Analysis of Tungsten Long-term Retention and Re-emission with Ion-induced Defect Generation under Ion Oversaturation Condition

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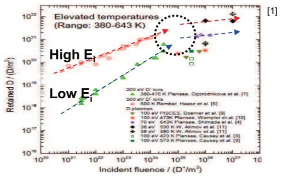
Motivation: Necessity of Dynamic Retention Analysis

Previous Long-term retention analysis

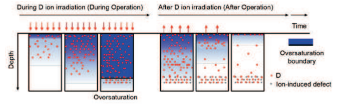


Reported 'fluence to retained-D scale' shows,

1) inflection point b/w low and high fluence.
2) Low retained-D for low E_i case but same infection point, implying that ion-induced defect effect with fluency dependency.

Defect generation mechanism

Tanabe et al and most PMI research reaches to consensus on ion-induced defect generation possibility by oversaturation even though low incident ion energy condition (E_i < 200eV). [2]



Diffusion of D atom into bulk induces **oversaturation**. and it generate ion-induced defect due to stress field [2].

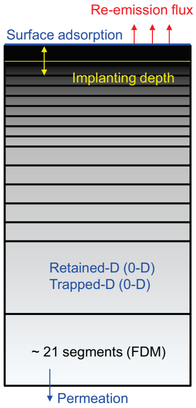
Long-term retention is equilibrium phase after dynamic retention and plasma permeation into material. So retention analysis is incomplete without dynamic retention analysis with consideration on defect generation.

➔ Present work concentrates on that **effect of ion-induced defect generation on dynamic and long-term retention with currently suggested ion-induced defect generation mechanism of oversaturation.**

[1] Z. Tian et al., Journal of Nuclear Materials 399 (2010) 101–107 Contents
[2] T. Tanabe, Phys. Scr. T159 (2014) 014044

Time-transient Retention Analysis with TMAP4 Simulation

Schematics of TMAP4 Simulation



Main control input

- **Control input.1: plasma exposure condition**
  1. T_{surf}, P_{surf}
  2. Ion incident condition

Parameter	Value
D Ion flux, Γ_i	8.4x10²¹ D⁺/m²-s
$f_{backscattering}$	0.486 (TRIM result)
Effective D ion flux, $\Gamma_{i,eff}$	4.090x10²¹ D⁺/m²-s
Impact energy, E_i	33 eV/D⁺
Ion implanting depth	1.5 nm

- **Control input.2: Trap/detrap condition**
  1. Trap type: vacancy, 10 at%
  2. Trapping rate (D-vacancy)

$$K_{trap} = 9.13\times10^{12}\exp(-0.39eV/kT)$$

  3. De-trapping rate (vacancy)

$$K_{de-trap} = 8.4\times10^{12}\exp(-1.45eV/kT)$$

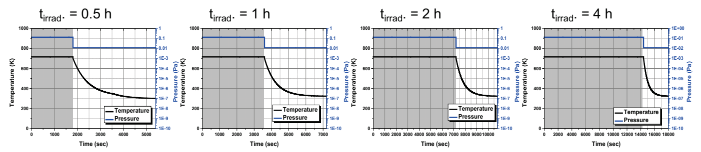
Main output

Long-term retention parameter
1) Soluted-D concentration
2) Trapped-D concentration

Dynamic retention parameter
1) Surface adsorption concentration
2) Surface re-emission flux
3) Backside permeation concentration

Assumed Input Condition for TMAP4 for No Defect in Tungsten

- **Simulation condition: D ion fluence 0.5-4.04 x10²⁵ D/m² (0.5 h - 4 h)**
- **Variable for plasma exposure: Exposure time (Fluence with const. flux)**
  - ➔ **Total simulation time = Plasma exposure time (0.5, 1, 2, 4 h) + Cooling time (1 h)**
- **Input control variable: Temp., Press., Fluence, Trapping/Detrapping**

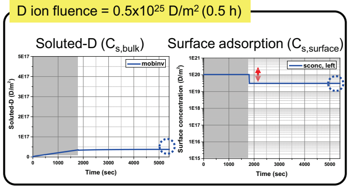


Condition		0.5 h	1 h	2 h	4 h
	Irradiation Time [h]	0.5 h	1 h	2 h	4 h
	Cooling time [h]	1 h	1 h	1 h	1 h
	D ion fluence [D/m²]	0.50 x 10²⁵	1.01 x 10²⁵	2.02 x 10²⁵	4.04 x 10²⁵
1. without ion-induced defect generation by oversaturation		No trapping $K_{trapping}=0$ $K_{de-trapping}=0$			

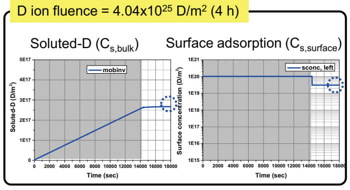
Long-term Retention Estimation without Defect

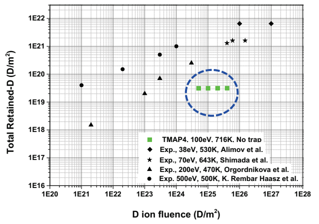
- **No defect condition ➔ Long-term retention = Solute-D + Surface adsorption**

D ion fluence = 0.5x10²⁵ D/m² (0.5 h)



D ion fluence = 4.04x10²⁵ D/m² (4 h)





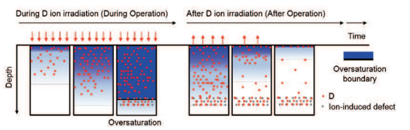
Simulated long-term retention amount can't follow the previous experimental finding especially,

1. **Slope of fluence to retained-D amount**
2. **Absolute total amount**

➔ Retention should be estimated with defect effect according to experimental findings on defect generation fluence condition.

Current Consensus on Ion-induced Defect Generation Mechanism

Oversaturation and typical defect type as a D trapping site



D atom diffuse into bulk induced oversaturation, and it generate ion-induced defect due to stress field [3].

➔ there's no study to estimate the effect of oversaturation on retention.

Category	Typ.conc. [at. %]	Defect type	Generation mechanism	Reemis. energy [eV]	Form of retained-D	TDS peak	Ref.
Intrinsic defect	4 x 10⁻⁴ to 10⁻²	Point defect, grain boundary,	purity, surface preparation	~ 0.85	D_2 (pore)	340-560 K	[3]
					D (chemisorp.)	600 K	[6]
Ion-induced defect	~ 10	dislocation	Cascade collision	0.85	D (dislocation)	~700 K	[5]
		vacancy	**Oversaturation**	**1.45**	**D (vacancy)**	**800-900 K**	**[5]**
n-induced defect	~ 1	vacancy clusters, voids	Displacement event	1.8-2.2	D, D_2 (vacancy)		[4] [6]

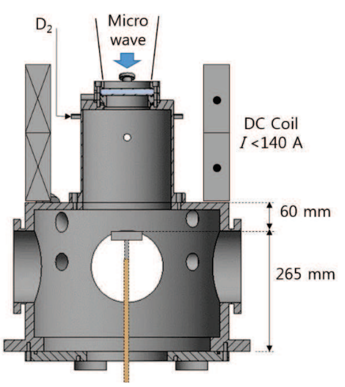
- Ion-induced defect is most effective long-term retention contributor when there's negligible intrinsic defect and n-induced defect such as ITER condition. And it proportional to ion fluence up to 10 at%.

➔ **ion-induced defect can be increased up to 10 at% after oversaturation for sufficient ion fluence.**

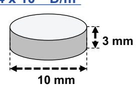
[3] T. Tanabe, Phys. Scr. T159 (2014) 014044
[4] Joachim Roth and Klaus Schmid, Phys. Scr. T145 (2011) 014031 (9pp)
[5] Ogorodnikova O V, Roth J and Mayer M, 2003 J. Nucl. Mater. 313–316 469
[6] Tyburska B, Alimov V Kh, Ogorodnikova O V, Schmid K and Ertl K 2009 J. Nucl. Mater 395 150

Demonstration of Defect Generation during Plasma Exposure

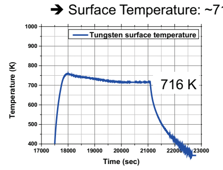
Electron Cyclotron Resonance (ECR) Plasma (1D SOL Simulator)



- Electron Temperature (T_e) : 4.7 eV
- Target Bias: -87 V (Plasma potential +13 V)
- Ion impact energy (E_i): 100 eV (Low E_i)
- Ion flux (Γ_i): 2.8 x 10²¹ m⁻²s⁻¹
- Ion fluence: 0.5 - 4.04 x 10²⁵ D/m²
- Tungsten Specimen

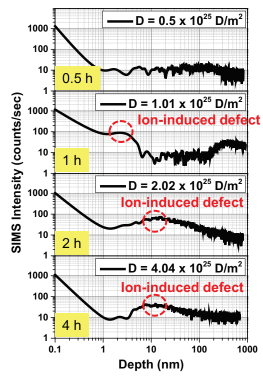


- $T_{surf.}$ during irradiation from pyrometry
  - ➔ Surface Temperature: ~716 K

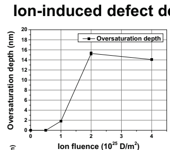


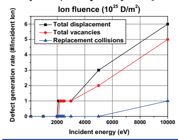
Threshold D Ion Fluence for Defect Generation by Oversaturation

Retained-D depth profile



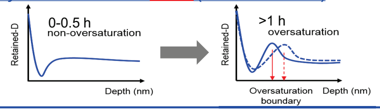
Ion-induced defect depth and type





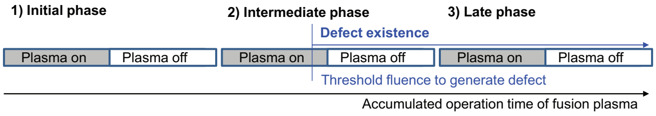
- From SIMS results, location of defect is between 1 nm to 16 nm with similar peak intensity
  - ➔ No difference in defect quantity
- It generated by oversaturation not cascade collision due to low ion impact energy than damage threshold energy. ➔ vacancy

Ion induced defect generation by oversaturation: b/w 0.5-1 h (0.5-1.01x10²⁵ D/m²)



Condition to Evaluate Defect Generation Effect on Dynamic Retention

- Defect existence condition ➔ Long-term retention = surface adsorption + soluted-D + trapped-D
- Ion-induced defect generation fluence is assumed as 0.75 x 10^{25} D/m^2 from SIMS results.
- 3 typical condition of retention with defect generation in shot-by-shot operation of fusion plasma (Repetition of plasma on-plasma off).



Simulation Condition	Irradiation Time [h]	0 – 2700 s	2700 – 14400 s(45m - 4h)
	D ion fluence [D/m^2]	0.50 – 0.75x 10^{25}	0.75 -4.04 x 10^{25}
Condition.1) Initial phase		No trapping (No defect)	
Condition. 2) Intermediate phase		No trapping	Trapping/De-trapping (from 45 min)
Condition. 3) Late phase		Trapping/De-trapping	

From 1) dynamic retention with no trap case can be understood.

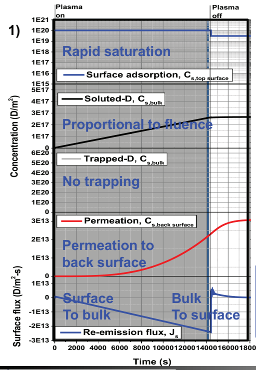
From 2) changes of dynamic retention due to defect generation during plasma-on phase can be analyzed.

From 3) changes of dynamic retention with tungsten with defect (generated early shot) can be estimated.

Changes in Dynamic Retention by Defect Generation

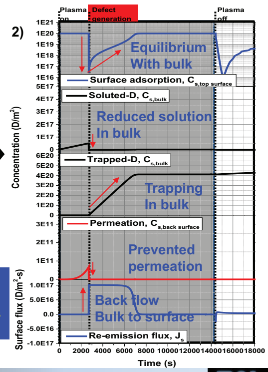
Simulation Condition	Irradiation Time [h]	0 – 2700 s	2700 – 14400 s(45m - 4h)
	D ion fluence [D/m^2]	0.50 – 0.75x 10^{25}	0.75 -4.04 x 10^{25}
1) Initial phase		No trapping	
2) Intermediate phase		No trapping	Trapping/De-trapping (from 2700 s)

Simulation range



Changes in Dynamic retention 1) ➔ 2)

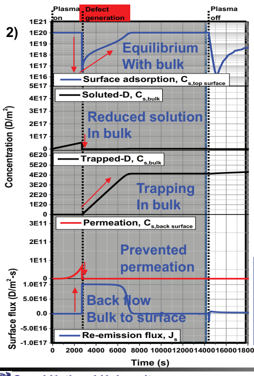
Defect generation during plasma exposure Makes prominent changes in all dynamic properties



Changes in Dynamic Retention by Defect Generation

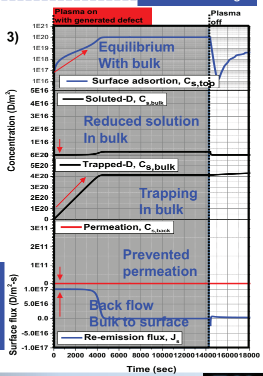
Simulation Condition	Irradiation Time [h]	0 – 2700 s	2700 – 14400 s(45m - 4h)
	D ion fluence [D/m^2]	0.50 – 0.75x 10^{25}	0.75 -4.04 x 10^{25}
2) Intermediate phase		No trapping	Trapping/De-trapping (from 2700 s)
3) Late phase		Trapping/De-trapping	

Simulation range



Changes in Dynamic retention 2) ➔ 3)

For long-term operation, the result for intermediate defect generation and initial defect result in consistent result.



Effect of Defect Generation on Dynamic Retention Property

Surface

Mass conservation at the surface

$$\sum_i a_{m_s}\left(P_{m_i}K_{d_{m_i}} - K_{r_{m_i}} - C_{s,surface}C_j\right) + D_s\left(\nabla C_{s,bulk} + C_{s,bulk}\frac{Q_s^*}{kT^2}\nabla T\right) + \sum_j \frac{C_{s,surface}C_jD_s}{N\lambda} + C_{s,surface}\lambda\left[\nu_0 \exp\left(-\frac{E_b}{kT}\right) - \upsilon_s\right] = 0$$

① net flux to surface of molecule ② atomic flux diffusing from bulk to surface ③ rate of recombination ④ rate of dissociation

Re-emission flux↑ (bulk-to-surface)

↑ Enhanced 'Backflow' ↑ from bulk to surface

↑ Negative gradient of D concentration (-∇C)

↑ Trapped-D ↑(> Surface), Soluted-D ↓

D atom flux From surface to bulk

↓ D oversaturation In bulk

↓ Ion-induced defect generation

Bulk

Sivert's law: $C_{s,bulk} = K_s P_m^{1/2}$

$C_t^e = C_t^0 - \sum_j C_j^t$: Balance of trapping site

Conservation eq. of atom 's': $\frac{\partial C_{s,bulk}}{\partial t} + \nabla \cdot J_s = S_s - \frac{\partial C_{s,bulk}^t}{\partial t}$ $\quad \frac{\partial C_{s,bulk}^t}{\partial t} = \alpha_{t_s}\frac{C_t^e}{N}C_{s,bulk} - \alpha_{r_s}C_{s,bulk}^t$: Balance of trapped-D

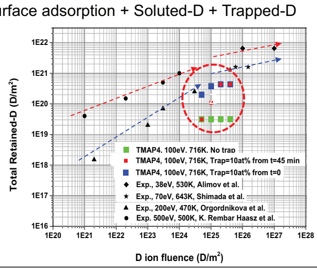
Defect generation changes surface mass conservation as well as bulk trap-to-solution balance.

Long-term Retention Estimation with Consideration on Defect

Fluence to Retained-D Scaling

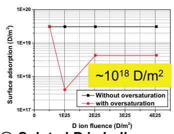
Total retained-D

= Surface adsorption + Soluted-D + Trapped-D

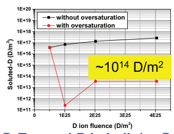


- Along 3 typical condition of 1)Green, 2)Red, 3)Blue, 3) late phase has greatest correspondence with experimental and it explains saturated trend of experiments ➔ Saturation trend mainly comes from trapping limit of trapped-D as shown in ③ ➔ it implies that under fusion-relevant plasma, tungsten has ion-induced defect from the early state of operation.

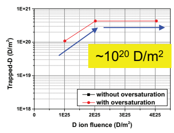
① Surface adsorption



② Soluted-D in bulk



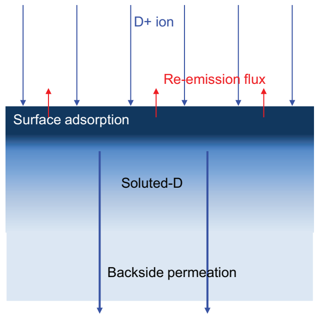
③ Trapped-D in bulk (>>①,②)



Schematics Diagram for Dynamic Retention Changes by Defect

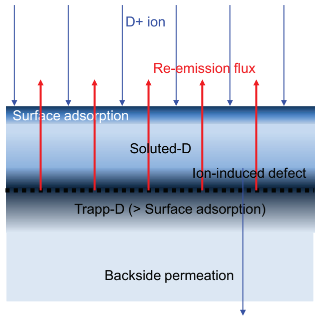
- **Lower fluence than defect generation fluence (Φ_D < 0.75 x 10^{25} D/m^2)**

Retention	Low conc. High conc.
= Surface adsorption + Soluted-D	



- **Higher fluence than defect generation fluence (Φ_D > 0.75 x 10^{25} D/m^2)**

Retention	Low conc. High conc.
= Surface adsorption + soluted-D + trapped-D	



Conclusion

- Typical 3 condition was evaluated with different defect generation (existence) time based on threshold fluence of ion-induced defect generation. ➔ For long-term operation, the result for intermediate defect generation and initial defect result in consistent result.
- Saturation trends of previous experimental researches are clarified in this work. The trends comes from mainly trapping limit of defect rather than solubility limit of D in tungsten.
- Additionally, unexpected phenomenon was expected, which is enhanced surface re-emission flux by 'backflow'. It was observed in TMAP4 simulation based on higher bulk-to-surface D atom flux due to formation of negative concentration gradient from bulk to surface by trapping.
- This work suggests defect generation affect the changes of on-pulse retention dynamic as well as long-term retention amount. so it is significant to not only safety limit but also operation stability.