Deuterium retention in tungsten irradiated by heavy ions

M. Sakamoto, H. Tanaka, S. Ino,
H. Watanabe[1], M. Tokitani[2]

Plasma Research Center, University of Tsukuba, Tsukuba, Japan
[1] RIAM, Kyushu University, Kasuga, Fukuoka, Japan
[2] National Institute for Fusion Science, Toki, Gifu, Japan

3rd RCM of CRP on Irradiated Tungsten
IAEA Headquarters, Vienna, Austria
27-30 June 2017

Contents

- Main objective of this study
- Experimental set up
- Tungsten sample (TEM observation)
- Effect of heavy ion irradiation on retention in W
  - ✓ Damage level dependence
  - ✓ Flux dependence
  - ✓ Sample temperature dependence
  - ✓ Simulation results
- Summary

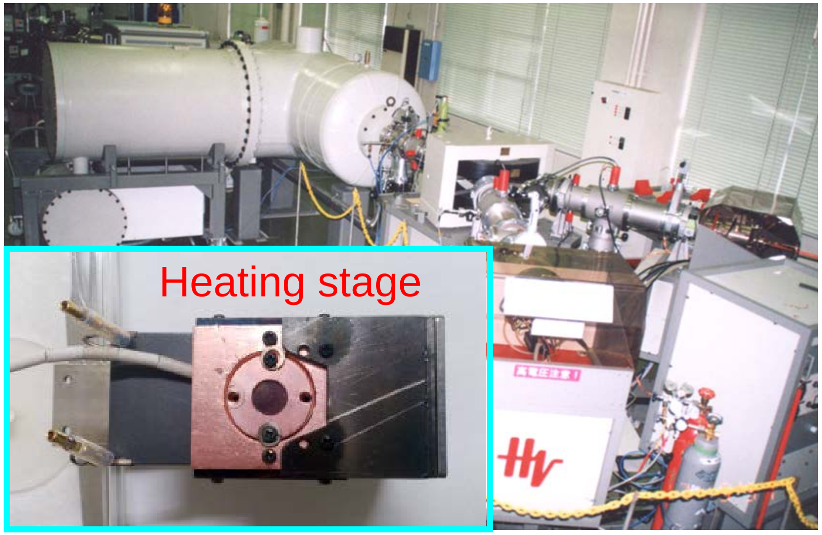
Main objective of this study

To understand the effect of neutron and surrogate irradiation upon microstructure of tungsten and

To understand the effect of the damage of tungsten on hydrogen isotope retention through the observation of change in the microstructure

In the last CRP held in Soul, I presented D retention in W exposed to He plasma in addition to preliminary results of D retention in W irradiated by heavy ions.

Experimental setup: Cu^{2+} Irradiation



Tandetron Accelerator (Kyushu Univ.)

Ion: Cu^{2+}

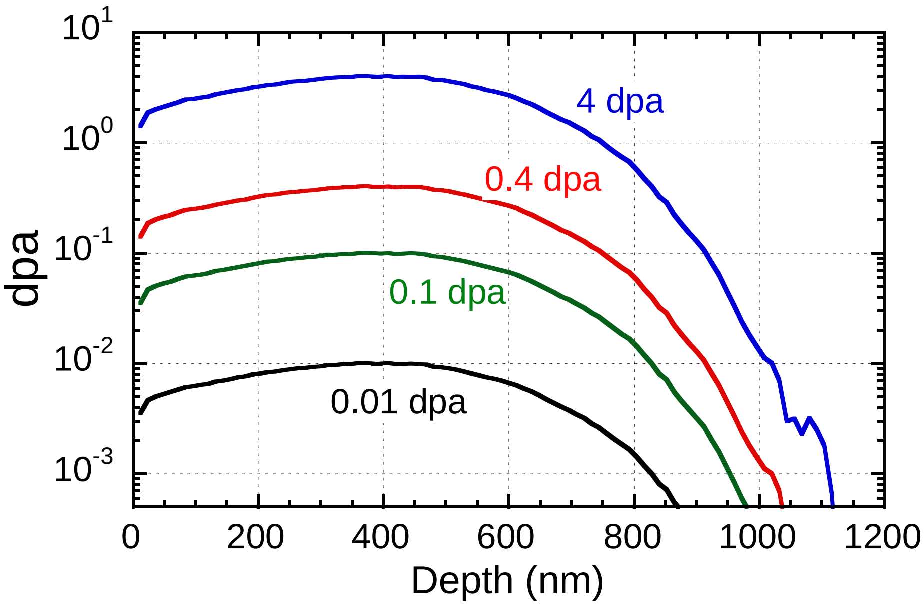
Energy: 2.4 MeV

Flux: up to 5 x 10^{15} Cu^{2+}/m^2 s

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

Sample Temp.: up to 873 K

Exposure area: 8 mmϕ

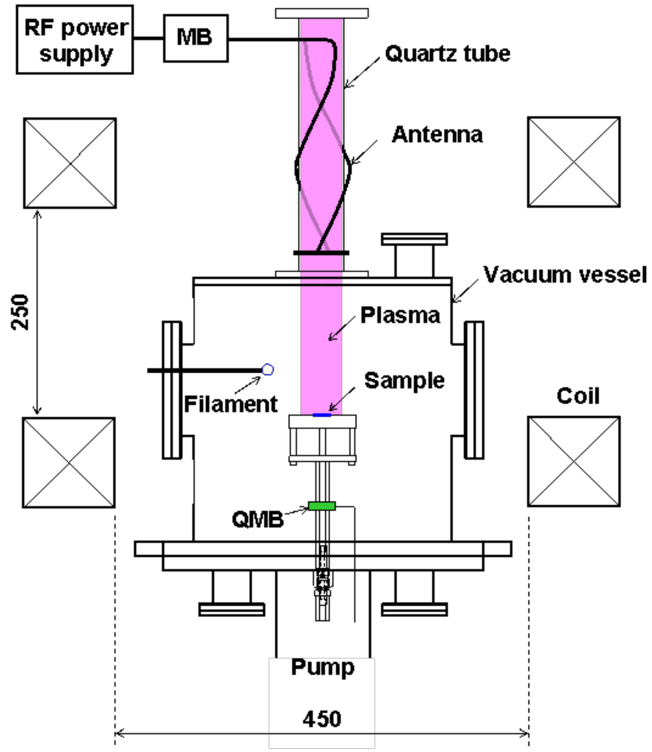


- Estimated depth profile of displacement damage in W.
- Calculation using the SRIM code with displacement energy of 55 eV.
- The peak damage region is ~400 nm.

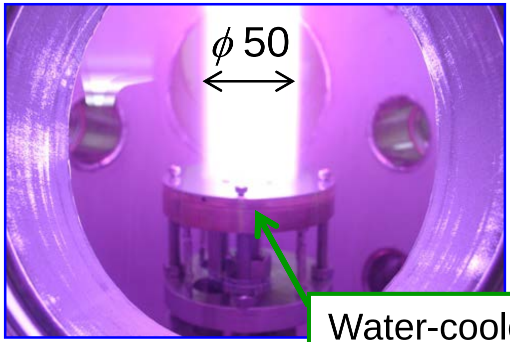
Experimental setup: Deuterium Irradiation

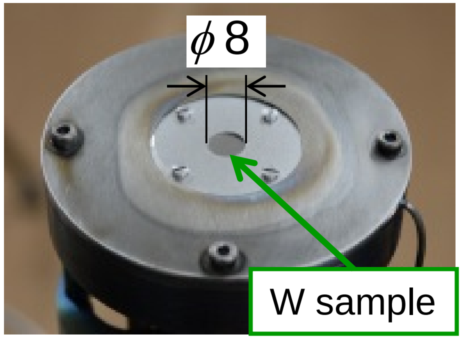
➢ Plasma exposures have been performed using a compact PWI simulator APSEDAS.

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)



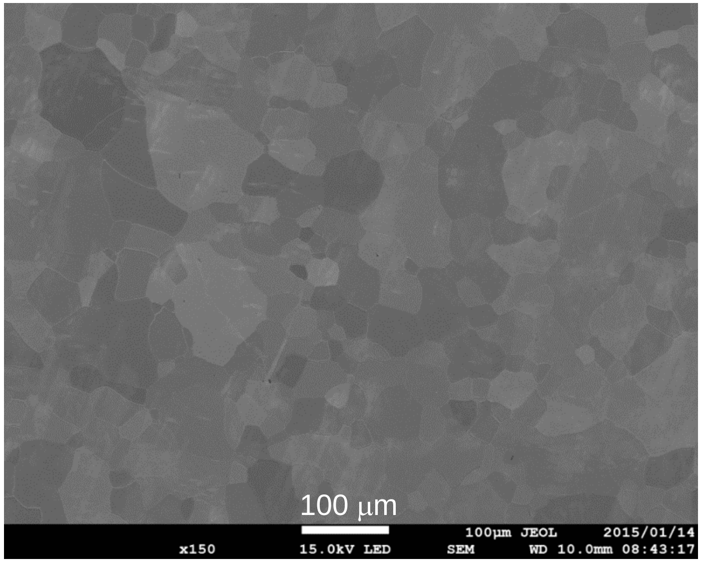


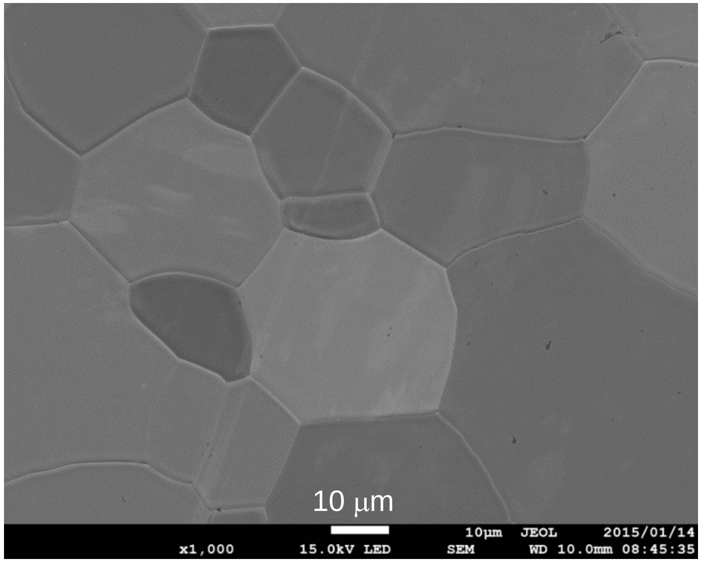
Examined Materials

Base Materials:

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

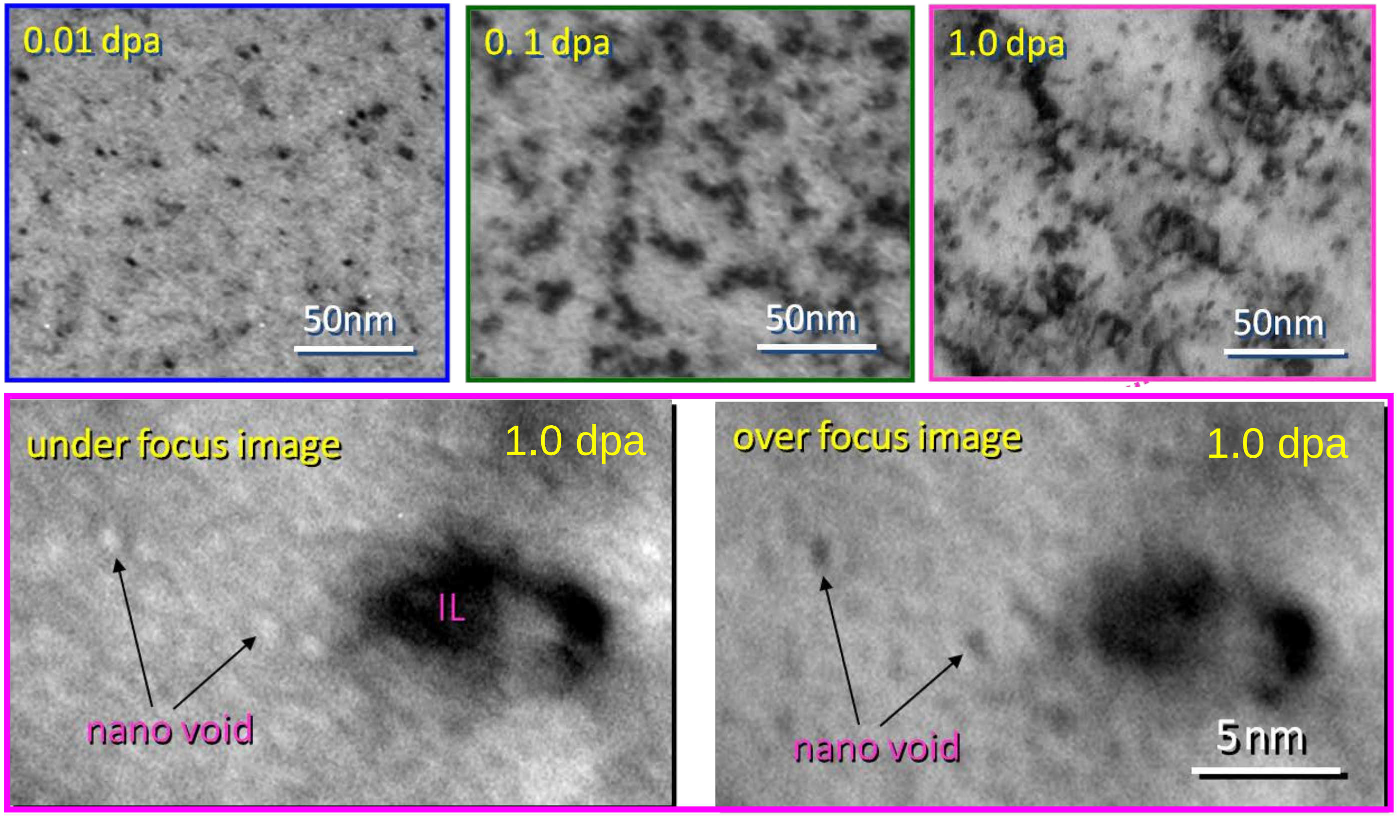
- Annealed at ~ 2000 ^{o}C for 1 h (Re-crystallized)
- The surface was mechanically polished to a mirror finish.





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

Damage evolution by 2.4 MeV Cu^{2+} irradiation



Irradiation at room temp.

- Most of interstitial loops (IL) is considered to 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 additional loops were formed in the vicinity of pre-existing dislocation loops and dislocations and aligned to coalesce with each other.
- Nano-voids (d<1nm) are observed in 1 dpa case and they formed densely.

Experimental Procedure

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

- (1) dpa dependence: 0.01 ~ 4 dpa
- (2) flux dependence: 1 ~ 5 x 10^{15} Cu^{2+}/m^2 s
- (3) sample temperature: RT, 500 K, 873 K

↓

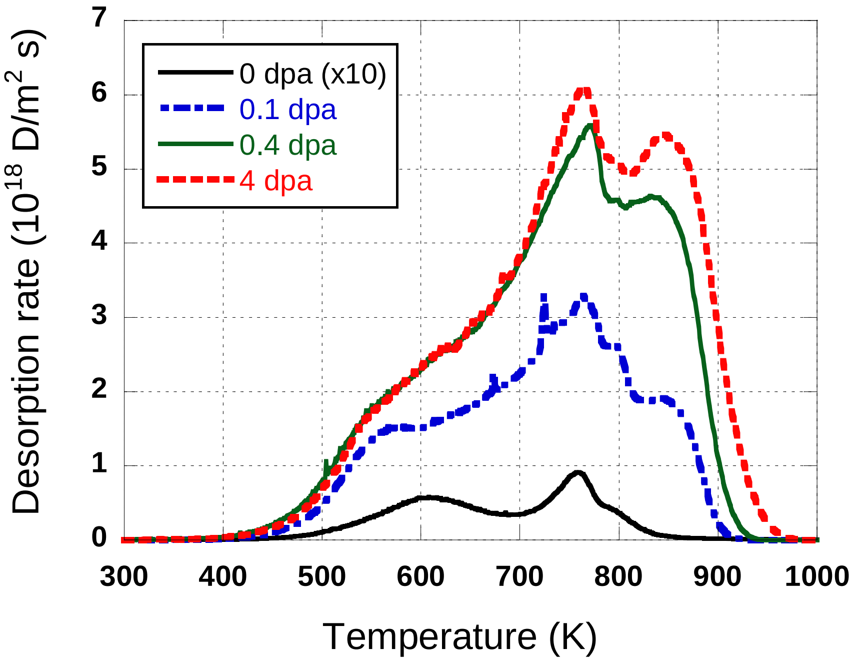
D plasma irradiation

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

↓

TDS measurement

Damage level dependence of TDS Spectrum



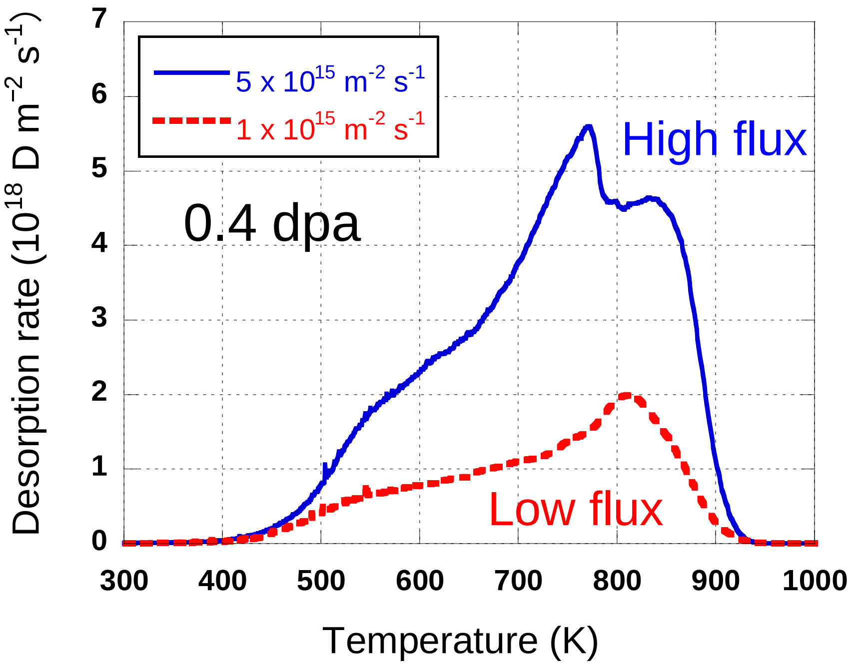
dpa	0 dpa	0.1 dpa	0.4 dpa	4 dpa
Peak 1	613	600	624	634
Peak 2	759	761	770	765
Peak 3		861	858	863

5 x 10^{15} Cu^{2+}/m^2 s

- There is three main peaks in the spectra: ~620 K, ~760 K and ~860K.
- Temperatures at 1st and 2nd peaks are the same as those of non-irradiated W.
- The 3rd peak newly appears due to 2.4 MeV Cu^{2+} irradiation and is considered to be caused by vacancy clusters and voids.
- 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.

Flux dependence of TDS Spectrum

Comparison between high and low fluxes of Cu^{2+} ions



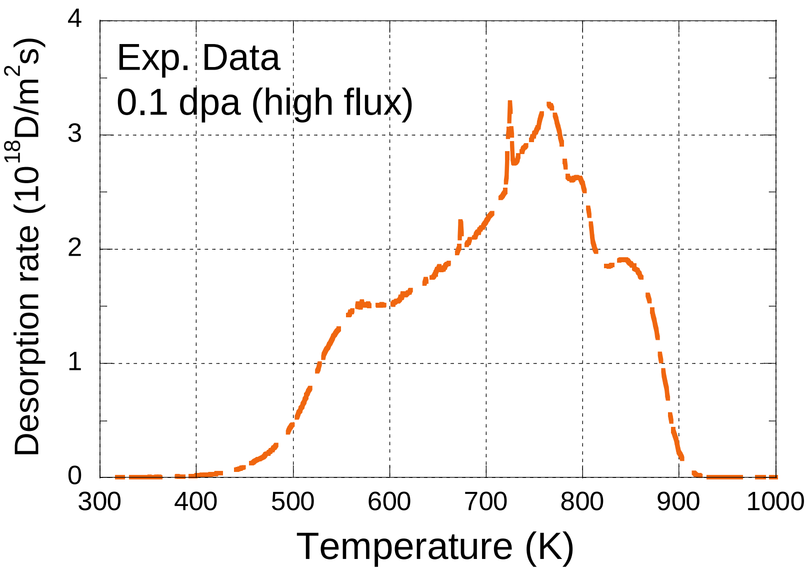
Flux	1 x 10^{15} $m^{-2}s^{-1}$	5 x 10^{15} $m^{-2}s^{-1}$
Peak 1	553 K	624 K
Peak 2	724 K	770 K
Peak 3	825 K	858 K

- Comparison between high and low fluxes of Cu^{2+} ions indicates that D retention in W irradiated with low flux Cu^{2+} ions becomes 3.5 times lower than that for high flux irradiation.
- The desorption spectrum of the low flux irradiation also consists of three stages of desorption.
- The temperatures at the three peaks for the low flux case became lower than those of the high flux irradiation.

Comparison of dpa dependence between high and low irradiation fluxes

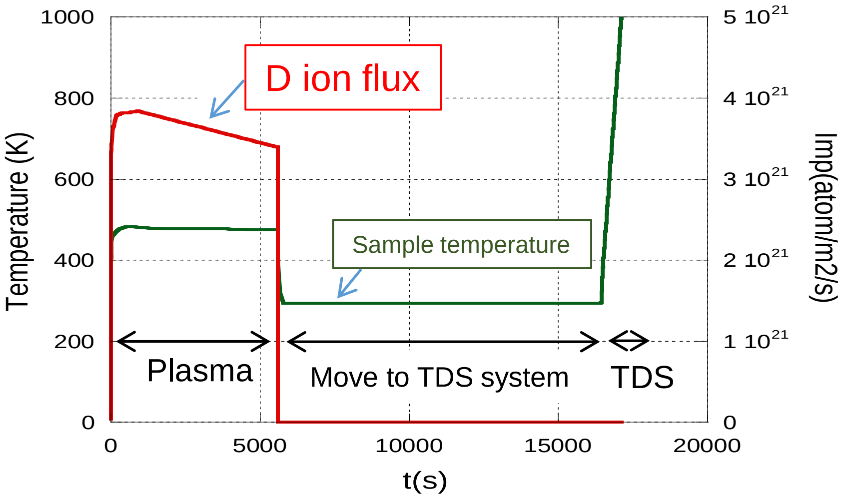
- In the case of high flux irradiation, D retention increases with the damage level but it saturates around 0.4 dpa.
- This suggests that newly introduced defects are cancelled by already existing vacancies and voids with high density.
- In the case of low flux irradiation, D retention increases with the damage level up to 2 dpa and no saturation is observed.
- It is found that defect formation due to heavy ion irradiation depends on not only a damage level but also flux of the high energy ions.

Simulation of TDS spectra using an HIDT code

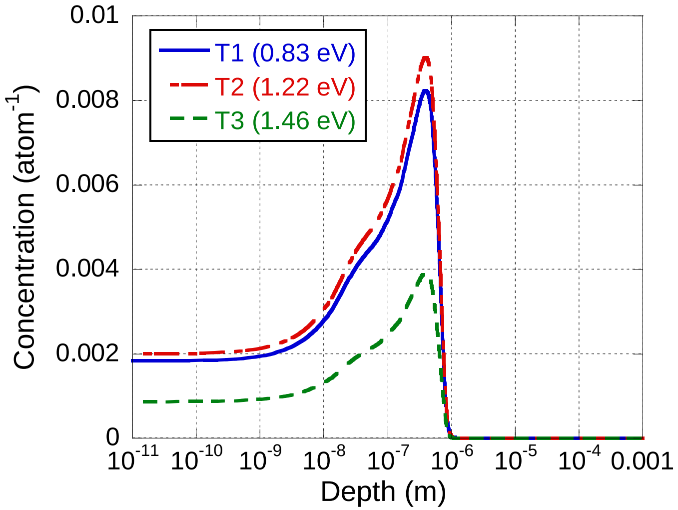


- The HIDT (Hydrogen Isotope Diffusion and Trapping) code has been developed in Shizuoka Univ.
  (ref: Y. Oya et al., JNM 461 (2015) 336.)
- Input parameters:
  D = 2.9 x 10^{-7} m^2/s (Frauenfelder)
  Kr = 3.0 x 10^{-25} m^4/s (Pick)
- Time evolution of sample temperature and D ion flux for the simulation was the same as those of the experiments.
- A depth profile of defect concentration is similar figure to the dpa profile calculated by the SRIM code.

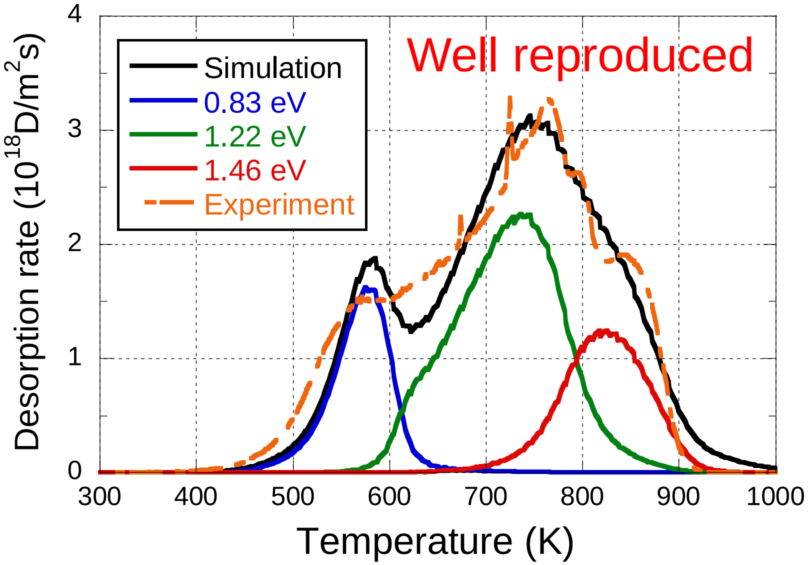
Sample temperature & D ion flux vs time



Defect concentration of each binding energy

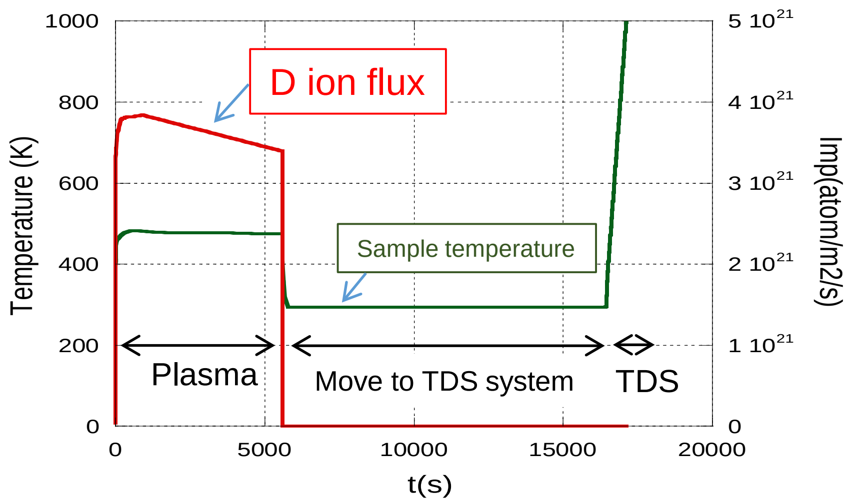


Simulation of TDS spectra using an HIDT code

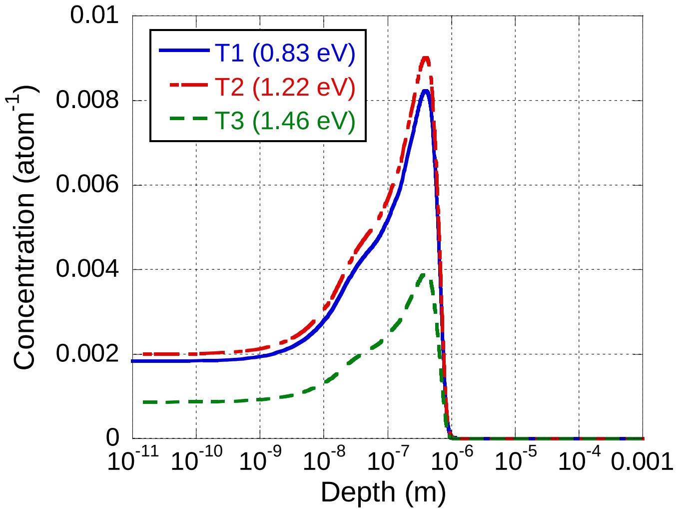


- The HIDT (Hydrogen Isotope Diffusion and Trapping) code has been developed in Shizuoka Univ.
  (ref: Y. Oya et al., JNM 461 (2015) 336.)
- Input parameters:
  D = 2.9 x 10^{-7} m^2/s (Frauenfelder)
  Kr = 3.0 x 10^{-25} m^4/s (Pick)
- Time evolution of sample temperature and D ion flux for the simulation was the same as those of the experiments.
- A depth profile of defect concentration is similar figure to the dpa profile calculated by the SRIM code.

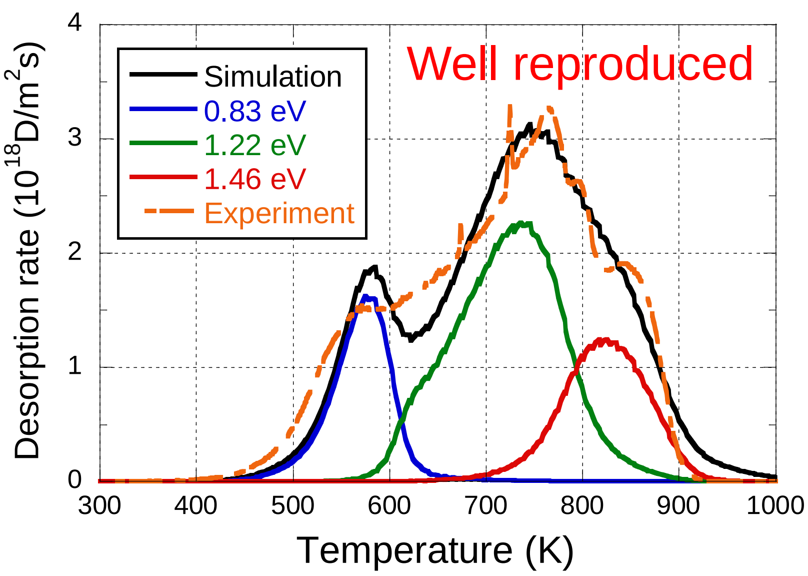
Sample temperature & D ion flux vs time



Defect concentration of each binding energy



Simulation of TDS spectra using an HIDT code



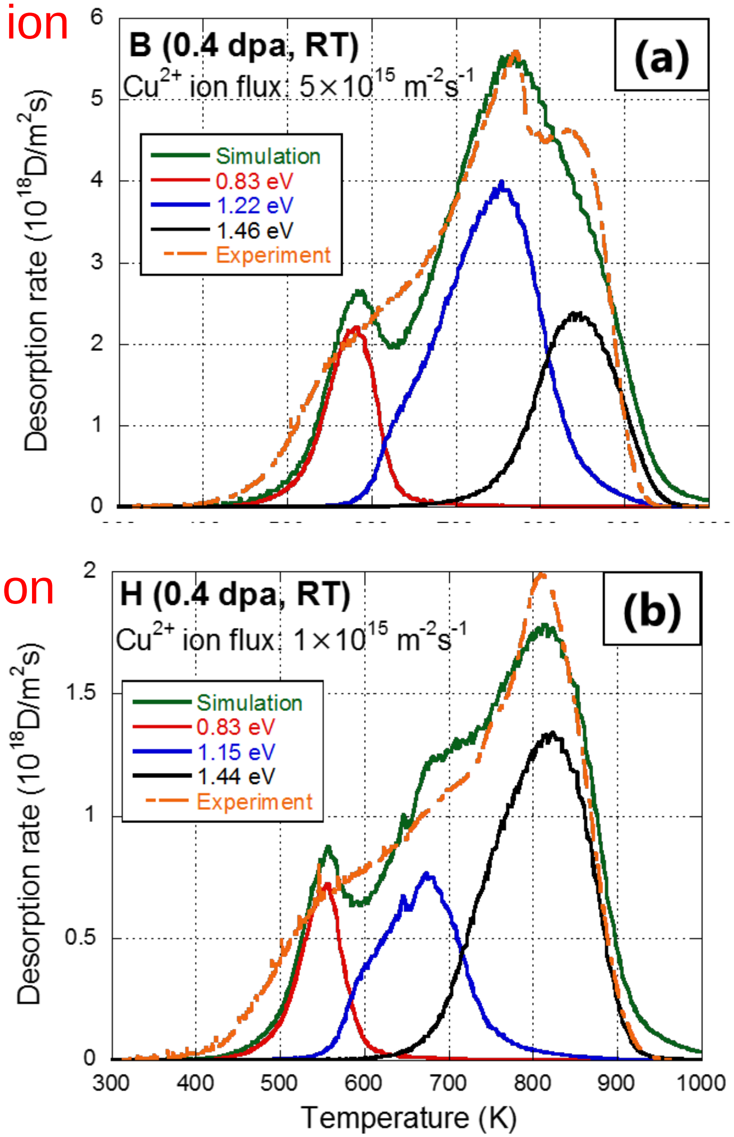
- The HIDT (Hydrogen Isotope Diffusion and Trapping) code has been developed in Shizuoka Univ.
  (ref: Y. Oya et al., JNM 461 (2015) 336.)
- Input parameters:
  D = 2.9 x 10^{-7} m^2/s (Frauenfelder)
  Kr = 3.0 x 10^{-25} m^4/s (Pick)
- Time evolution of sample temperature and D ion flux for the simulation was the same as those of the experiments.
- A depth profile of defect concentration is similar figure to the dpa profile calculated by the SRIM code.

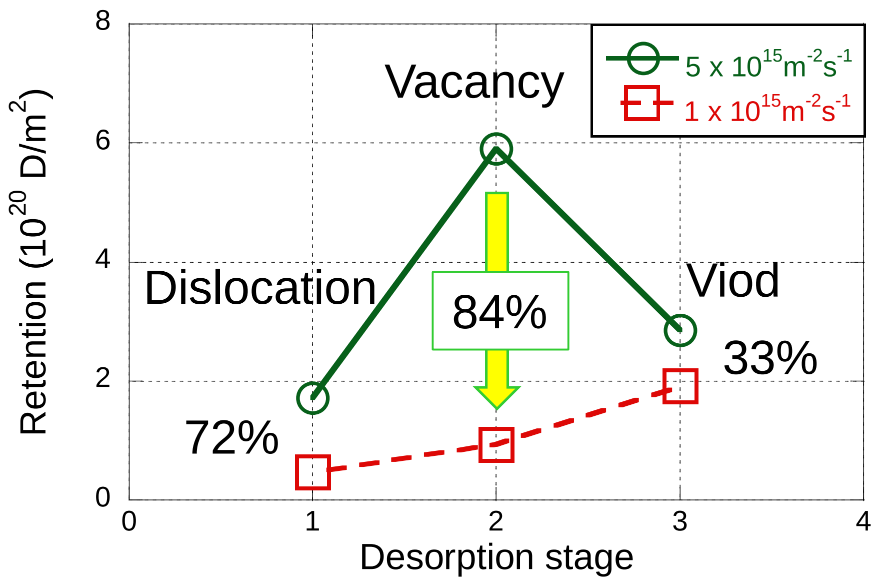
Peak	Binding energy	Corresponding defects
1st	0.83 eV	dislocation loops & grain boundary
2nd	1.22 eV	vacancy
3rd	1.46 eV	vacancy cluster & void

Simulation of TDS spectra using an HIDT code

High ion flux

Low ion flux



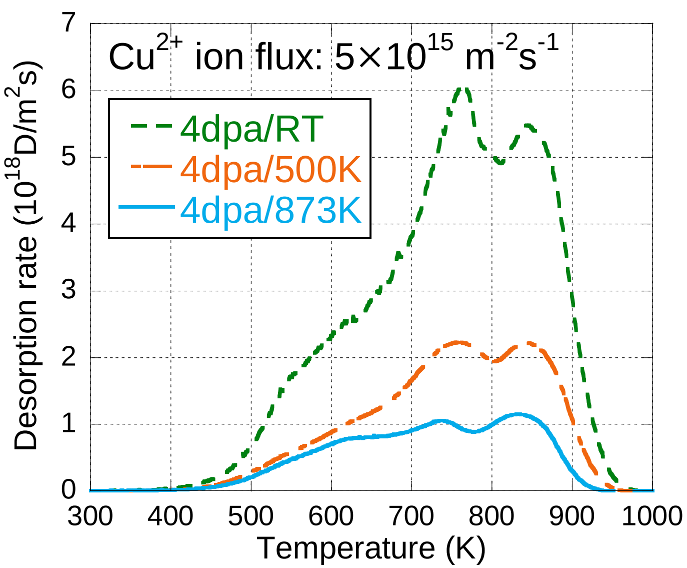


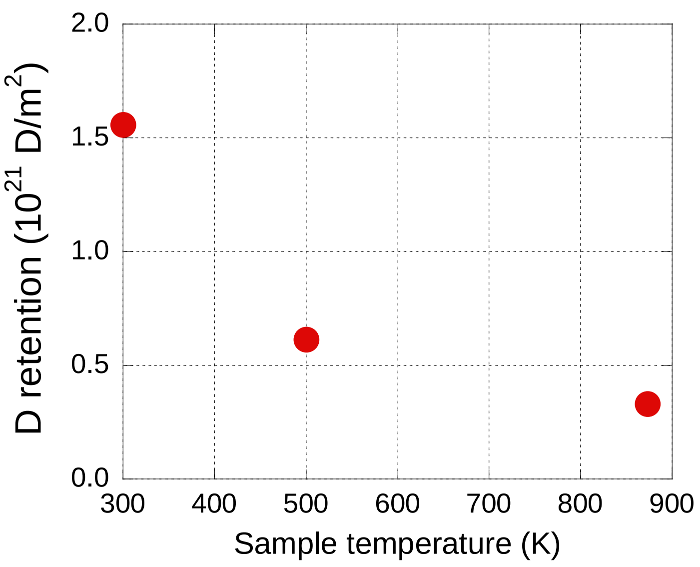
D retention in the 2nd stage (i.e. vacancy) significantly decreased with decrease in ion flux (i.e. dpa rate), while D retention in 3rd stage decreased not so much.

Speculation

High dpa rate: vacancies may be remained.
Low dpa rate: voids may be produced.

Dependence of D retention on sample temperature druing Cu^{2+} ion irradiation





- Cu^{2+} irradiation was carried out at the sample temperature of RT, 500 K and 873 K.
- Desorption rate decreased as a whole with the sample temperature during the irradiation.
- For the sample temperature of 500K, 873 K, D retention decreased by 60 % and 80 %, respectively.

Two possible candidates

1. Interstitial and vacancy may annihilate each other with increase in the sample temperature.
2. At the sample temperature of 873 K, Interstitials may escape to grain boundary and remained vacancies coalesce to become void.

Question

Which is dominant for D retention, large number of vacancies or smaller number but bigger voids?

Summary

Deuterium retention in tungsten (W) samples irradiated by 2.4 MeVCu^{2+} ions has been investigated to study effects of the damage on hydrogen isotope retention in W.

TEM observation

- Most of the dislocation loops (ILs) were nucleated by cascade collisions.
- Additional loops were formed in the vicinity of pre-existing dislocation loops and dislocations and aligned to coalesce with each other.
- Nano-voids (d<1nm) are observed in 1 dpa case and they formed densely.

TDS spectra

- There exists three desorption peaks in the TDS spectra . The desorption peak around 850 K newly appeared due to Cu^{2+} ions irradiation. This peak must be related to voids.

Flux and damage level dependences

- When the Cu^{2+} ion flux decreased by 5 times, the D retention decreased by 3.5 times, indicating clear flux dependence.
- In the case of higher ion flux, D retention 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.

Summary (continued)

Flux and damage level dependences

- On the other hand, In the case of low flux irradiation, D retention increases with the damage level up to 2 dpa and no saturation is observed.

Sample temperature dependence

- Desorption rate decreased as a whole with the sample temperature during the irradiation.

Simulation using the HIDT code

- TDS spectrum was reproduced by using the HIDT code using experimental data. The binding energies of three desorption peaks are 0.83 eV, 1.22 eV and 1.46 eV.

Speculation

High dpa rate: vacancies may be remained.
Low dpa rate: voids may be produced.

It is necessary to know effects of the number of defects and their size on D retention.
It is planed to measure TEM using samples which have irradiated at different sample temperature.