Hydrogen isotope retention in W irradiated by heavy ions and helium plasma

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- Summary

Experimental setup: Cu^{2+} Irradiation

Tandetron Accelerator (Kyushu Univ.)

Ion: Cu^{2+}

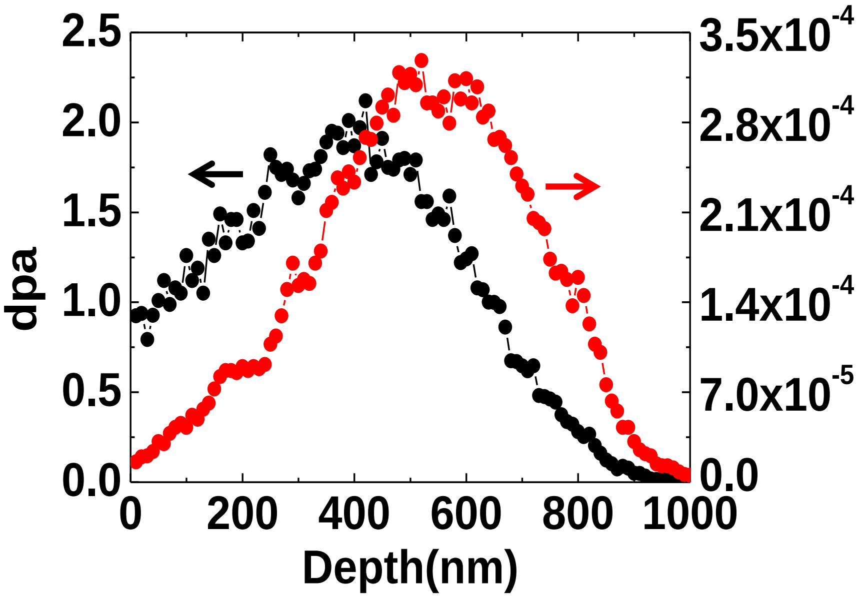
Energy: 2.4 MeV

Damage level: up to 4 dpa (max: > 100 dpa)

Sample Temp.: up to 600 ^{o}C

Exposure area: 8 mmϕ

2.4MeV-Cu^{+} (1x$10^{19}Cu^{2+}/m^2$) ➔ W



- Estimated depth profile of displace-ment damage and implanted ion range distribution in W.
- Calculation using the SRIM code with displacement energy of 55 eV.
- The peak damage region is ~400 nm.

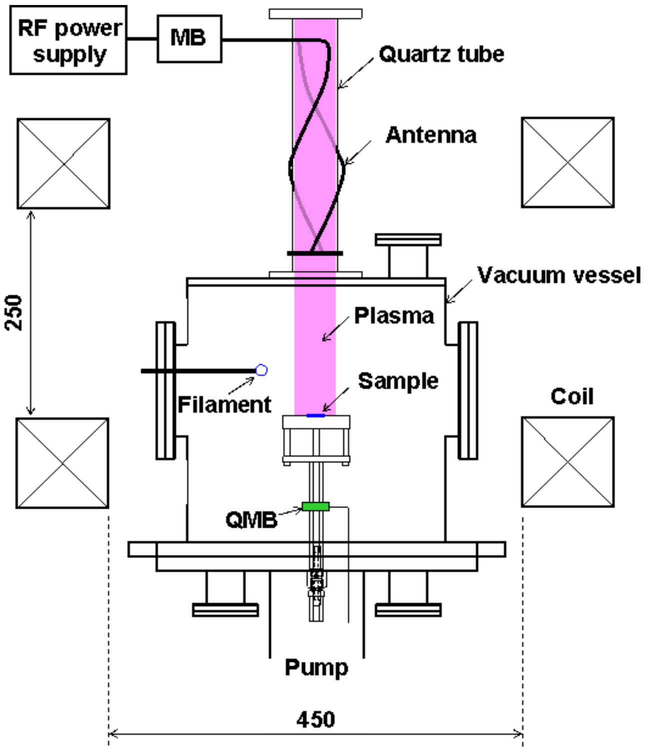
Experimental setup: Deuterium Irradiation

- High energy D ion beam irradiation has been done in Kyushu Univ.

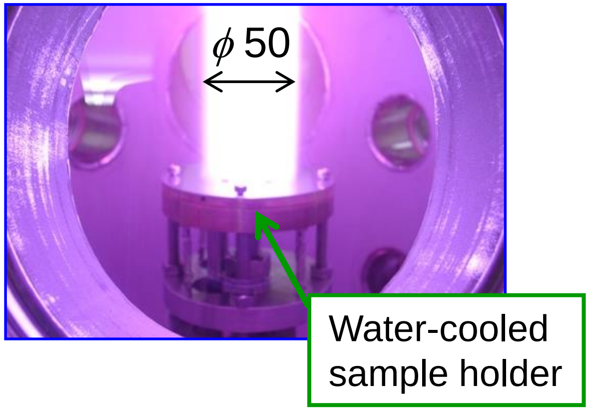
  Energy: 2 keV-D_2^+, Fluence: 1×10^{21} D_2/m^2

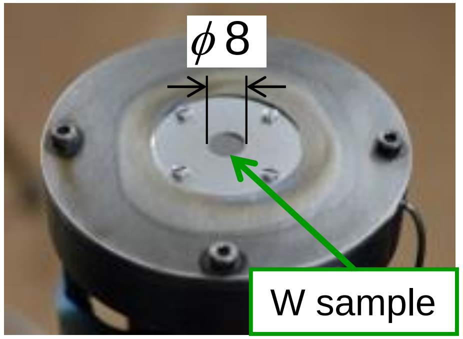
- Plasma exposures have been performed using a compact PWI simulator APSEDAS.

APSEDAS (PRC, Univ. of Tsukuba)



- Plasma is produced by RF (13.56 MHz) wave power (< 5kW, typical: 800 W steady state).
  ==> No electrode, No impurity such as carbon, Clean environment
- Magnetic field: < 0.05 T
- Water cooled sample stage
- Plasma diameter: ~50 mm
- Exposure area: ϕ8 mm (uniform exposure)





Examined Materials

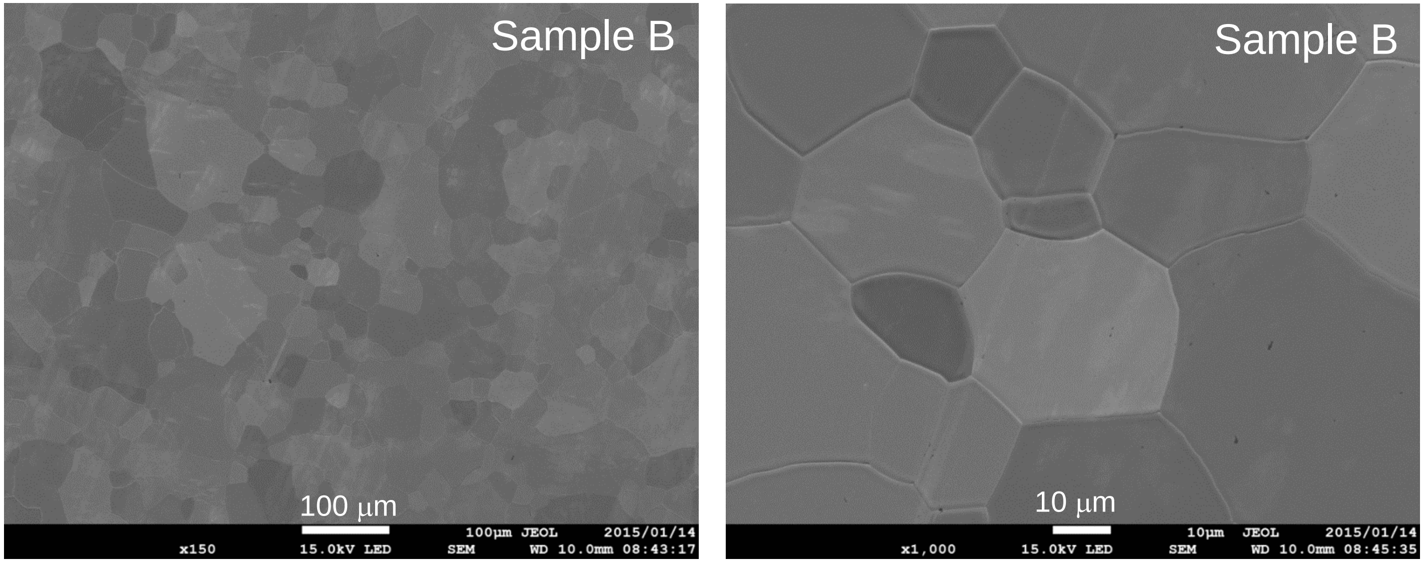
Base Materials:

(A) 0.1mm-thick tungsten sheet (99.95% pure, Nilaco Co.)

- Annealed at ~2000 ^{o}C for 20 min (Re-crystallized)

(B) 1mm-thick tungsten disc (99.99% pure, A.L.M.T. corp.)

- Annealed at ~ 2000 ^{o}C for 1 h (Re-crystallized)



The grain size is in the range of 10 μm to ~100 μm.

Experimental Procedure

■ Sample A:

(A) 0.1mm-thick tungsten sheets (99.95% pure, Nilaco Co.)

Annealed at ~2000 °C for 20 min (Re-crystallized)

↓

2.4 MeV Cu^{2+} irradiation → Thinning with twin-jet electropolishing (~100 nm)

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2 keV-D_2^+ irradiation by the ion beam

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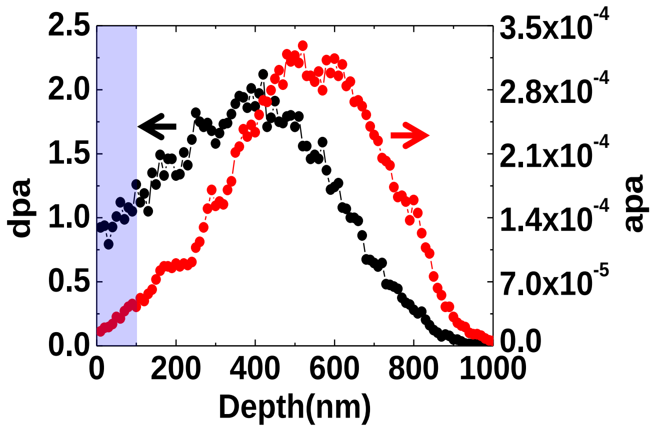
TDS measurement

Thinning with twin-jet electropolishing (~100 nm) → TEM observation



Experimental Procedure

- Sample B:

(B) 1mm-thick tungsten discs (99.99% pure, A.L.M.T. corp.)

Annealed at ~2000 ^{o}C for 1 h (Re-crystallized)

↓

2.4 MeV Cu^{2+} irradiation

↓

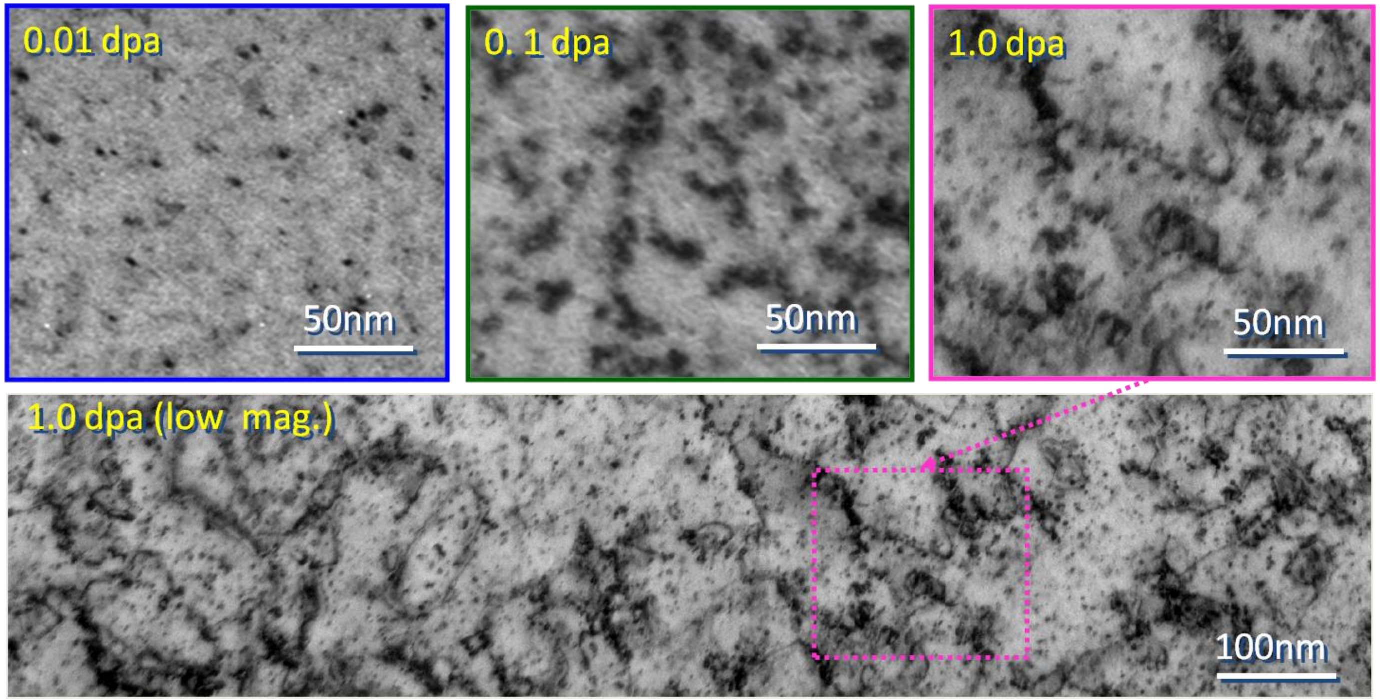
D plasma irradiation

↓

TDS measurement

Damage Evolution (Sample A)

300K , 2.4MeV-Cu^{2+}, 0.007~0.7dpa, about $1x10^{-4}$dpa/s

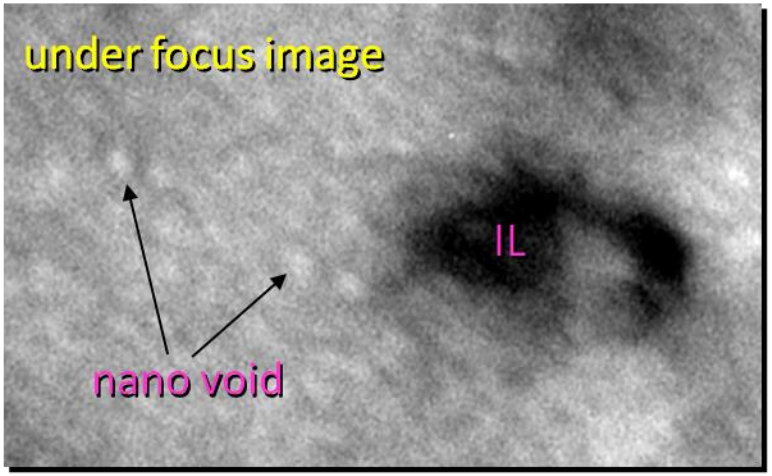


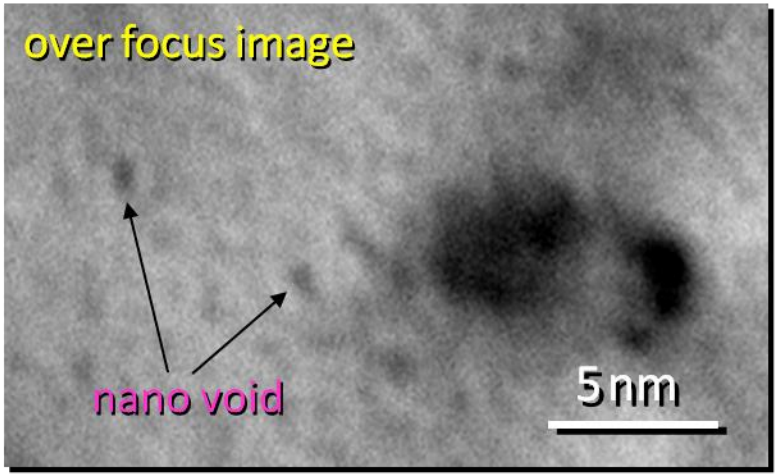
- Most of the interstitial loops (IL) must be nucleated by cascade collisions, since the density of ILs was two order of magnitude higher than the estimated value using a rate theory assuming that the two free interstitials act as nuclei for ILs.
- Each IL can not grow larger individually but aligned ILs grow by coalescing. Concentration of ILs and interstitials accumulated in ILs exceeds $2x10^{-7}$ and $5x10^{-4}$ (≧0.1dpa), respectively.
- Small ILs align and change to larger IL by coalescing each other.

After H. Watanabe et al. 16th ICFRM, 16-405.

Formation of nano-void

300K , 2.4MeV-Cu^{2+}, 1.0dpa, about $1x10^{-4}$dpa/s



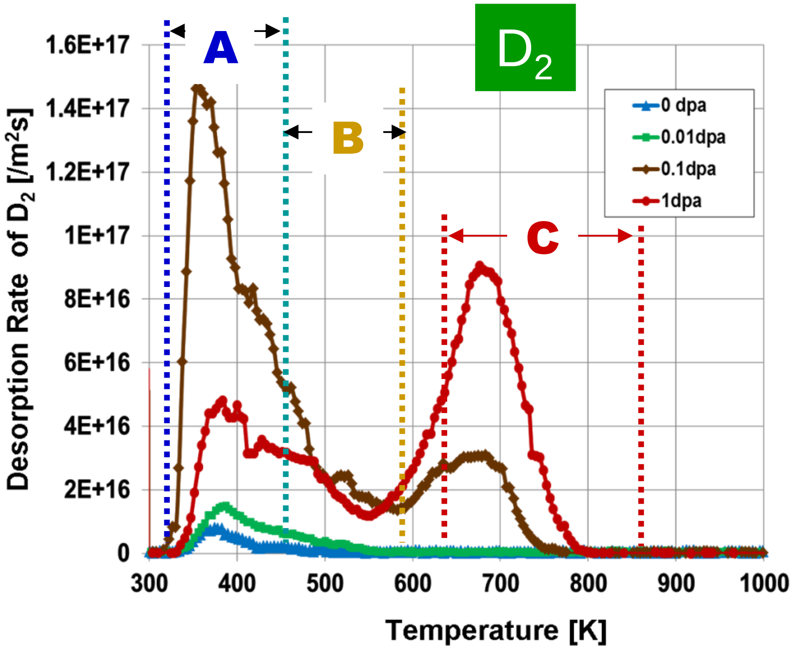


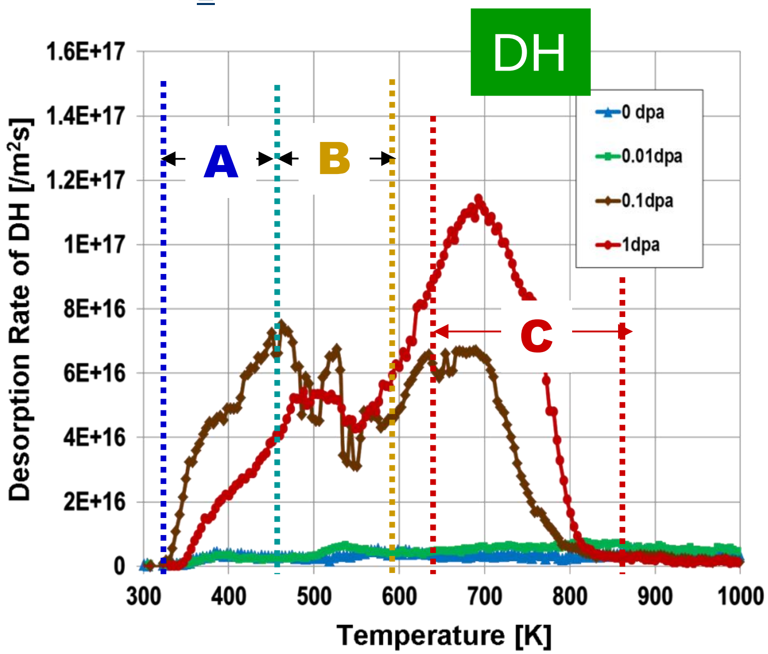
- Nano-voids (d<1nm) are observed in 1 dpa case and they formed densely.
- Nano void formation following 2.4 MeV-Cu^{2+} irradiation at room temperature seems to be attributed to the cascade collisions and radiation induced diffusion of the vacancies.

After H. Watanabe et al. 16th ICFRM, 16-405.

TDS Spectra after 2 keV-D_2^+ irradiation (Sample A)

2.4MeV-Cu^{2+}, 0.01~1.0dpa @300K + 2keV-D_2^+, $1x10^{21} D_2^+/m^2$ @300K





- There seems to be mainly two peaks in the spectra.
- In the case of 0.1 dpa, the trapping of the peak A is greater than that of the peak C. In the case of 1 dpa, on the other hand, the peak C is greater than the peak A.
- Judging from the TEM observation and the computer simulation, trapping sites of A and C seems to be vacancies (+ their very small clusters?) and nano voids, respectively.

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D plasma irradiation & TDS

- Sample B:

  (B) 1mm-thick tungsten discs (99.99% pure, A.L.M.T. corp.)

Annealed at ~2000 ^{o}C for 1 h (Re-crystallized)

↓

2.4 MeV Cu^{2+} irradiation

↓

D plasma irradiation

↓

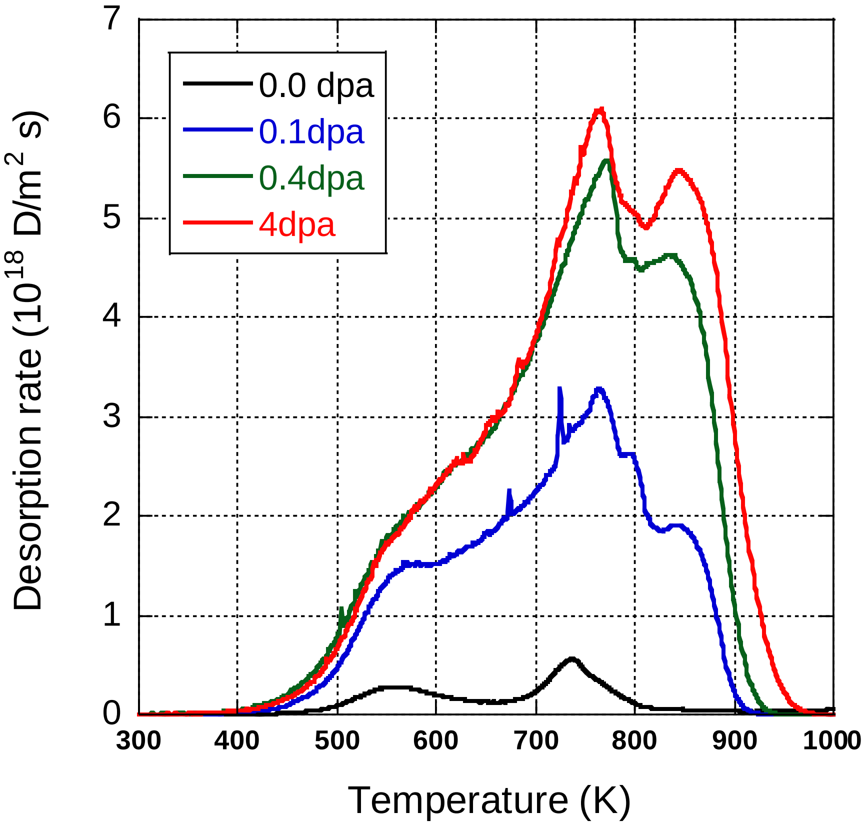
TDS measurement

Plasma parameters

Electron density (m^{-3})	2.7 x 10^{17}
Electron temp. (eV)	10
Space potential (V)	30
Flux (m^{-2} s^{-1})	3.7 x 10^{21}
Fluence (m^{-2})	2.0 x 10^{25}
Surface temp. (K)	480

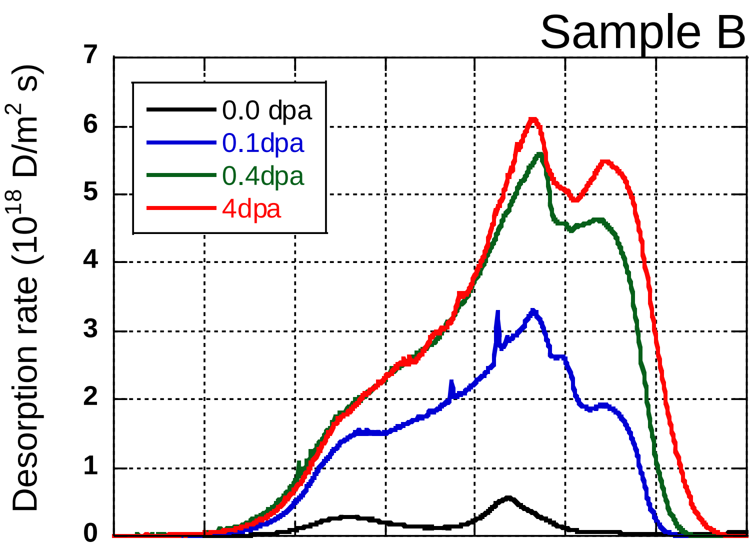
- Low energy and high flux plasma was exposed to the sample B.
- The D atoms seem to diffuse over the damage peak (~400nm), since the fluence was high and the surface temperature was also high.

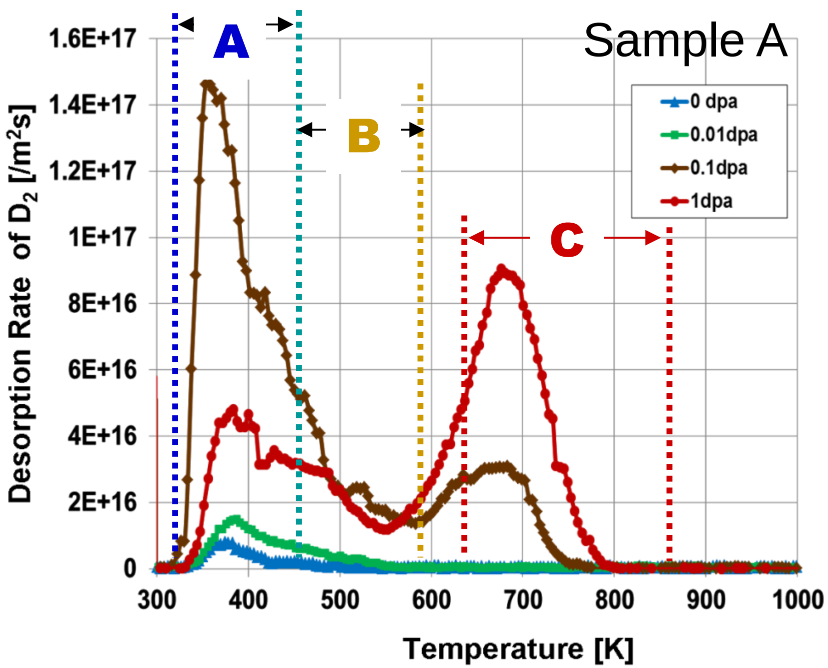
Dependence of TDS Spectrum on the damage level



- There seems to be three main peaks in the spectra and the peak around 850 K newly appeared due to 2.4 MeV Cu^{2+} irradiation.
- Retention in the damaged W increases with the damage level but it saturates around 0.4 dpa, suggesting that newly introduced defects may be cancelled by already existing vacancies and voids with high density.

Comparison between samples A & B





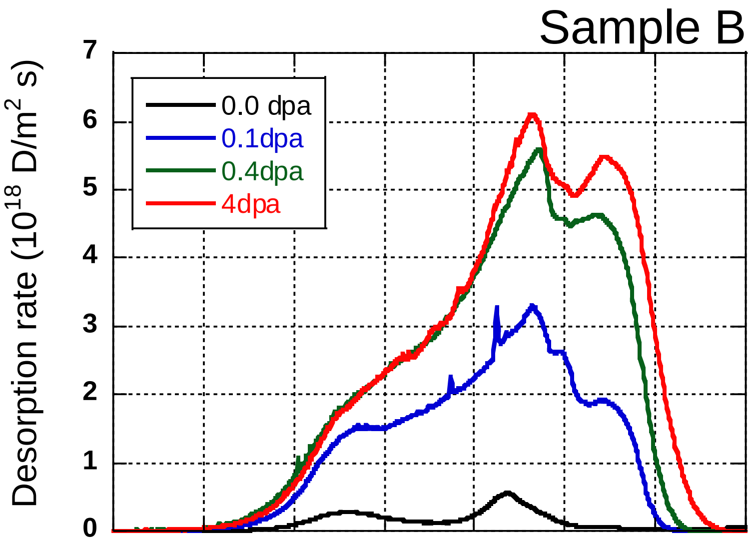
Sample	A	B
Irradiation	Ion beam	Plasma
Incident energy (eV)	1000	30
Fluence (D m^{-2})	2.0×10^{21}	2.0×10^{25}
Surface temp. (K)	300	480

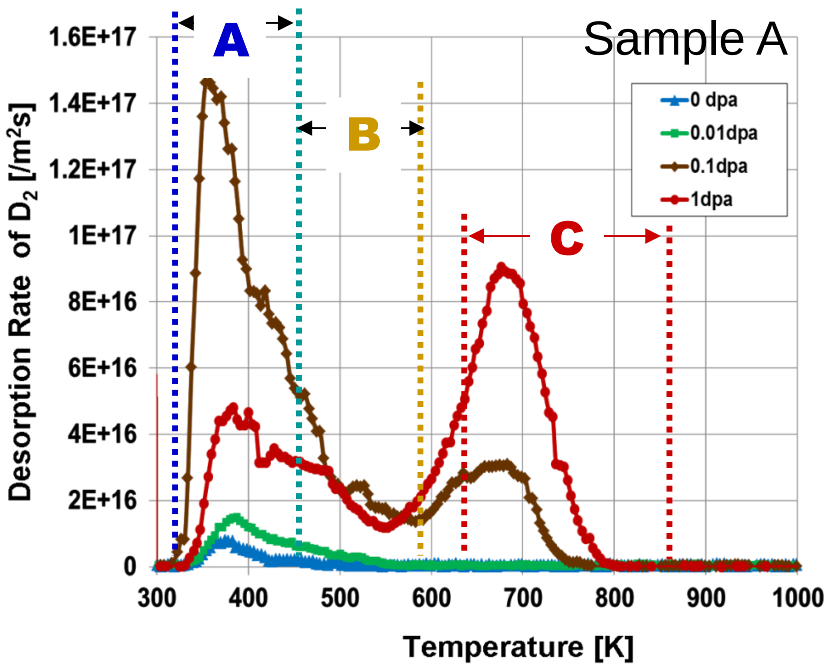
- The desorption rate is much higher in the plasma irradiation (i.e. sample B).
- In the case of ion beam irradiation, no desorption around 850 K was observed.

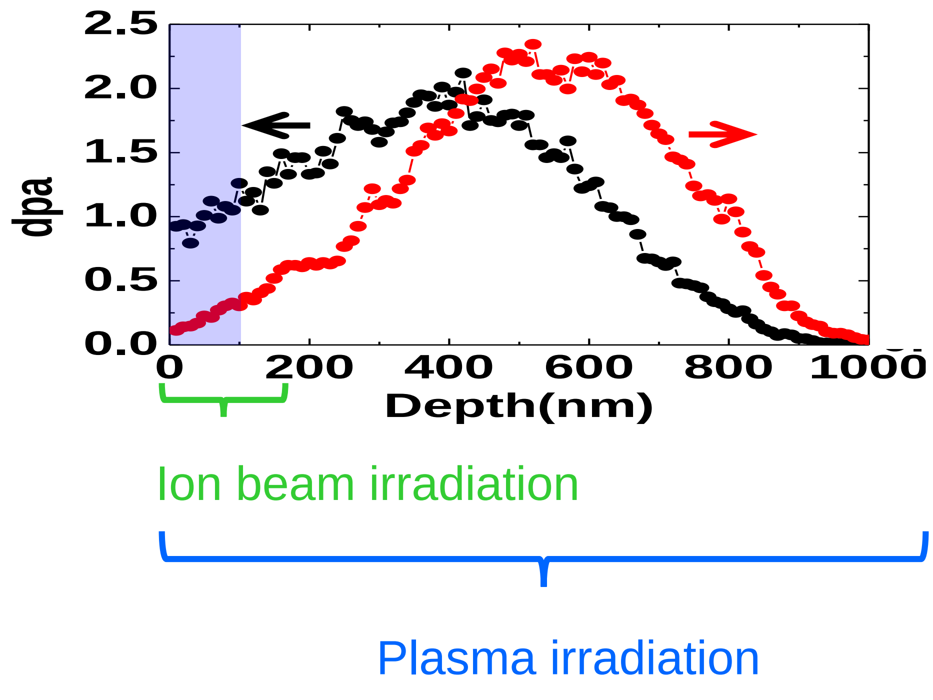
Question: Where did D atoms exist?

- In the case of plasma irradiation, the D atoms seem to diffuse over the damage peak (~400nm), since the fluence was high and the surface temperature was also high.
- On the other hand, in the case of ion beam irradiation, the D atoms may not reach the damage peak due to the low fluence.

Comparison between samples A & B







- The trapping around 850 K in the sample B (i.e. plasma irradiation) is considered to be attributed to the defects (i.e. voids) around the damage peak.

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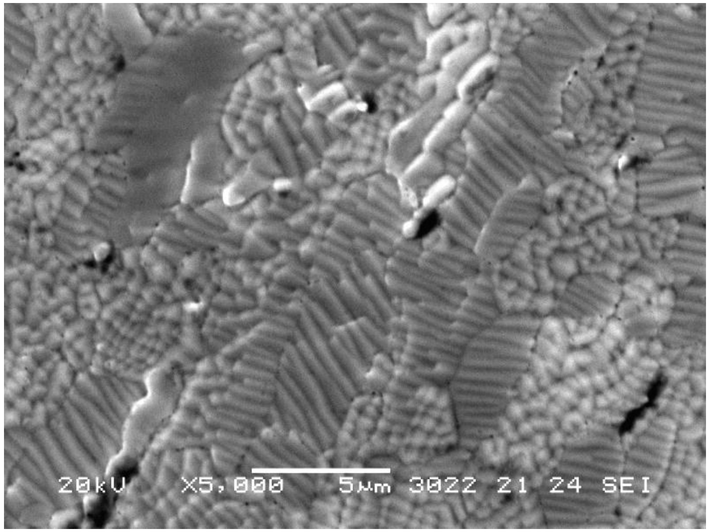
Procedure (1): He bubble layer formation

Preparation

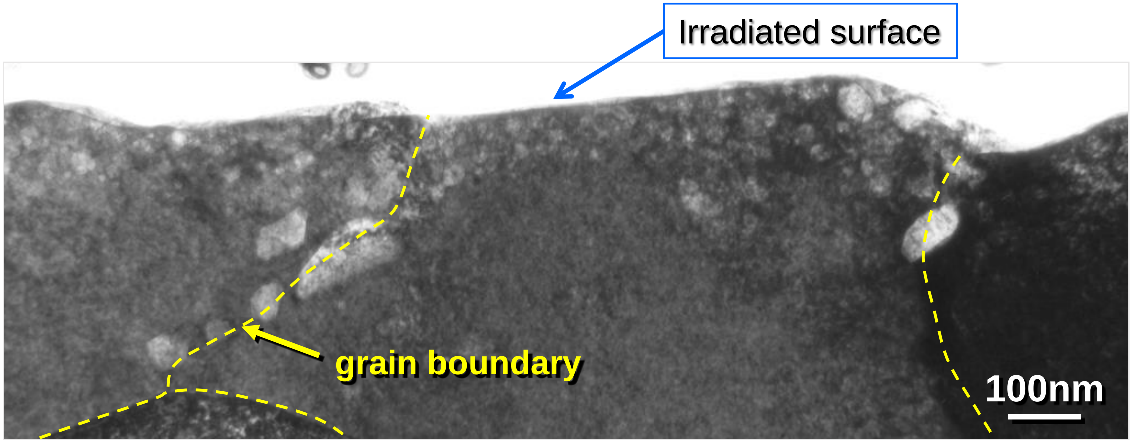
- Polycrystalline tungsten: 0.1 mm thickness (Nilaco. Co.)
- Ultrasonic cleaning with acetone and ethanol
- He plasma exposure without cooling
  - ✓ Surface temperature: 1700 – 1900 K
  - ✓ Ion energy: ~ 30 eV
  - ✓ Flux: ~ 2 x 10^{22} He/m^2s
  - ✓ Fluence: ~ 8 x 10^{25} He/m^2



SEM micrograph

He bubble formation

- Remarkable fine irregularities of < a few μm on the surface.
- Many bubbles with the size of ~10nm to ~200 nm were generated beneath the surface.



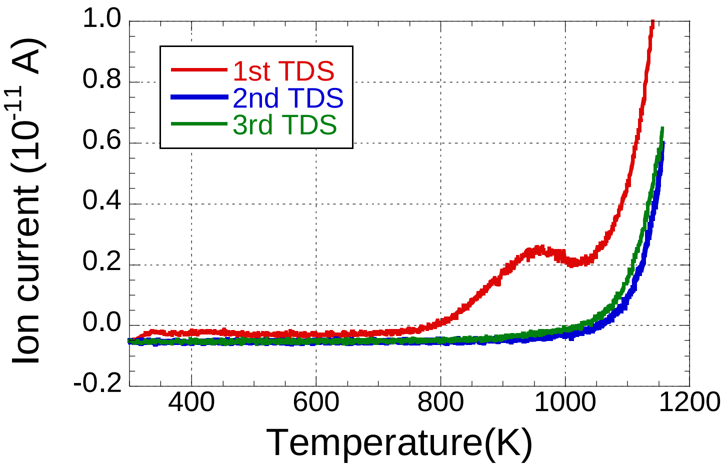
TEM micrograph after FIB process

Procedure (2): He desorption before D plasma exposure

- Three TDS measurements to check He desorption before D plasma exposure

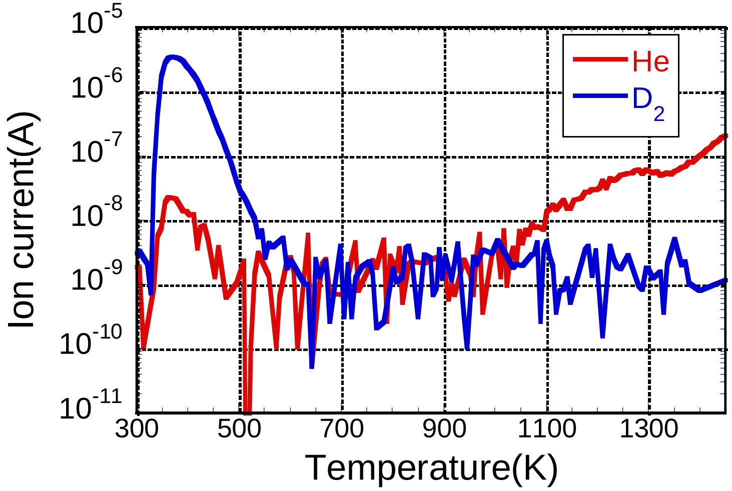
  ==> Helium was desorbed in the first TDS, but no He desorption was observed below ~900 K after 2nd TDS.

TDS spectra after He plasma exposure



- After three TDS measurements, D plasma was exposed to the sample and then TDS spectra of He and D_2 were measured simultaneously with a high-resolution QMS.
  - Contribution of He desorption to the signal of M/e=4 is considerably low (< 1%) when the temperature is below 900 K.
  - Maximum temperature of the TDS measurements with the normal-resolution QMS was set to <1000K (typical:773 K).

TDS spectra of He and D_2 after He and D plasma exposures

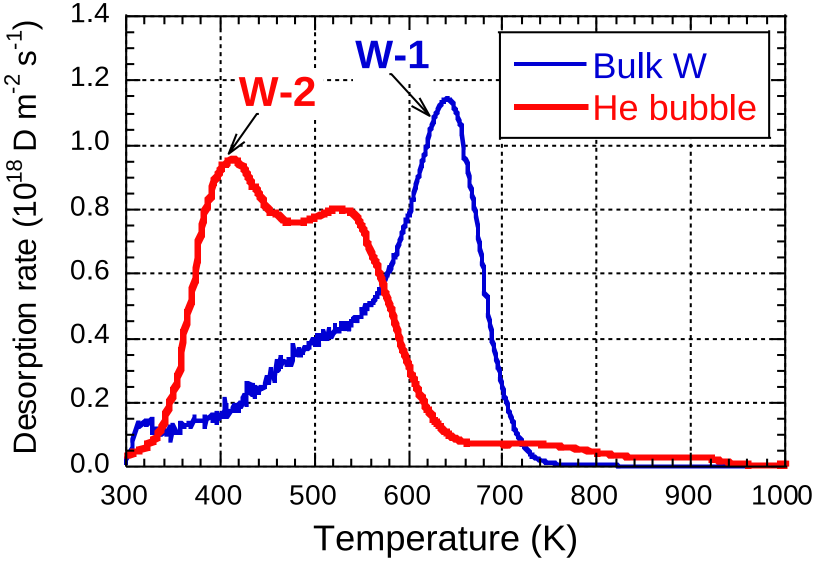


Deuterium plasma exposure

Summary of surface condition and D plasma parameters

Sample name	W-1	W-2
Pre-treatment	Annealing (1173K, 1h)	He exposure (1700K~1900K, 20-40eV, 4~9 X 10^{25} m^{-2})
Surface condition before D plasma exposure	Nothing	Fine irregularity He bubble, Hole
D plasma exposure		
Surface temp. (K)	500	510
Electron density (m^{-3})	2.5 x 10^{17}	2.4 x 10^{17}
Electron temp. (eV)	8	8
Space potential (V)	27	29
Flux (m^{-2} s^{-1})	3 x 10^{21}	2.9 x 10^{21}
Fluence (m^{-2})	2.1 x 10^{25}	2.3 x 10^{25}
Retention (m^{-2})	2.2 x 10^{20}	7.3 x 10^{20}

TDS spectra of bulk W (W-1) and W with He bubbles (W-2)



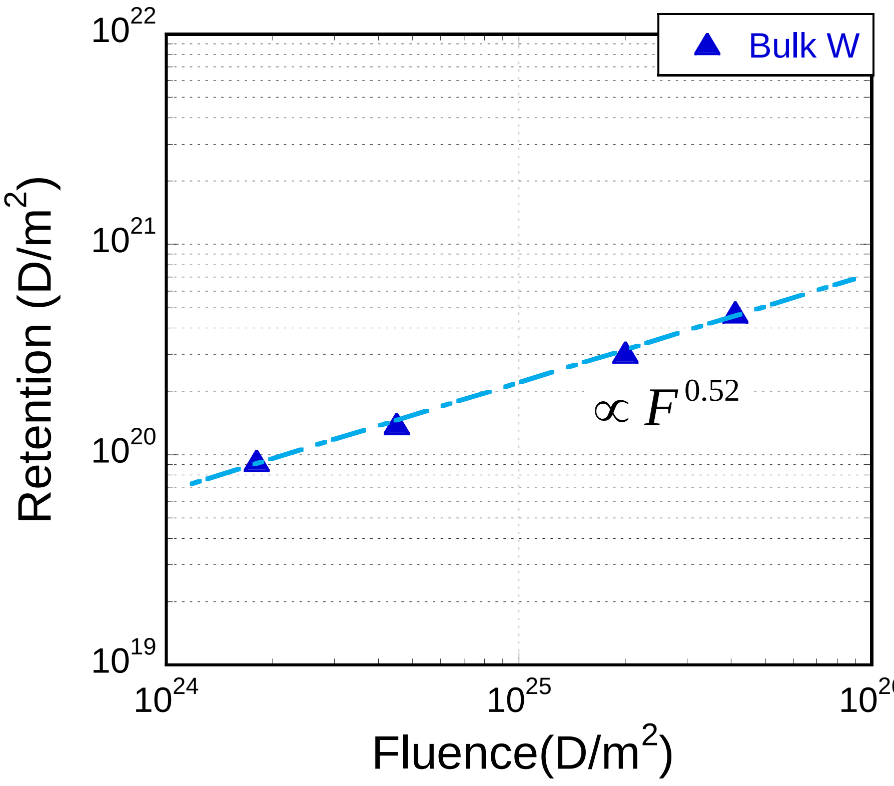
➢ The D retention in W with He bubbles increased and the desorption peak shifted toward the lower temperature, suggesting that a little weak trap sites were generated beneath the surface due to the He bubbles.

Fluence dependence

Bulk W:

- The D retention increased with square-root dependence of the fluence, indicating that diffusion is a dominant process of the D retention in the bulk W.

D retention of the bulk W as a function of the fluence



Fluence dependence

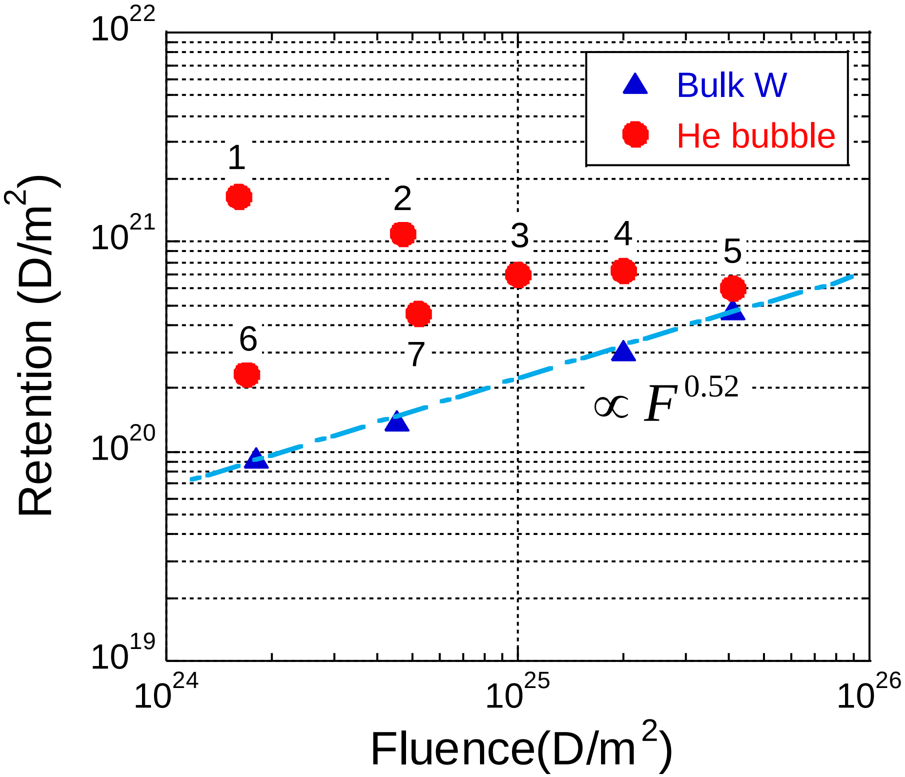
Bulk W:

- The D retention increased with square-root dependence of the fluence, indicating that diffusion is a dominant process of the D retention in the bulk W.

With helium bubbles:

- The D plasma exposure and subsequent TDS measurement was repeated 7 times using the same sample.
- The D retention is one order of magnitude larger than that of the bulk W in the low fluence case. It decreased gradually with fluence and became almost constant.
- 6th and 7th data are lower than 1st and 2nd data, respectively, suggesting annealing effect of D plasma exposure.

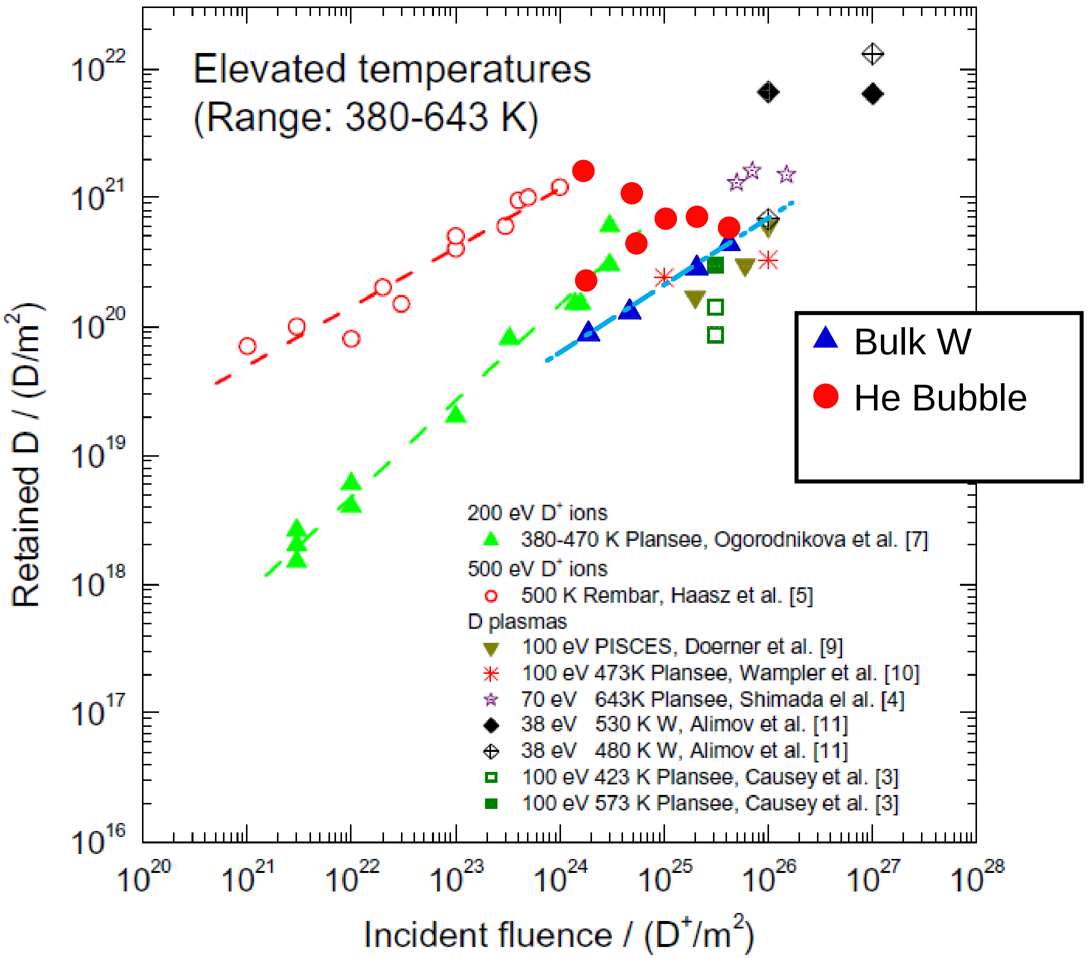
D retention of the bulk W, W with He bubbles and W with W deposition as a function of the fluence



The number indicates the order of the measurements.

Enhancement and suppression of D retention

- The helium bubbles trap considerable D atoms but simultaneously they prevent the diffusion of the D atoms to the deeper region and also generate a diffusion path to the surface for desorption of the mobile D atoms.



Z. Tian et al., JNM 399 (2010) 101. and references written therein

Summary

- Deuterium retention in tungsten (W) samples irradiated by Cu^{2+} ions and helium plasma has been investigated to study effects of the damage on hydrogen isotope retention in W.

2.4 MeV Cu^{2+} ion irradiation

- The TEM observation revealed that most of the dislocation loops (ILs) were nucleated by cascade collisions and each IL can not grow larger individually but aligned ILs grow by coalescing. Nano-voids (d<1nm) are observed in 1 dpa case and they formed densely.
- Retention in the damaged W increases with the damage level but it saturates around 0.4 dpa, suggesting that newly introduced defects may be cancelled by already existing vacancies and voids with high density.
- The desorption peak around 850 K appeared in the TDS spectra of plasma irradiation, while it did not appear in the case of ion beam irradiation.
- From comparison between TDS spectra of ion beam irradiation and plasma irradiation, the trapping around 850 K in plasma irradiation is considered seems to be attributed to the defects (i.e. voids) around the damage peak.

Summary (continued)

He plasma irradiation (He bubble effect)

- As for W with He bubbles, the D retention was one order of magnitude higher than that of the bulk W in the low fluence case. It decreased gradually with fluence and became almost constant, suggesting that the helium bubbles trap considerable D atoms, but simultaneously they work as a diffusion barrier against deeper penetration of D and generate a diffusion path for desorption of the mobile D atoms.