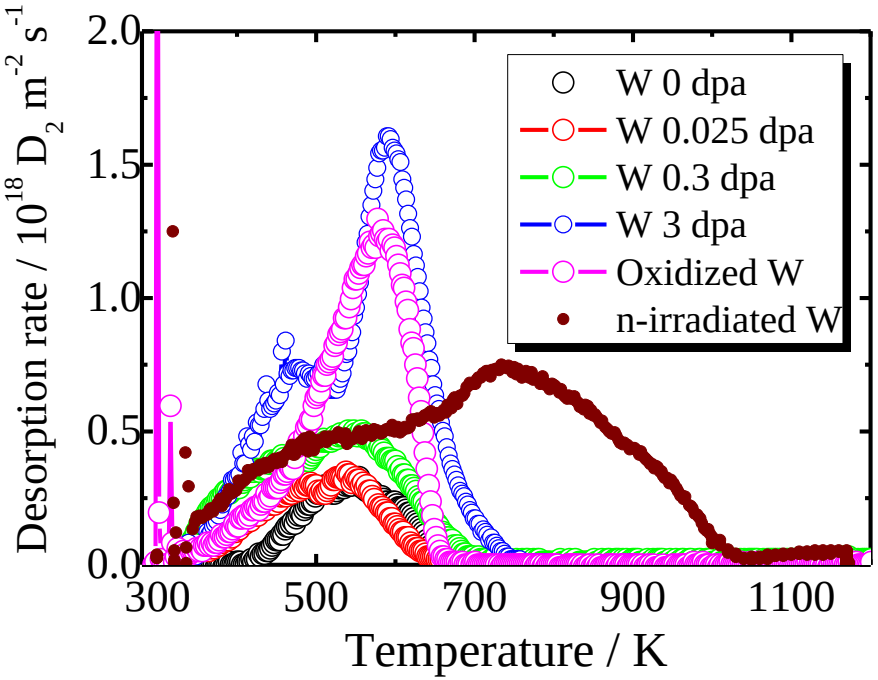
The additional peak was found at lower temperature side.

The D desorption was increased as the number of dpa increased.

The oxidized sample (673 K 2 hour)

The desorption stage was the same as the un-irradiated sample. The D accumulation was due to the surface trapping and/or diffusion barrier.

TDS spectra for neutron irradiated W and ion implanted W



D_2 TDS spectra for n-irradiated W and ion implanted W

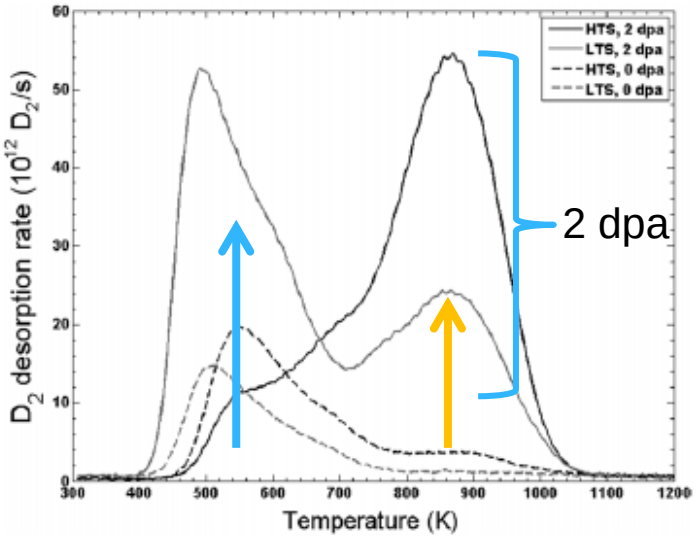


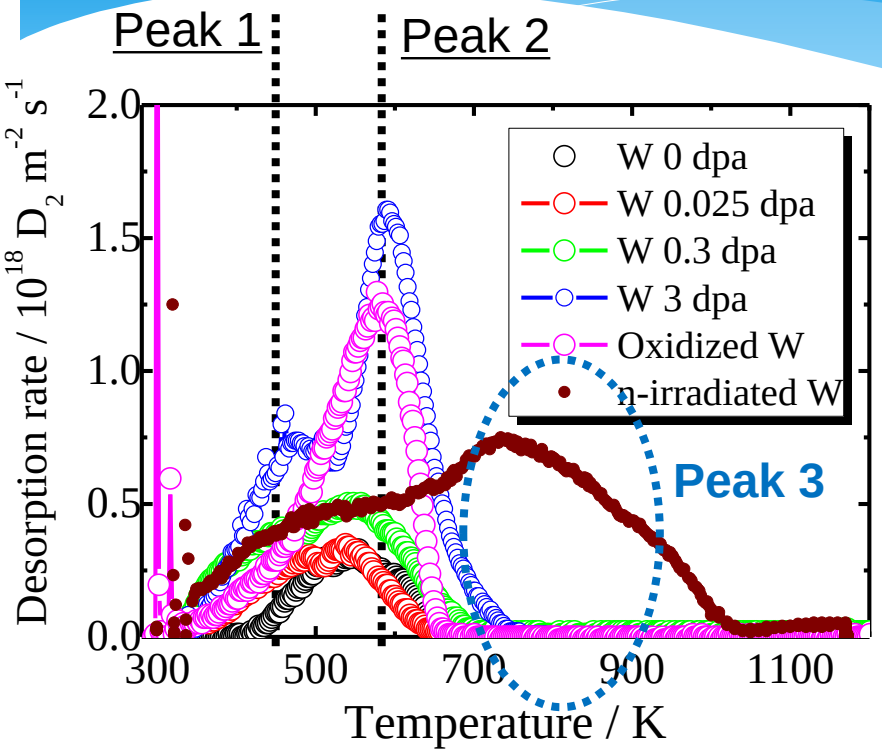
Figure 9. Thermal desorption spectra for irradiated (2 dpa) and unirradiated (0 dpa) W targets in the low-temperature (LTS) and HTSs.

- The peaks of TDS spectrum for ion-implanted samples were concentrated in the lower temperature less thank 700 K.
- It looks like the TDS spectrum for LTS sample for Reference.

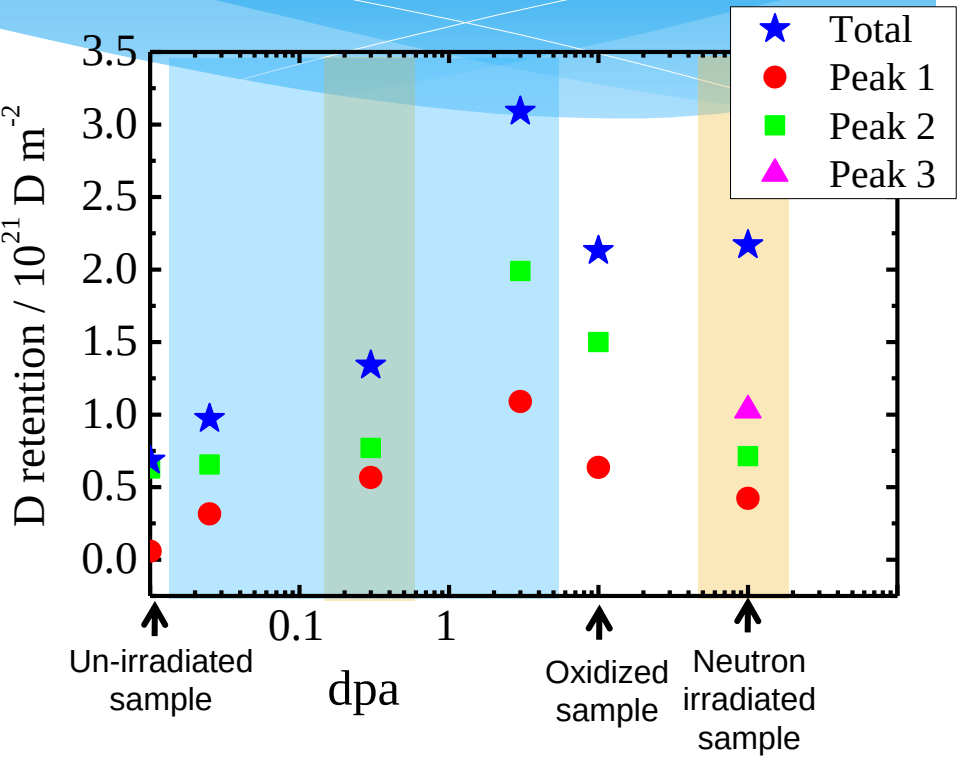
- The additional desorption peak was found for neutron-irradiated samples, which is almost the same as the HTS (2 dpa), but the ion-irradiated sample with 3 dpa does not show this higher desorption.

G. M. Wright, et al., Nucl. Fusion 50 (2010) 075006.

TDS spectra for neutron irradiated W and ion implanted W



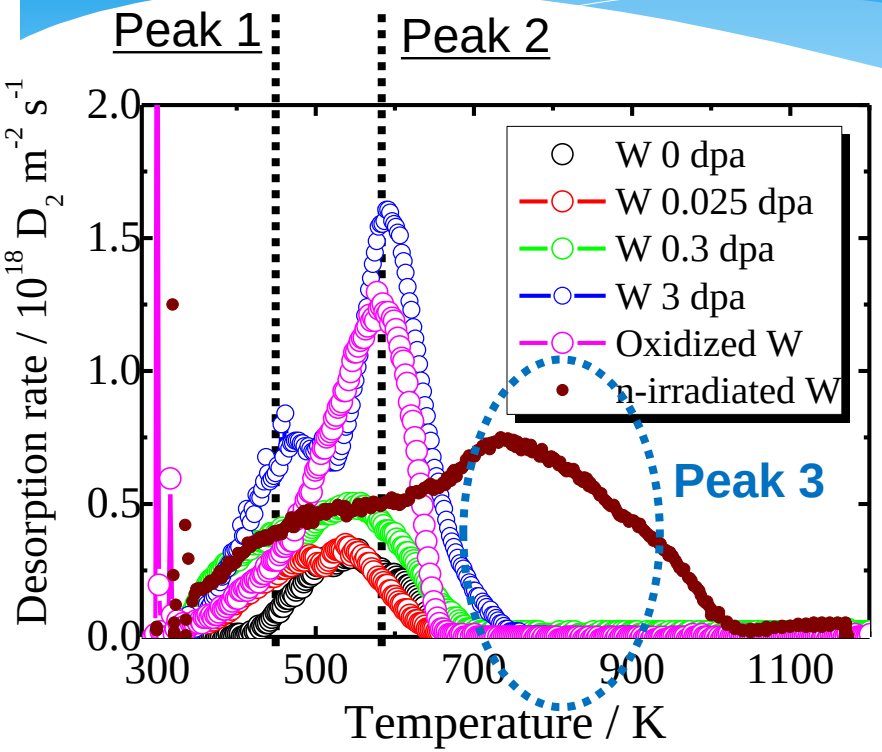
D_2 TDS spectrum for damaged W



D retention for various W samples

- Two desorption peaks were observed for ion implanted W and oxidized one. However, that for neutron irradiated one was consisted of more than three stages.
- Deuterium retention in both peaks increased as the number of dpa increased. (Peak 2 was remarkably increased)
- The TDS spectrum for oxidized sample does not show the additional desorption peak, showing that no surface effect for D trapping.
- **Peak 3 was only appeared for n- irradiated W.**

TDS spectra for neutron irradiated W and ion implanted W



D_2 TDS spectrum for damaged W

Peaks 1 and 2 ⇨ (1-1.5 eV)

Trapped by monovacancy

Peak 3 ⇨ High energy trap site (~ 2 eV)

Trapped by void formed from vacancies clustering

- Two desorption peaks were observed for ion implanted W and oxidized one. However, that for neutron irradiated one was consisted of more than three stages.
- Deuterium retention in both peaks increased as the number of dpa increased. (Peak 2 was remarkably increased)
- The TDS spectrum for oxidized sample does not show the additional desorption peak, showing that no surface effect for D trapping.
- **Peak 3 was only appeared for n- irradiated W.**

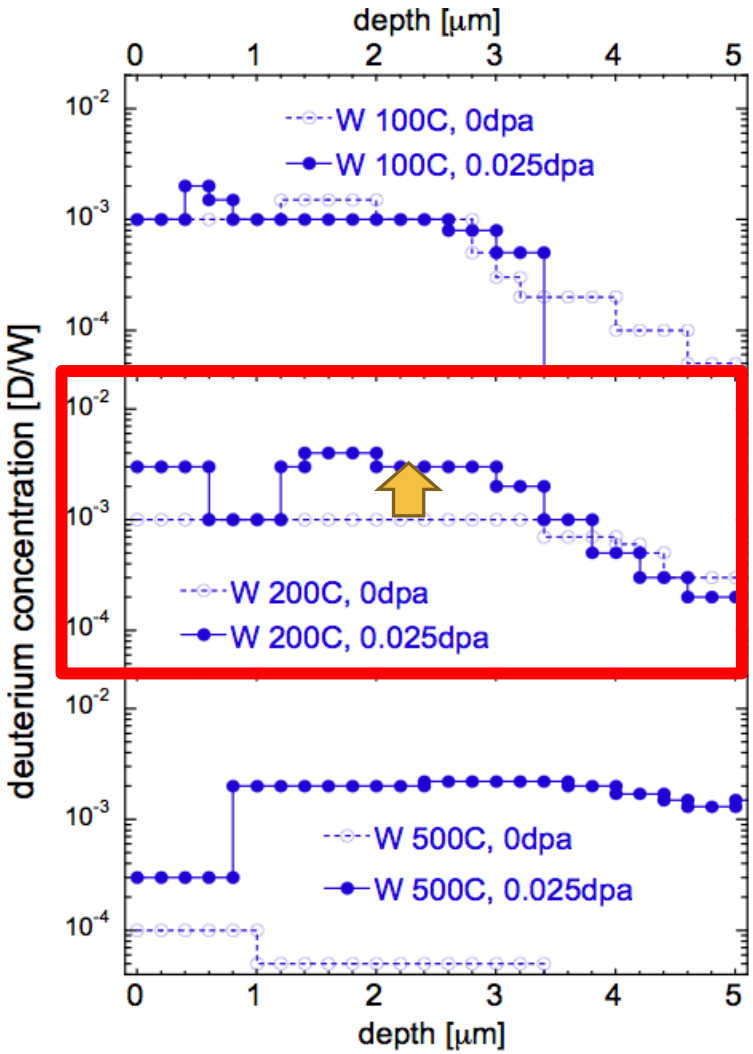
Comparison of D retention for various W samples

Sample ID	Temperature	D fluence [D m^{-2}]	D retention [D m^{-2}]	ratio (retained/implanted)
Neutron irradiation (0.025 dpa)	473 K	3.6 X 10^{25}	2.17 X 10^{21}	6.03 X 10^{-5}
Ion implantation 3 dpa	473 K	6.0 X 10^{25}	3.10 X 10^{21}	5.17 X 10^{-5}
Ion implantation 0.3 dpa	473 K	5.9 X 10^{25}	1.34 X 10^{21}	2.27X 10^{-5}
Ion implantation 0.025 dpa	473 K	5.7 X 10^{25}	9.73 X 10^{20}	1.71X 10^{-5}
Un-irradiated W	473 K	6.2 X 10^{25}	6.85 X 10^{20}	1.10X 10^{-5}

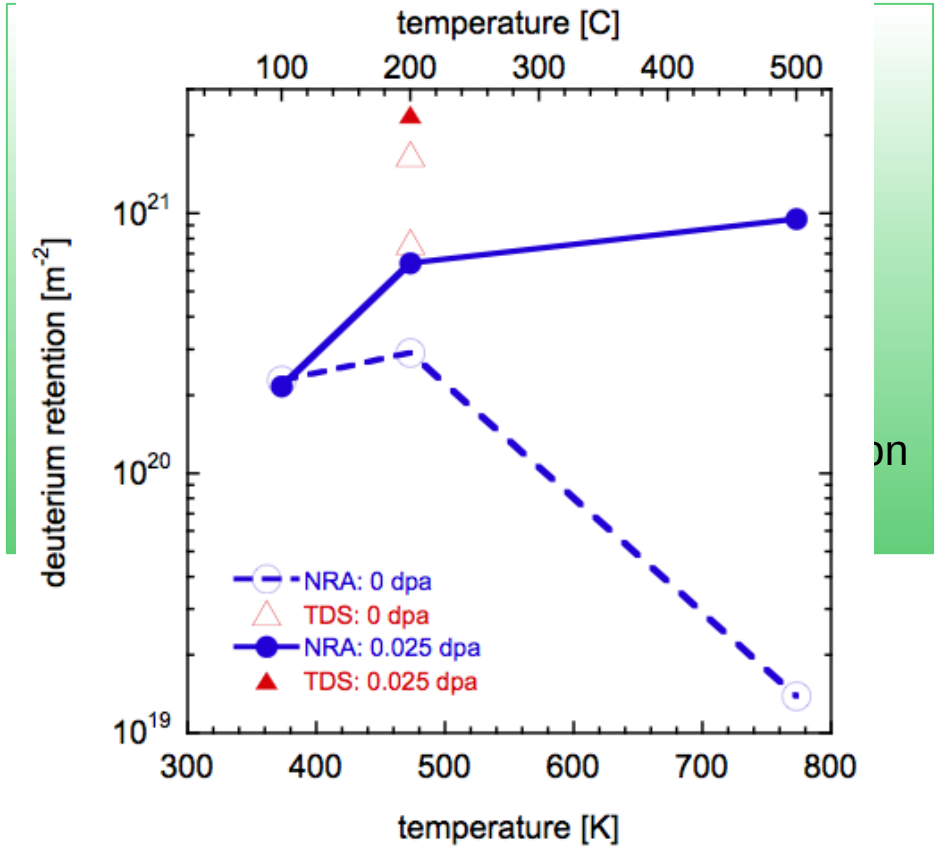
3 times

1 [illegible] times

D depth profiling for neutron-irradiated W by NRA



D depth profile for neutron-irradiated W



Comparison of D retention for NRA and TDS

P50B Masashi Shimada et al.
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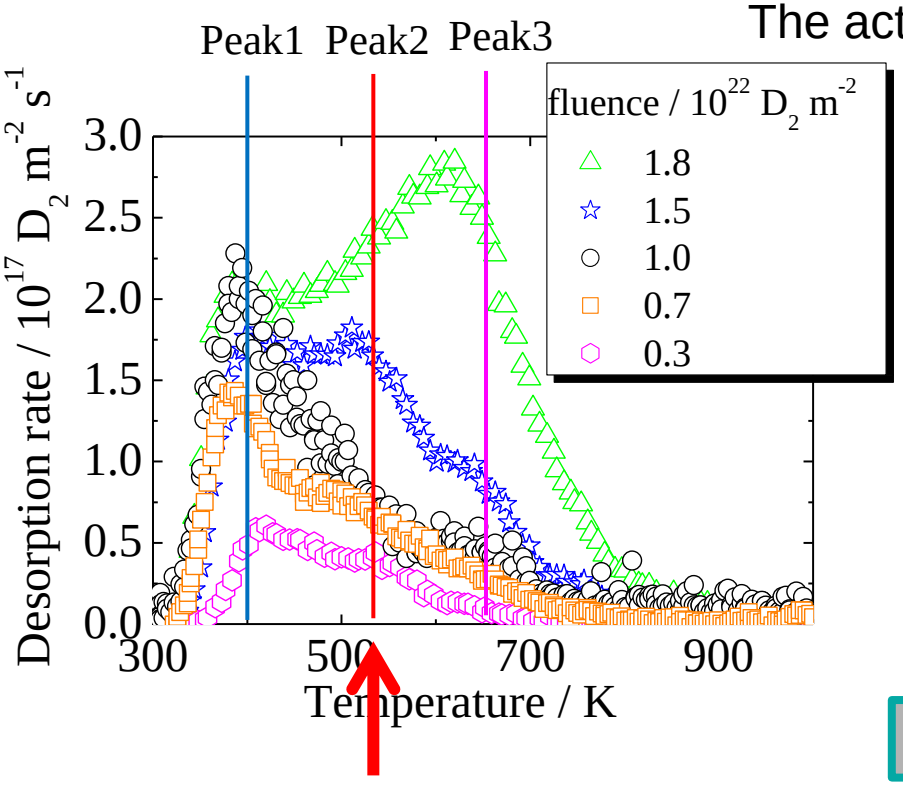
Estimation of activation energy by D_2^+ implantation

1 keV D_2^+ implantation was performed for same W sample and TDS experiment was performed at Shizuoka University.
The desorption stages were almost consistent with D plasma exposure samples, but the peak intensities for each desorption stages were different.



The activation energy for Peak 2 was estimated.

Assuming to be 2nd order reaction (surface recombination)

Polanyi-Wigner equation

$$\ln \nu(t) = \ln\left(-\frac{d\theta}{dt}\right) = \ln \nu_2 + 2\ln\theta + \frac{-E_a}{RT} \times \frac{1}{T}$$

$\nu(t)$: Surface D concentration
θ : Surface coverage
ν_2 : Frequency factor
Ea : Activation energy for D desorption

Activation energy

Peak2 (550 K) : vacancy ⇒ 1.48 eV

The activation energy for Peak 1 should be much lower than that for Peak 2 (1.48 eV), which would be caused by multi-trapping in a monovacancy.

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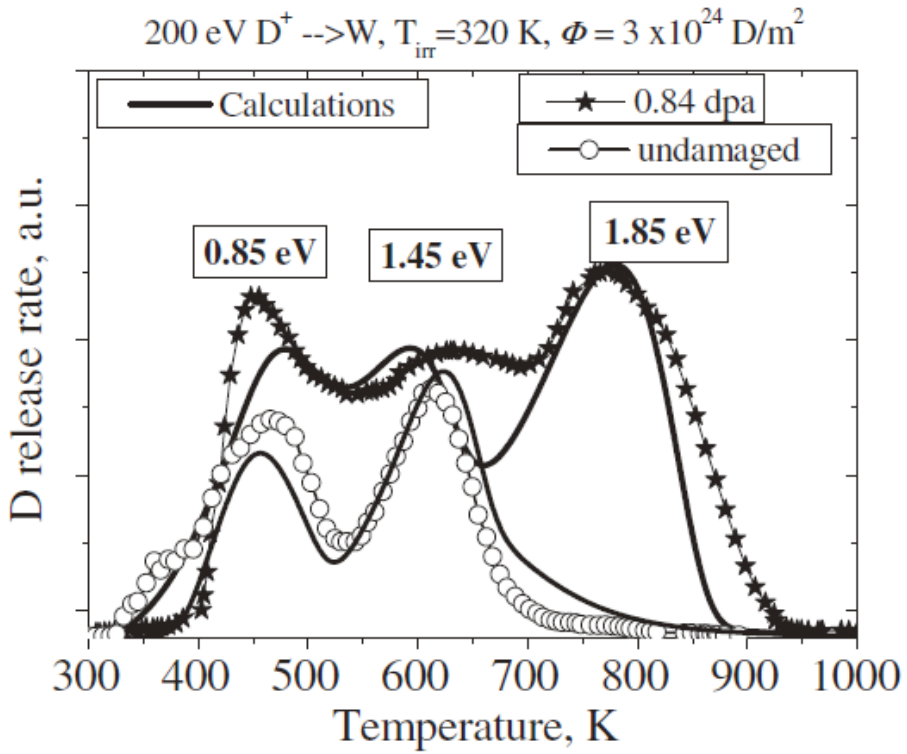
D concentration
verage
y factor
n energy for D desorption
n energy



O.V. Ogorodnikova et al., J. Nucl. Mater. In press.

Peak2 (550 K) : vacancy ⇒ 1.48 eV

The activation energy for Peak 1 should be much lower than that for Peak 2 (1.48 eV), which would be caused by multi-trapping in a monovacancy.

Hydrogen – vacancy binding energy

Unit : eV

N	E_b (Heinola et al)	E_b (D.F. Johnson et al.)
1	1.43	1.41
2	1.41	1.40
3	1.22	1.14
4	1.11	1.14
5	1.00	0.91
6	0.47	0.79

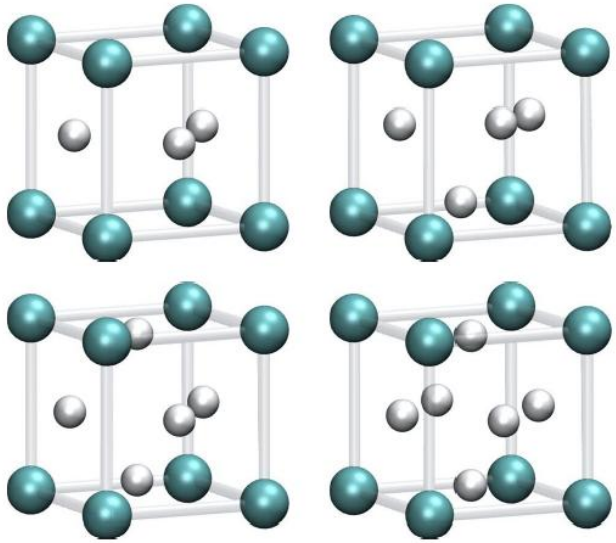


FIG. 2. (Color online) Geometries for the lowest-energy sites of three to six hydrogen atoms in a W monovacancy.

If a vacancy is produced, at least 6 H atoms can be trapped simultaneously at a single vacancy.
1 and 2 H atoms are trapped with identical energies, whereas ensuing H atoms are bound with lower energy.

→ Enhance D retention

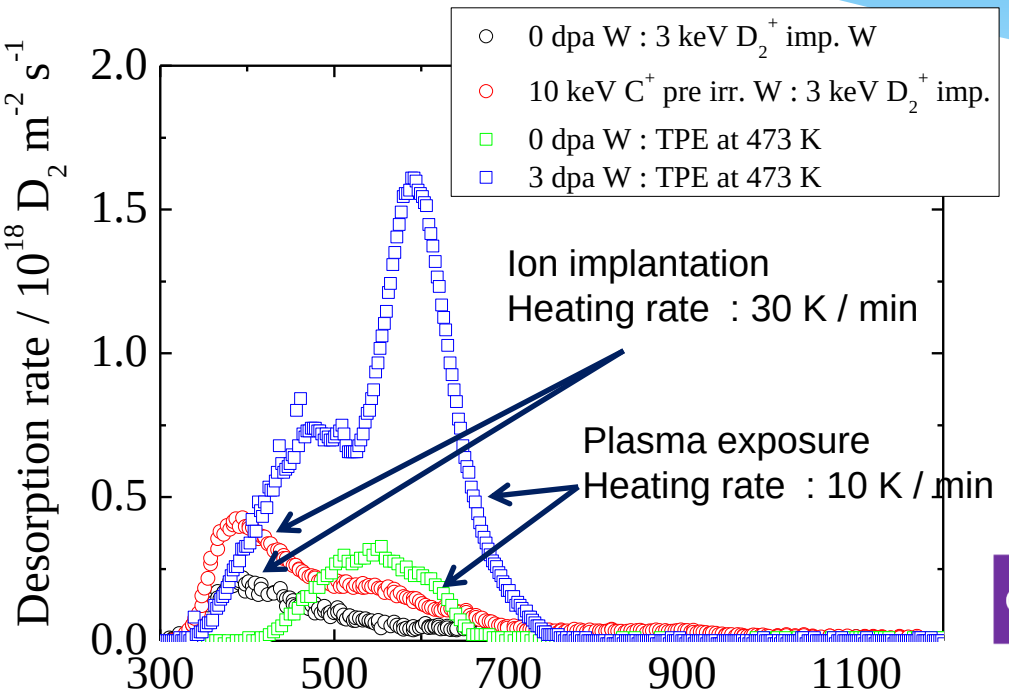
The damaged samples would enhance total D retention and induce the D accumulation.

↓

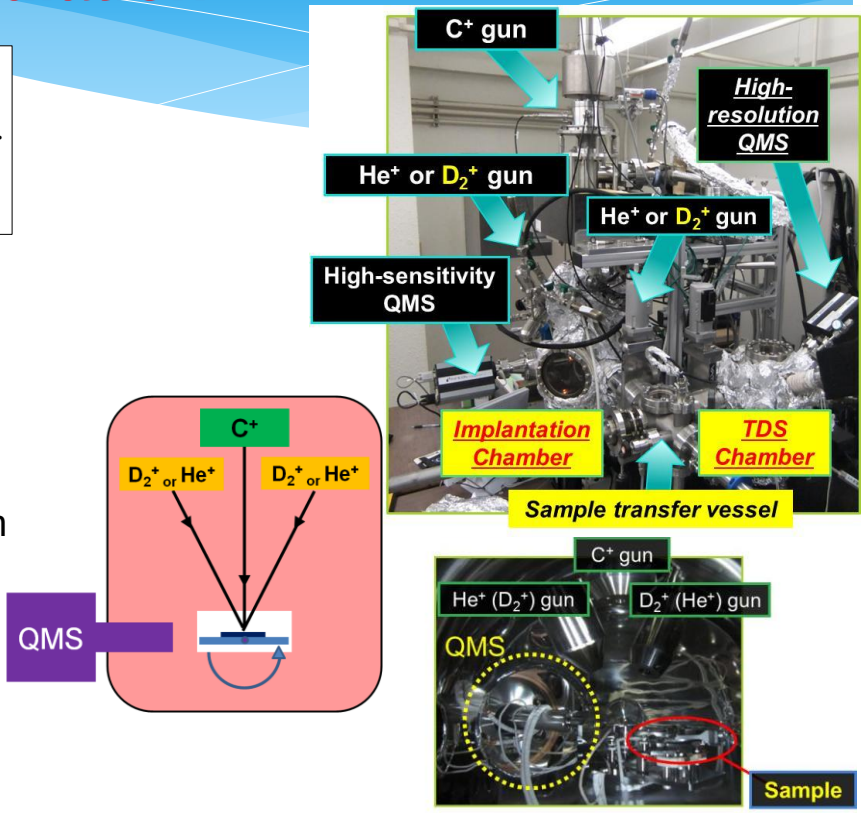
Additional desorption stage at lower temperature side (Peak 1) in TDS

K. Heinola, et al., Phys. Rev. B 82 (2010) 094102.
D. F. Johnson, J. Mater. Res. 25 (2010) 315.

Comparison of TDS spectra for D plasma exposure and D_2^+ ion irradiation



D_2 TDS spectra for damaged W

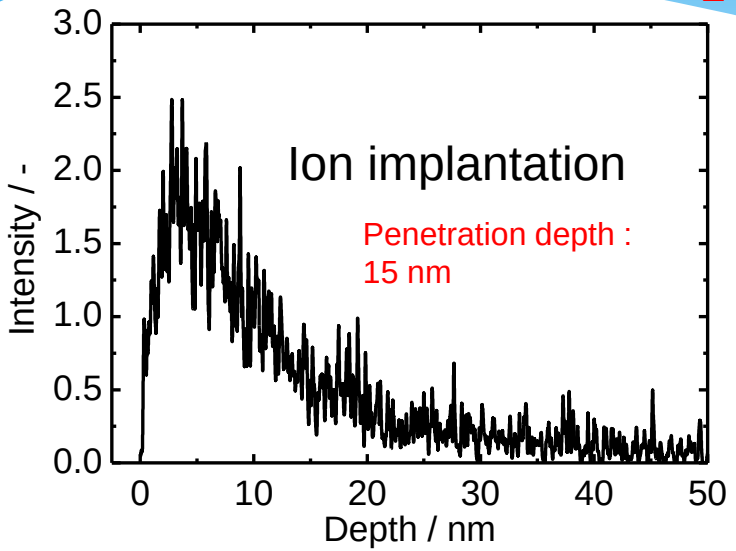


C^+ and D_2^+ implantation system at Shizuoka University

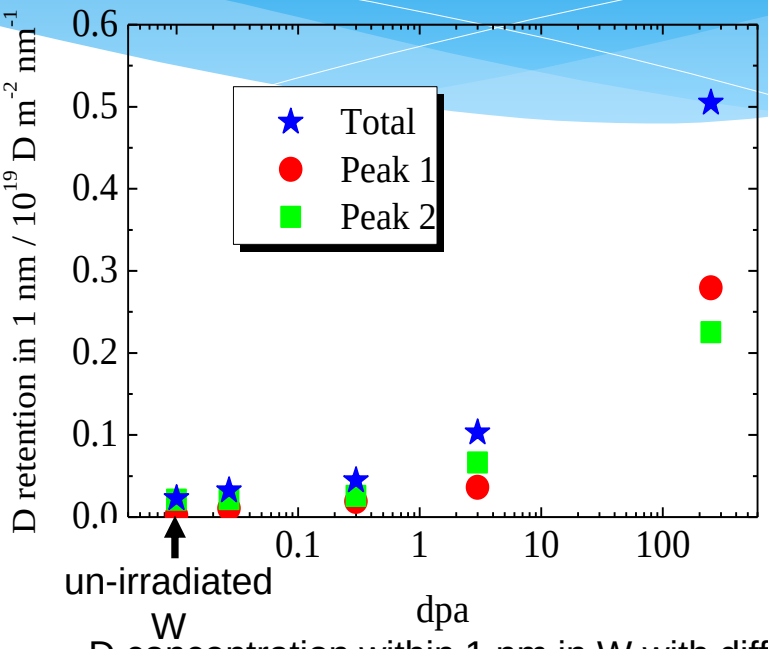
10 keV C^+ imp. : 243 dpa around depth of 15 nm
2.8 MeV Fe^{2+} imp. : 3 dpa around depth of 500 nm (Estimated by TRIM)

Peak temperatures for D_2^+ ion implanted sample were similar for D plasma exposed sample, but the D retention for these peaks were quite different.

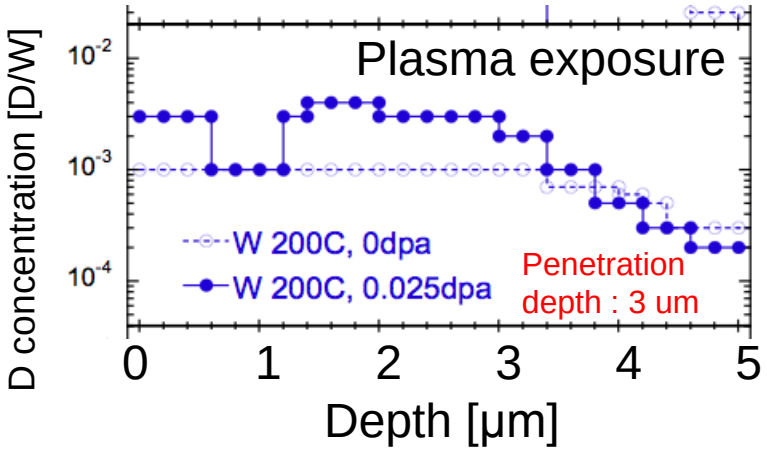
Dpa dependence on D retention for D plasma exposed W and D_2^+ ion irradiated W



D depth profile for C^+ implanted W observed by GD-OES



D concentration within 1 nm in W with different dpa

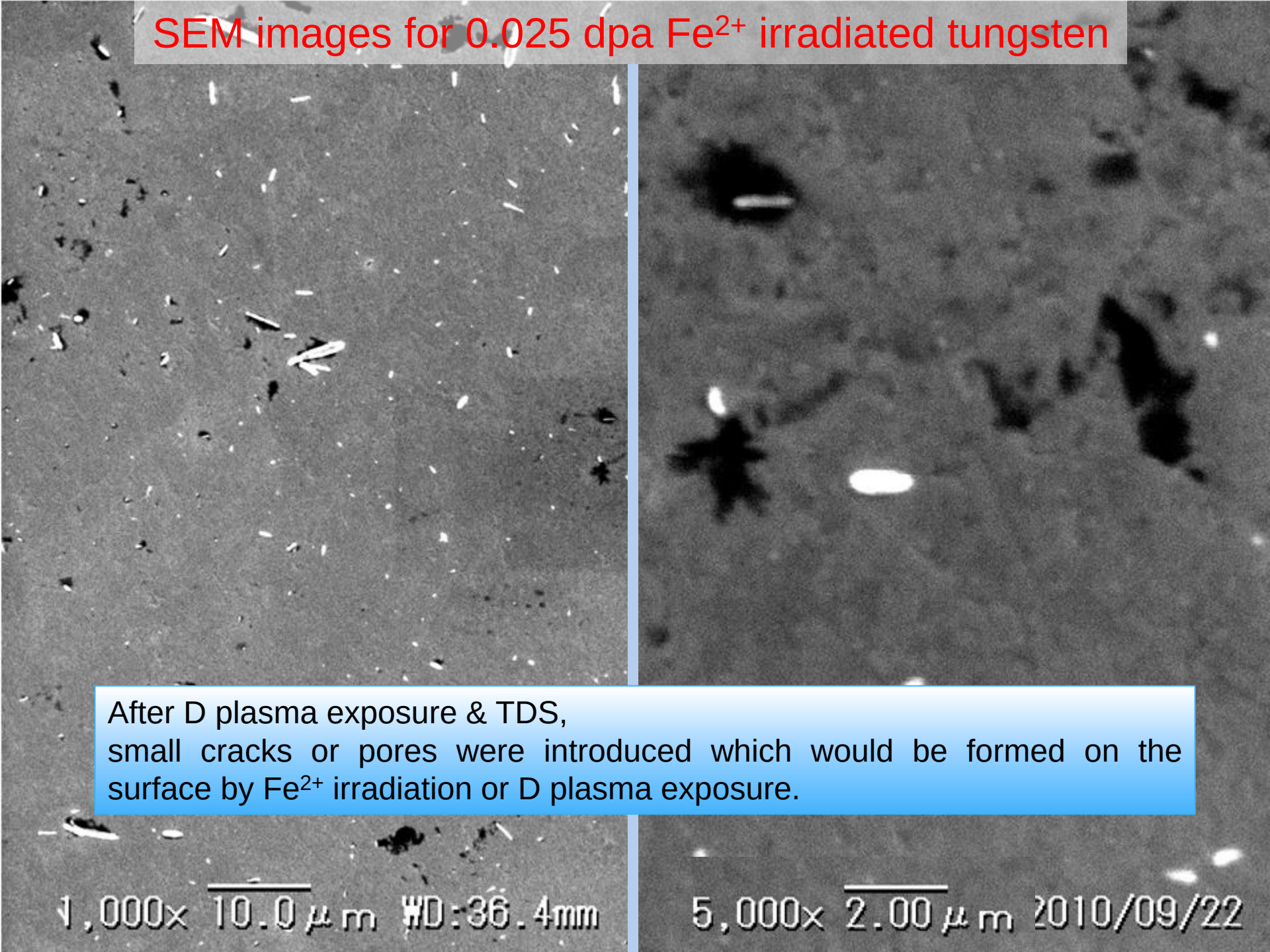


D depth profile for neutron irradiated W

D retention for C^+ implanted W was adjusted by the depth of plasma exposure case.
D retentions within 1 nm (D concentration for D plasma exposed W and D_2^+ ion irradiated W) were compared.

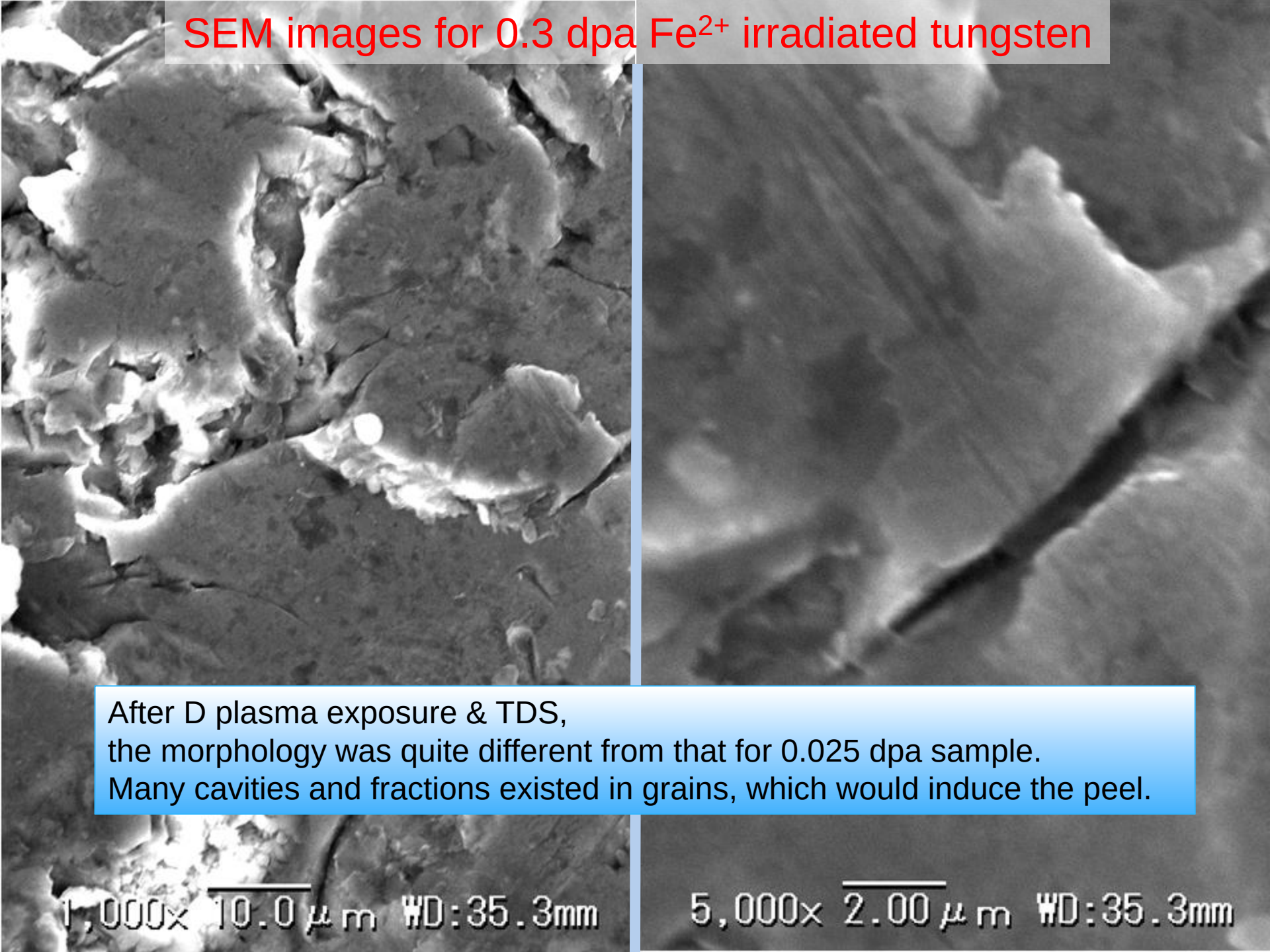
D retention clearly increased with increasing of dpa

SEM images for 0.025 dpa Fe^{2+} irradiated tungsten



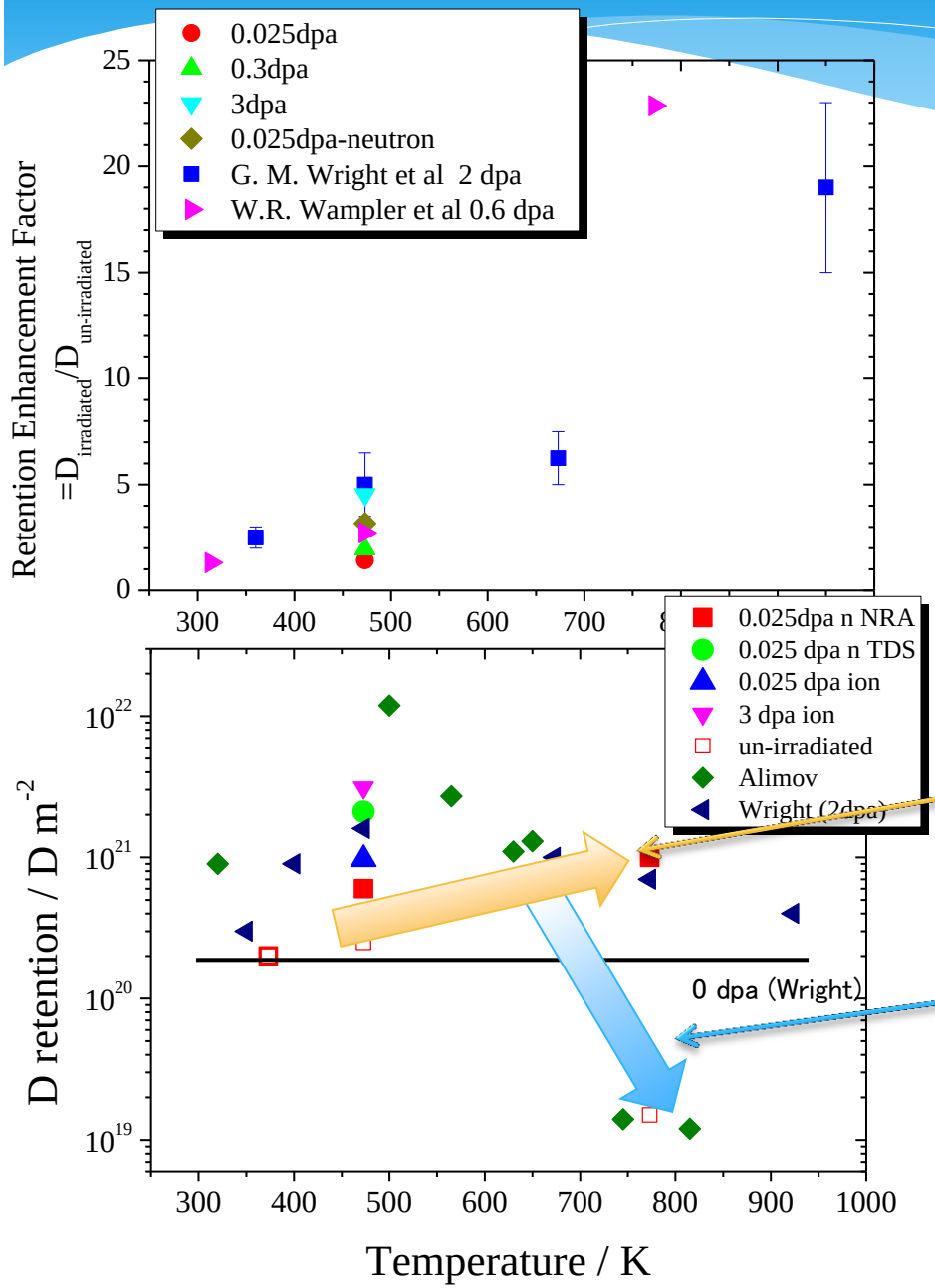
After D plasma exposure & TDS,
small cracks or pores were introduced which would be formed on the surface by Fe^{2+} irradiation or D plasma exposure.

SEM images for 0.3 dpa Fe^{2+} irradiated tungsten



After D plasma exposure & TDS,
the morphology was quite different from that for 0.025 dpa sample.
Many cavities and fractions existed in grains, which would induce the peel.

Comparison of Retention Enhancement Factor and D retention



Retention enhancement factor and D retention derived in the present study are almost consistent with the previous data.

However, we have to remember that the shape of TDS spectra is different.

Different D trapping sites should exist.

Irradiation case
D retention would be increased or remained.

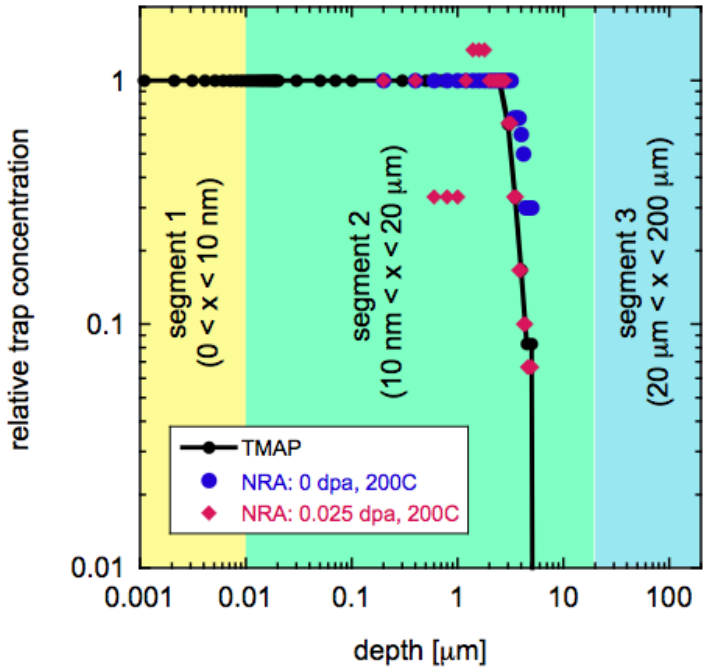
Un-irradiation case
D retention is clearly decreased as the temperature increased.

WR. Wampler et al. hys. Scr. T138 (2009) 014037.
V. Kh. Alimov et al. J. Nucl. Mater. In press.
G. M. Wright, et al., Nucl. Fusion 50 (2010) 075006.

TMAP simulation for the desorption of D in neutron-irradiated W

Assumptions:

- Simulate previously obtained TDS spectrum at 200 C (0 dpa/0.025 dpa)
- Use 3 segments ($0<x_{seg1}<10nm$, $10nm<x_{seg2}<20\mu m$, $20\mu m<x_{seg3}<200\mu m$)
- Use normalized NRA depth profile up to < 5 μm
- Set very low trap concentration ($D/W=10^{-6}$) in deeper bulk (x > 5 μm)
- Frauenfelder's Diffusivity
- R. Anderl's Recombination Coefficient
- Traps are fully filled with deuterium from surface (saturable traps only)
- Only three different trapping energies in TMAP7 (simulation limitation)



Normalized depth profiles used in TMAP simulation

Three different types of traps :

- Low energy trap (0.8-1.2 eV) :
  dislocation, Vacancy (2nd/3rd D atom)
- Medium energy trap (1.3-1.5 eV) :
  Vacancy (1st D atom), D_2 molecules from voids
- High energy trap (1.6-2.1 eV) :
  chemisorptions from voids, vacancy cluster

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TMAP simulation results

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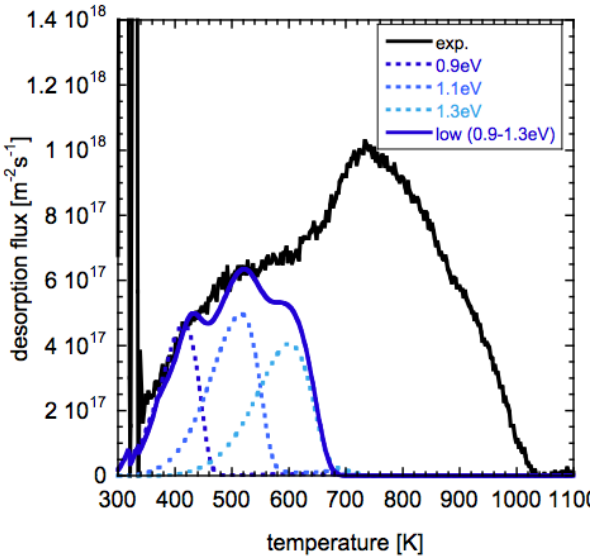
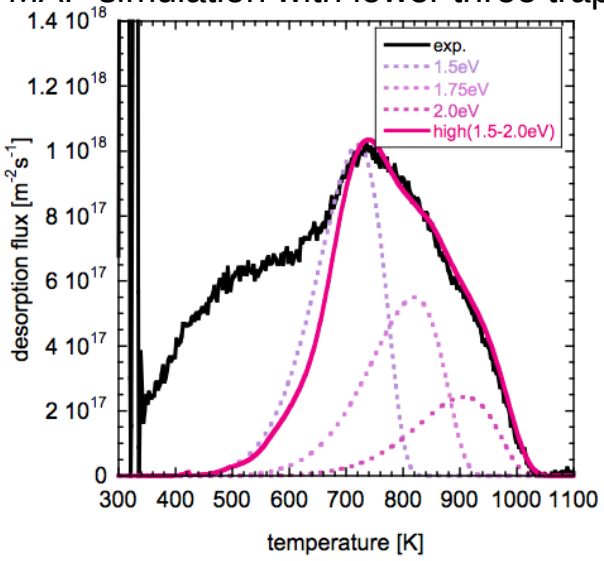
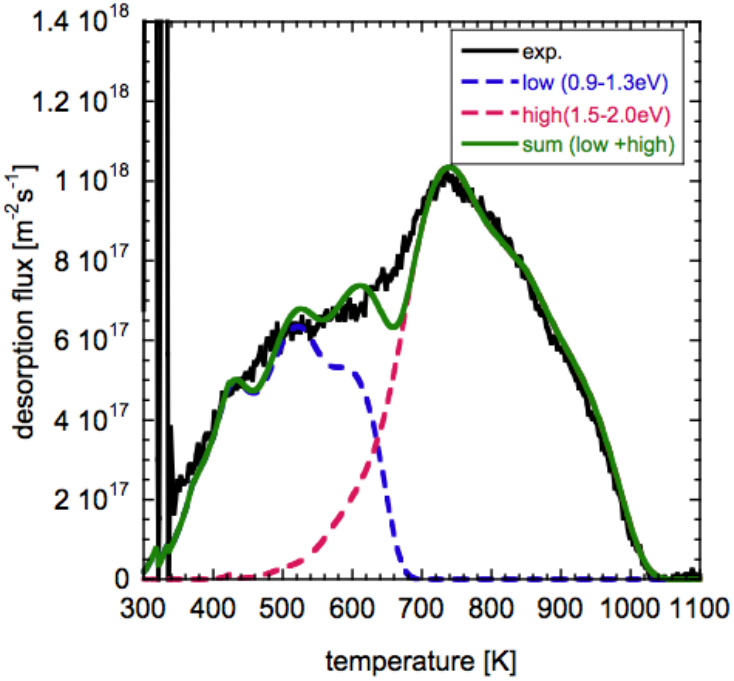


TMAP simulation with lower three traps



TMAP simulation with higher three traps



Summary of TMAP simulation of TDS spectra for 0.025 dpa neutron-irradiated W

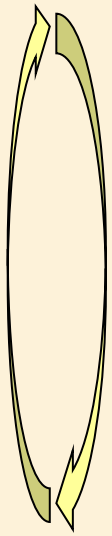
Four (0.9, 1.5, 1.75, 2.0 eV) traps are induced by neutron-irradiation, confirming that different trapping mechanisms exist for neutron-irradiated tungsten.

Monte Carlo code to simulate PWI

<Overview>

- Kinetic Monte Carlo
- Potential energy curve has been derived mainly from DFT calculation results
- 2D periodic boundary conditions
- Deuterium flux: 1×10^{26} D m^{-2} s^{-1}
- Temperature: 473 K

<Procedure>



(1) D implantation event

(2) Diffusion events: migration as D atom in bulk & on surfaces

- ✓ trapping/detrapping by vacancies is considered
- ✓ *R. Frauenfelder's hydrogen diffusivity is used.*

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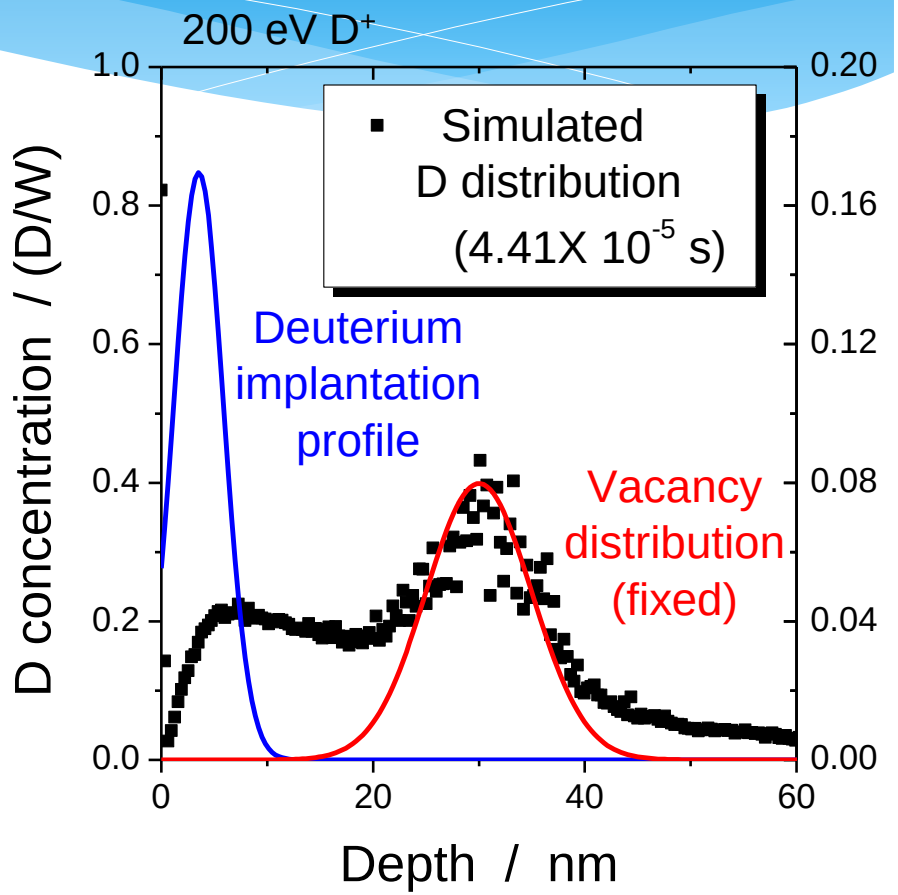
(3) Desorption events: recombination desorption as D_2

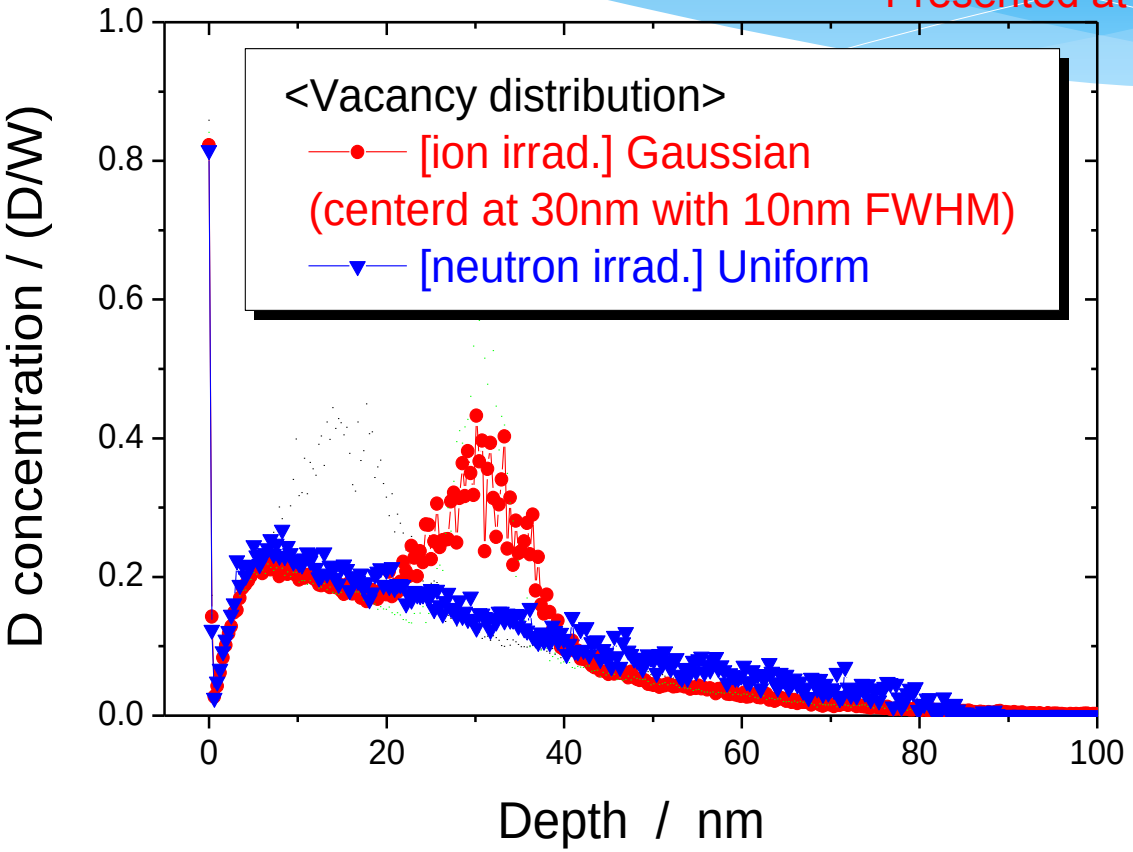


Comparison of simulated deuterium distribution with deuterium implantation profile and vacancy distribution

Dr. Oda, The University of Tokyo
Presented at T conference 2010

Calculation result: influence of vacancy distribution

Dr. Oda, The University of Tokyo
Presented at T conference 2010



Influence of vacancy distribution on simulated D distribution

- Defects created with ion irradiation and neutron irradiation were simulated by changing the defect distribution.
  - [ion] Implanted deuterium distribution is similar to defect distribution.
  - [neutron] Implanted deuterium concentration depends almost linearly on depth.

Conclusions

- The behavior of D retentions for Fe^{2+} irradiated tungsten with the damage of 0.025-3 dpa was compared with that for neutron irradiated tungsten with 0.025 dpa. In addition, surface morphology was clearly changed as the dpa increased.
- The D_2 TDS spectra for Fe^{2+} irradiated tungsten consisted of two desorption stages at 450 K and 550 K although that for neutron irradiated tungsten was composed of three stages and addition desorption stage was found around 750 K.

1st desorption stage : the accumulation of D in mono vacancy
2nd desorption stage : D trapping by intrinsic defects or vacancy
3rd desorption stage : D trapping by void or vacancy cluster (+ diffusion effect)

D_2 TDS spectra for ion-irradiated tungsten could not represent that for neutron-irradiated one, indicating that the deuterium trapping and desorption mechanism for neutron-irradiated tungsten has a difference from that for ion-irradiated one.

The simulation work represents the multiple D trapping states, especially neutron irradiation case.
The difference of vacancy distribution contributes the D trapping profiles, which would influence on D desorption behavior.

Thank you for your attention

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