Material mixing of tungsten with low Z materials -Carbon and Helium-

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ITER International Summer School 2009
"Plasma Surface Interaction in Controlled Fusion Devices"
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Outline of this talk

- Material mixing of C & W : introduction
- C deposition on W
- Effects of C & W mixing on retention/blistering
- Effects of simultaneous He bombardment to W on retention/blistering

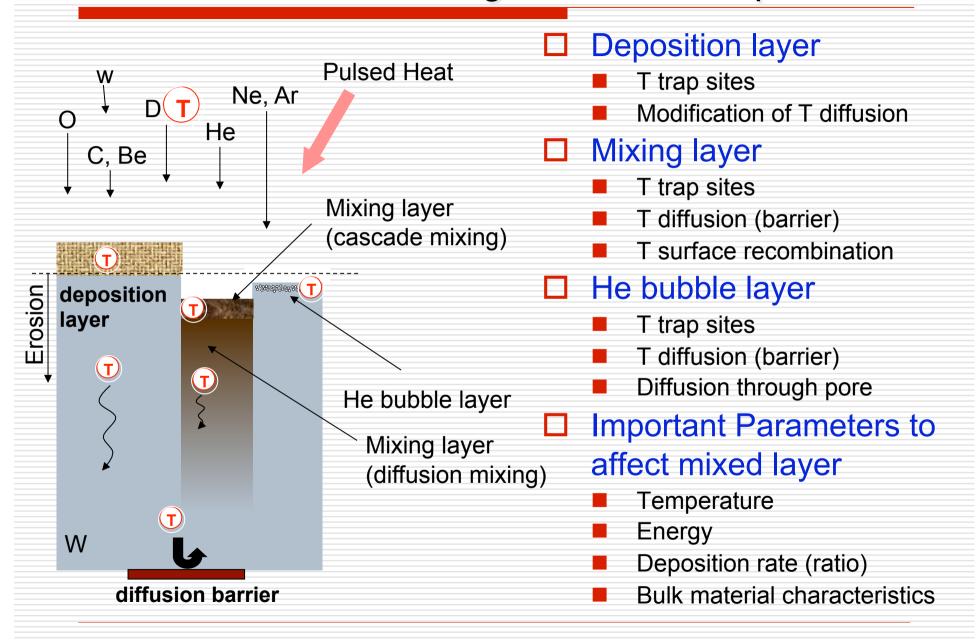
Wall material selection in ITER

- □ CFC : T retention problem (associated with significant erosion) could greatly reduces DT shots number
- Tungsten : several concerns such as Melting, Cracking, Helium embrittlement, Core plasma contamination.
- In terms of T retention, a full W wall is a better choice.
 - But, several issues need to be settled for the use of full W
 - In H phase, W, C and Be are used to learn ITER plasma operation toward full W.
- □ W+C (CFC) system is still one option for DT operation.
- ☐ Material mixing of C and W is a very important subject.
- □ In addition, He mixing effects are significant in T retention.
- □ W-Be and C-Be mixing are also an important issues.
 - Be issues will be discussed in Prof. Tynan's talk.

Research on W & C material mixing

- ☐ Many basic studies have been done in C+D→W, but still quite a few remaining issues (deposition, effects on retention)
- Several complicated processes need to be considered.
 - Dynamic mixing process
 - ☐ Mixed layer formation during ion irradiation (non-equilibrium state)
 - ☐ Multiple ion irradiation (D/T, He, Ar(edge cooling), C(wall), O, etc.)
 - Thermal processes of C (W, D) in mixed layer
 - Chemical sputtering of C in mixed layer and deposition layer
 - Depending on chemical state and micro-structure
 - Ion radiation enhanced processes
 - Radiation enhanced diffusion and segregation
- Necessary to consider actual conditions
 - Roughness(surface morphology)
 - Surface impurities (Ex. Oxide layer)

Effects of material mixing on T retention/permeation



Balance between C implantation and erosion

- □ C implanted
 - = <u>C injected</u> <u>C reflected</u>
- C erosion
 - by all ions
 - Physical sputtering
 - Radiation enhanced sublimation
 - by hydrogen isotope ions (oxygen)
 - Chemical sputtering
 - C in mixed layer
 - C in deposition layer
 - Sublimation (at elevated temperatures)
 - Flaking, exfoliation, or dust emission (for thick D deposition)
- ☐ C diffusion into the bulk

The simplest model for C balance (H+C ions)

$$\Gamma_{i} f_{C} \left(1 - R_{CC} \Delta_{C} - R_{CW} \left(1 - \Delta_{C} \right) \right) = \Gamma_{i} \left(\left(1 - f_{C} \right) Y_{HC} \Delta_{C} + f_{C} Y_{CC} \Delta_{C} \right)$$
Injected C C reflection by W (2) C sputtered by H(3) C sputtered by C (4)

 Δ_c : Surface coverage of C (Δ_c =1: fully covered by C)

[: Ion influx

f_C: C concentration in injected ions

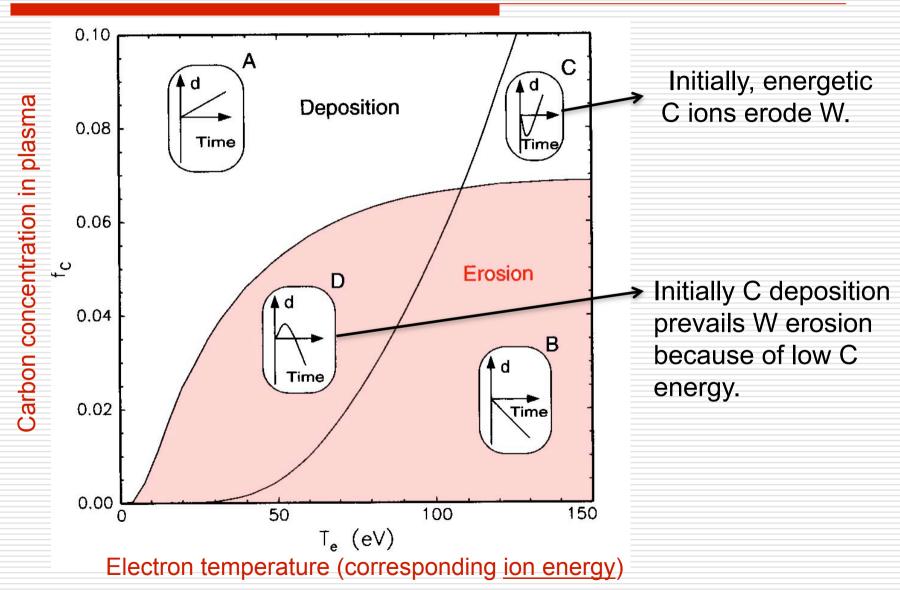
R_{CC}: C reflection coefficient on C, R_{cw}: C reflection coefficient on W

Y_{HC}: Sputtering yield of C by H, Y_{CC}: Sputtering yield of C by C (self-sputtering yield)

More complicated for real system

- How to determine the thickness of layers for △_c
 - □ Actually, for (1), (2), (3), and (4), thickness is different.
- Sputtering and reflection are not simple linear function of Δ_c .
- How does thermal effect play roles?
 - ☐ Surface segregation, diffusion

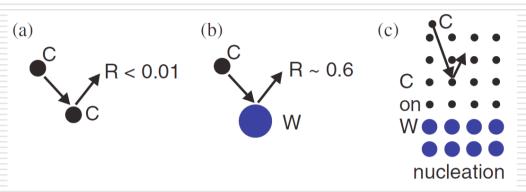
Initial surface evolution under D+C →W



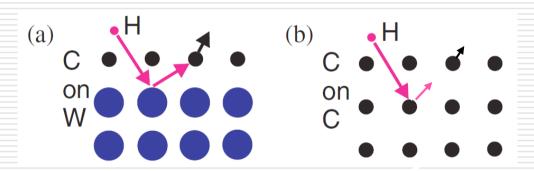
"Plasma Material Interaction in Controlled Fusion", D. Naujokes, Springer

Reflection and phys. sputtering of C on W

- □ Reflection coefficient is lower than that on W
 - R~0.6 (50eV C to W)
 - R~10-4(50 eV C to C)
- ☐ Carbon mono-layer is easily re-sputtered by reflected H from W substrate.
- Carbon deposition is more pronounced on graphite.



Difference in reflection

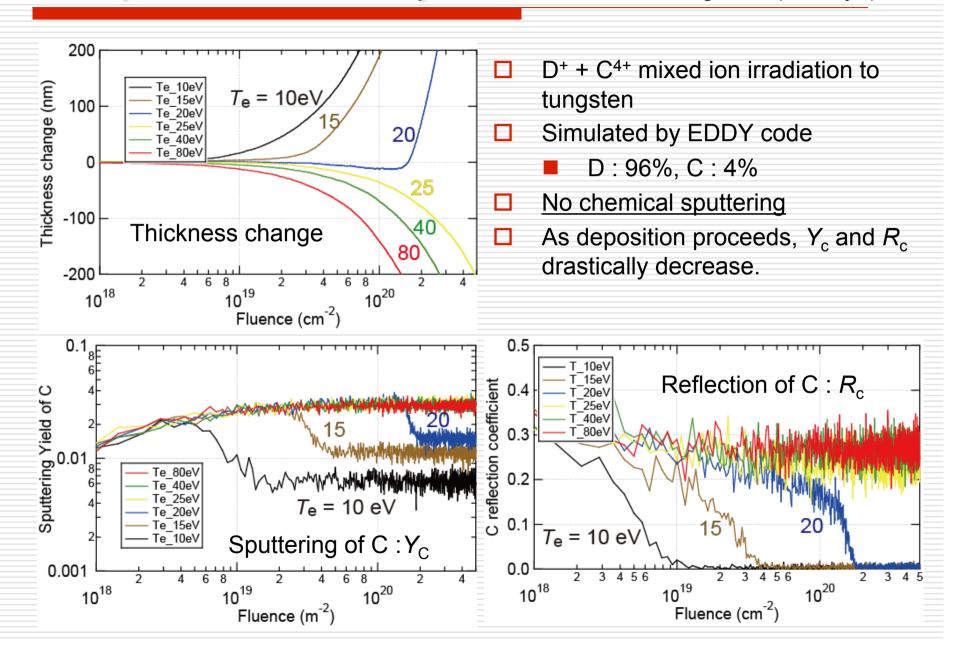


Enhancement of sputtering of surface C

A. Kreter, et al., Plasma Phys. Control. Fusion 48 (2006) 1401

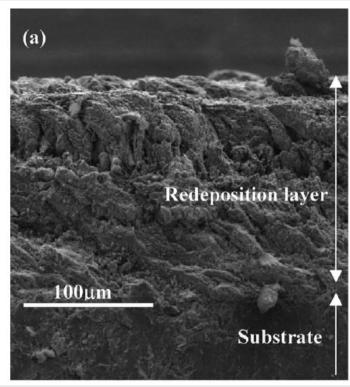
Computer simulation by EDDY

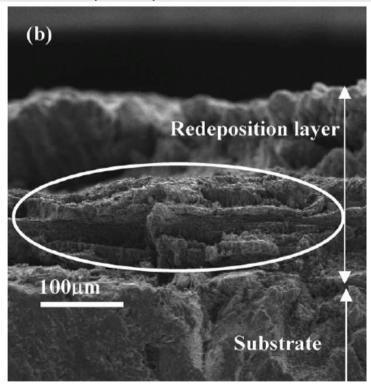
Acknowledgment (K. Ohya)



Deposition layer: quite different from solid C

- □ Different structure depending on Temperature, flux, D ratio, etc.
- ☐ C deposition layer is not dense (0.91 g/cm³ on JT-60U tiles (2.23 g/cm³ for graphite crystal)).
 - Y. Ishimoto et al., J. Nucl. Mater. 350 (2006) 301.





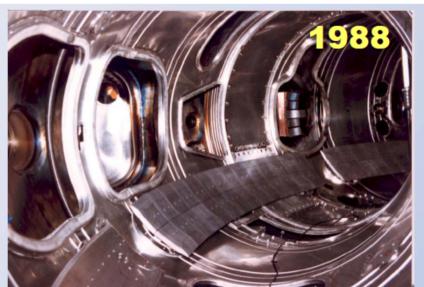
Structure of C deposition layer (JT-60U)

Some comments on erosion

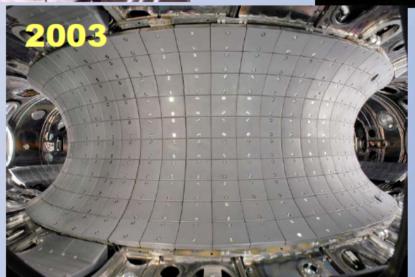
- □ Chemical erosion of C deposition layer
 - Depends on bulk properties (Soft C-H film (H/C~1), Hard C/H film (H/C~0.4))
 - W. Jacob, J. Nucl. Mater. 337-339 (2005) 839.
 - Local ¹³C deposition experiments and their simulations suggest enhanced re-erosion of C deposition layer.
 - ☐ A. Kirschner et al., J. Nucl. Mater. 328 (2004) 62.
- ☐ Chemical erosion of C in mixed layer with W
 - In general, C in mixed layer has lower chemical sputtering yields than graphite.
 - Temperature dependent C-selfsputtering was reported. But mechanism is not well known.
 - □ H. T. Lee, K. Krieger, J. Nucl. Mater. 390–391 (2009) 971.

Carbon deposition on W (TEXTOR test limiter experiments)

TEXTOR - a test bed for power exhaust concepts ...















High Z test limiter experiments

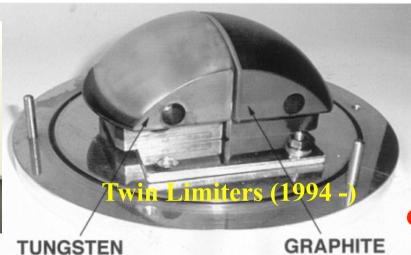






Main Poloidal limiters: VPS-W coated grahite (1998-1999)







Castellated W (1999-)

Subjects: Material test, erosion and transport, melt layer behavior, carbon redeposition

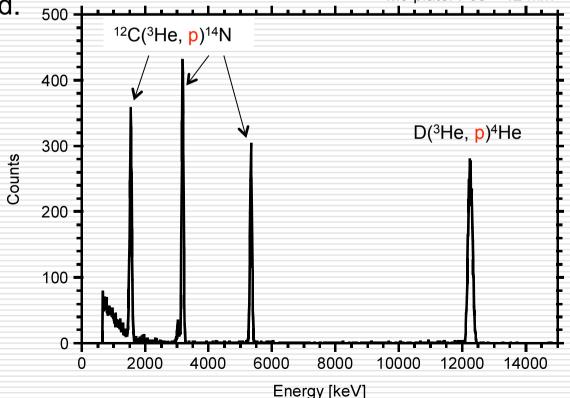
Experimental conditions for TEXTOR experiments

- Effects of surface roughness on C deposition
 - Tungsten
 - \square Roughness $R_a = 9 \sim 180 \text{ nm}$
 - Graphite (fine grained graphite)
 - \square Roughness $R_a = 70 \sim 700 \text{ nm}$
 - He plasma pre-exposed W
 - Nano-structure formed
- C deposition on tungsten at elevated temperatures
 - Temperature range
 - □ ~300 °C : ~ITER wall
 - □ ~550 °C : ~Chemical Sputtering peak
 - □ ~850 °C : Thermal diffusion + RES

NRA measurements (IPP Garching)

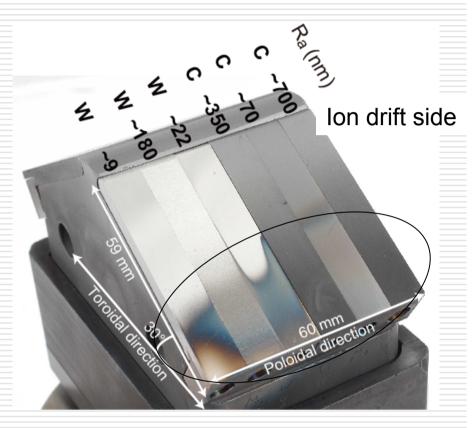
■NRA (Nuclear Reaction Analysis)

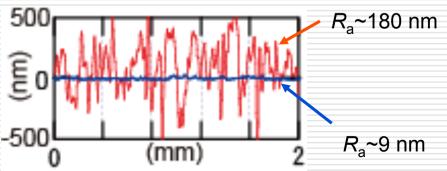
- Analysis beam: 2.5 MeV ³He⁺
- Protons produced by D(³He, p)⁴He & ¹²C(³He, p)¹⁴N nuclear reactions were detected.



Setup for study on surface roughness effects

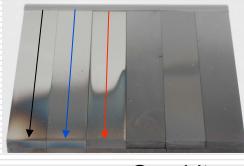
- □ Pure W samples
 - R_a ~9 nm, ~22 nm, ~180 nm
 - Difference in surface polishing
- ☐ Graphite (fine grained)
 - R_a ~70 nm, ~350 nm, ~700 nm
- Deposition mechanism
 - Lower T_e deeper into SOL
 - Higher carbon density deeper into SOL



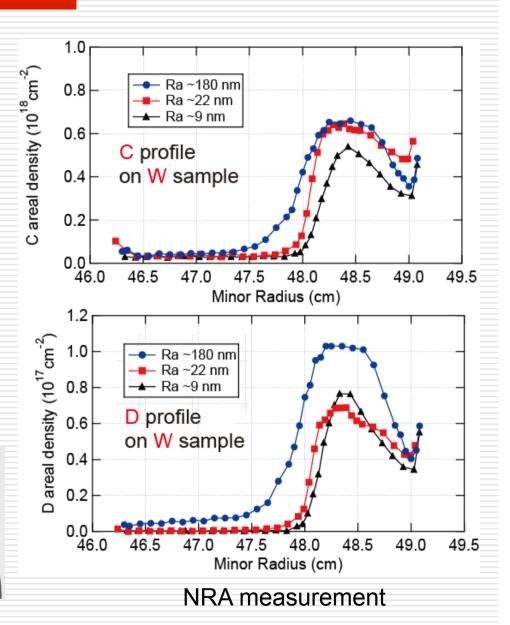


C deposition and D retention on W

- C deposition
 - Roughness enhances C deposition
 - $R_a\sim 180 \text{ nm} : \text{Long tail}$
 - Sharpe boundary between erosion and deposition
- D retention
 - similar to C deposition
 - no surface retention in erosion zone
 - D/C = 0.1~0.15



W Graphite



C/D ratio in C deposition layer

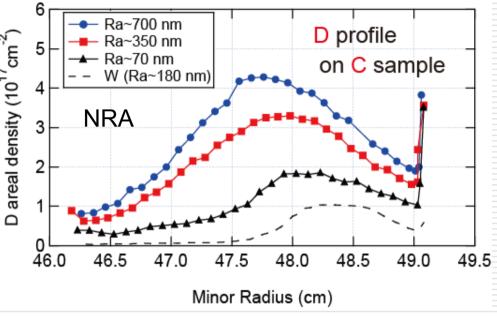
□ For the roughest case (R_a = 180 mm), the region of D/C ~ 0.1-0.15 extends to r ~ 46.5 cm, suggesting thin C deposition layer exists over wide area of the sample.

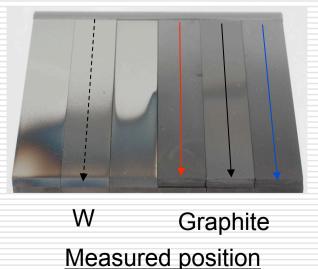


D/12C ratio

D retention (C deposition) on graphite

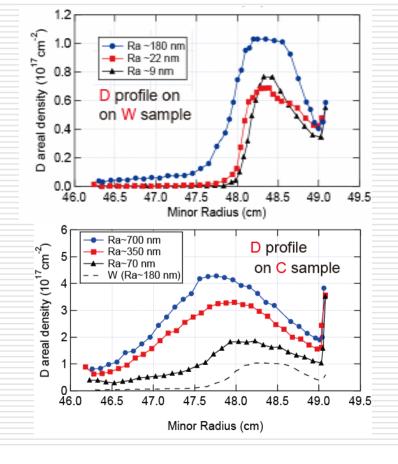
- C deposition on graphite
 - D retention was mainly in C deposition layer
 - D/C ~ const in deposition layer
 - □ D retention ~ C deposition
- Characteristics ofC deposition on graphite
 - Roughness enhanced C deposition also on graphite
 - No sharp transition between erosion and deposition

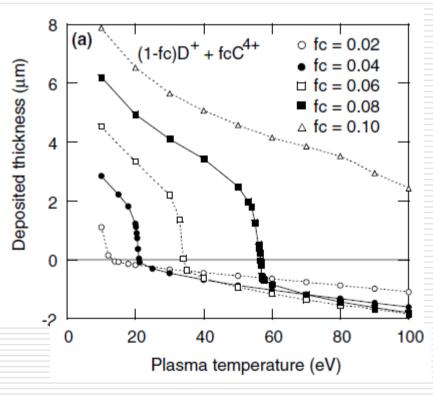




Sharp C deposition- erosion boundary on W

- C deposition rate is much higher on C than W.
- Once C deposition layer is formed, the deposition rate increases.
- Sharp boundary is formed.



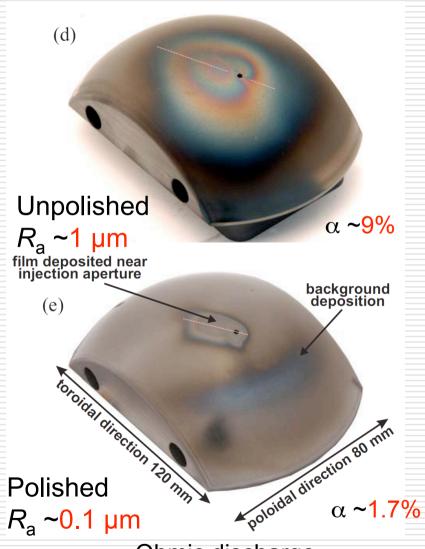


Simulation of C deposition

K. Ohya et al., J. Nucl. Mater. 337–339 (2005) 882

¹³CH₄ puff exp. with graphite limiter (TEXTOR)

- ☐ C deposition on graphite test limiter (TEXTOR exp.)
 - Deposition Efficiency α
 - Deposited ¹³C /injected ¹³CH₄
 - C on unpolished C (R_a ~ 1 µm)
 - **α ~9%**
 - C on polished C ($R_a \sim 0.1 \mu m$)
 - \square $\alpha \sim 1.7\%$
- ☐ Surface roughness seems to affect C deposition
 - Similar or larger than substrate effects (W or graphite)

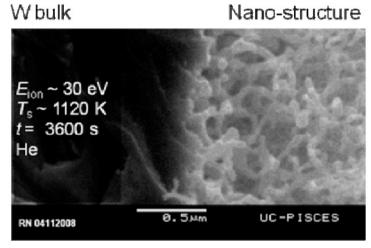


Ohmic discharge

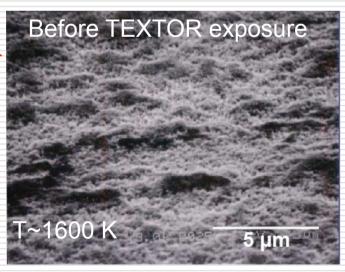
A. Kreter, et al., Plasma Phys. Control. Fusion (2008)

C deposition on He pre-exposed W

- ☐ He plasma pre-exposure
 - High density pure He plasma exposure in NAGDIS-II (Nagoya U.)
 - Black surface after ~1h exposure at 1300 °C (flux ~10²³ m⁻²s⁻¹)
 - ☐ He bubble and nanostructure formation
 - Surface structure removed before TEXTOR plasma exposure
 - □ Loosely bound nano-structure was wiped out mechanically
- □ Roughness of He exposed W
 - Roughness ~15 nm (after exp.)
 - ☐Small pits could be missing due to stylus type measurement



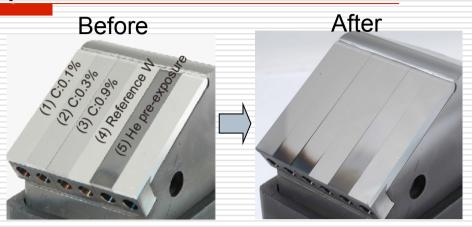
M. Baldwin et al., I-20, PSI18



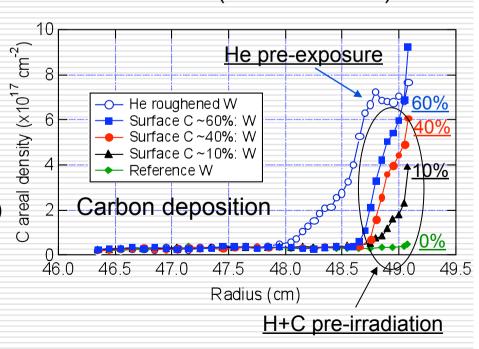
W surface in this work

C deposition on He pre-exposed W

- ☐ He pre-exposed W
 - Enhancement of C deposition
 - C profile : long tail
 - □ increase in deposition area
 - large enhancement of deposition despite small roughness (~15 nm)
- ☐ H+C pre-irradiated W
 - C deposition speed relates to surface C concentration
 - only 10% initial C affects deposition
 - □ No deposition on pure W (0%C)
 - $R_a \sim 10 \text{ nm}$ for each W

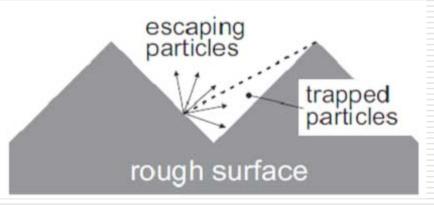


46 shots (Ohmic plasma) r = 46 cm (same as LCFS)



Explanation of roughness effect on deposition

- □ Roughness (0.01-1 µm) << Ion Lamor radius (0.1-1mm)
 - D ion flux and C ion flux did not change locally
 - Local shading effect of D ions may not occur
- Some of sputtered or reflected particles redeposited immediately.
 - Trapping rate depends on the morphology
 - He roughened surface was very fine and complicated structure
 - ☐ He induced roughness could have high trapping rate (C deposition)





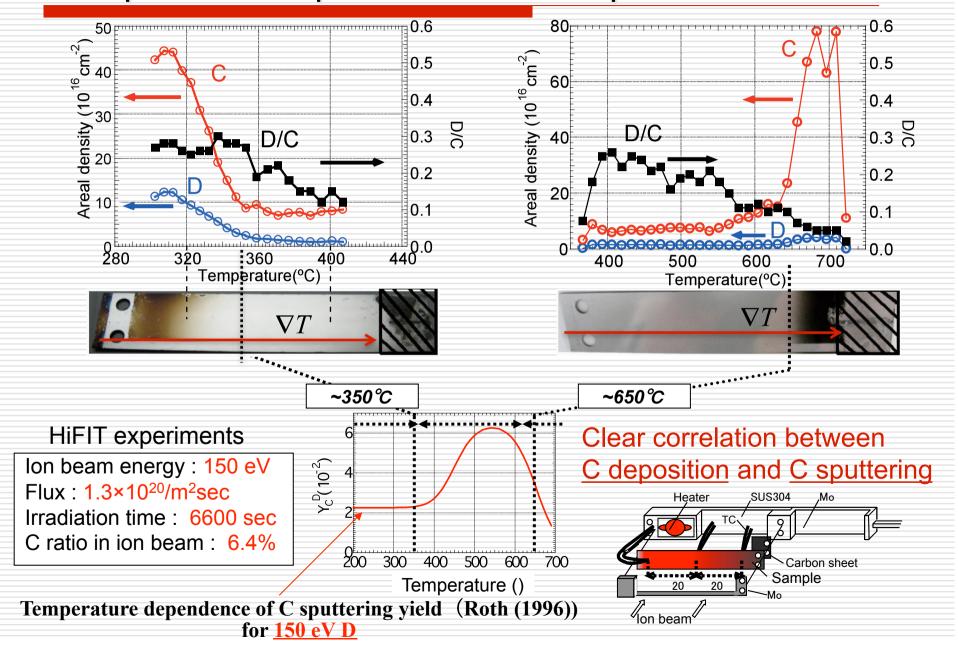
He roughened W surface

M. Kunster et al., Nucl. Instrum. Meth.B145 (1998)320.

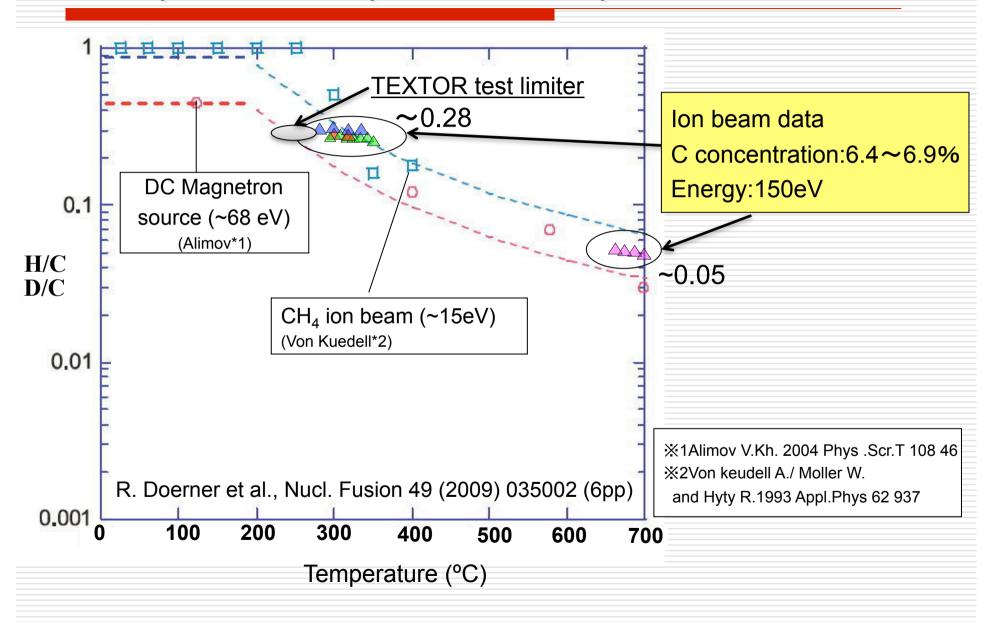
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 - \square Roughness $R_a = 70 \sim 700 \text{ nm}$
 - High density He plasma pre-exposed W
 - Nano-structure formed
- C deposition on tungsten at elevated temperature
 - Temperature range
 - □ ~300 °C : ~ITER wall
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Temperature dependence of C deposition

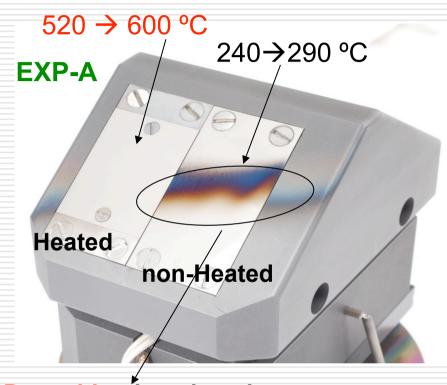


Comparison with previous C deposition data



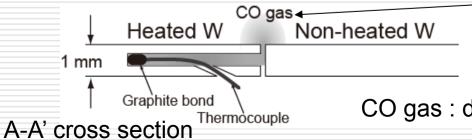
Partially heated limiter exp. for C deposition on W

770 → 930 °C



Deposition by edge plasma exposure

No deposition on the heated sample.



EXP-B

280 → 340 °C

EXP-B

A'

Heated

non-Heated

Deposition by edge plasma exposure

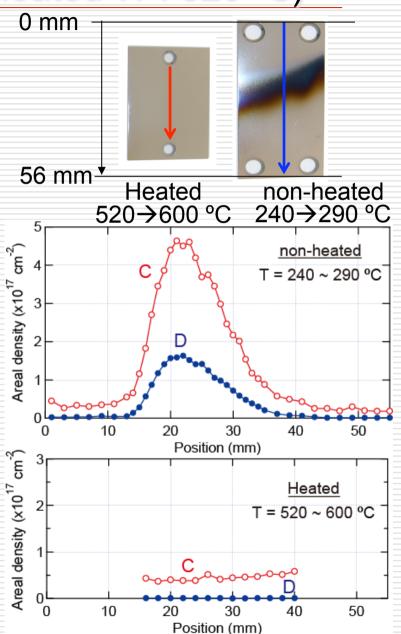
Deposition due to "gas puff" (CO)

No deposition on the heated sample.

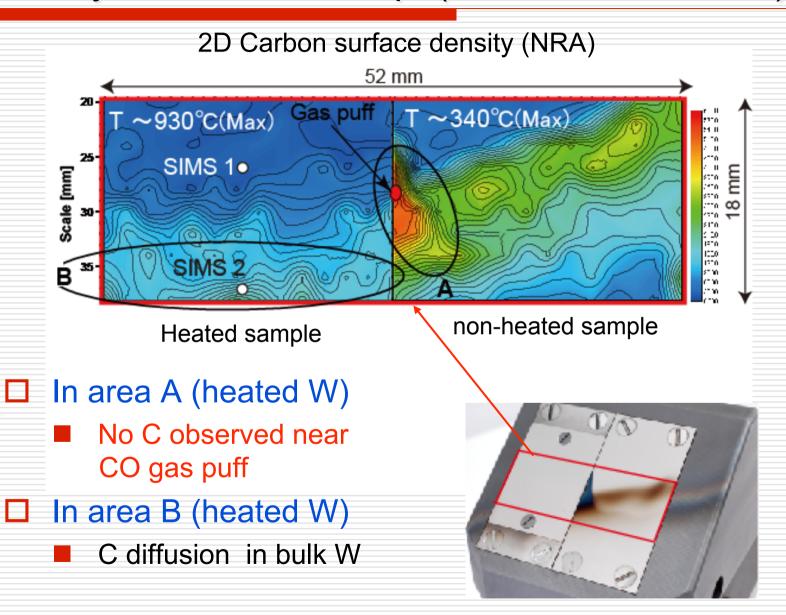
CO gas: desorbed above ~700 °C

Partially heated limiter exp. (heated W: 520 °C)

- □ non-heated W (240 °C~280 °C)
 - Beltlike C deposition (asymmetry)
 - D retention only on C deposition
 - D/C ratio ~ 0.3
- ☐ Heated W (520 °C~600 °C)
 - no C deposition
 - no near surface D retention
 - near peak T of chemical sputtering

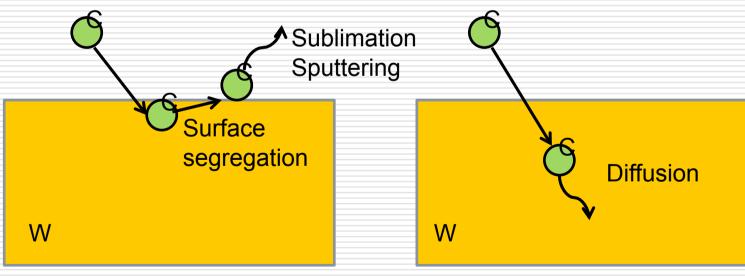


Partially heated limiter exp. (heated W: ~930 °C)



Possible reason for C behavior on high T tungsten

- □ Difference in Ion energy could be the reason
 - C in plasma : highly charged (~ +4), thermalized □ impact energy E ~ 580 eV (T_e~T_i~40 eV)
 - C+ or CO+ from CO gas : singly charged, not thermalized
 - \square impact energy E ~120 eV (T_e ~40 eV, T_i ~0 eV)
 - □ Ion range ~ less than a few ML
 - □ Implantation → Surface segregation → sputtering, sublimation

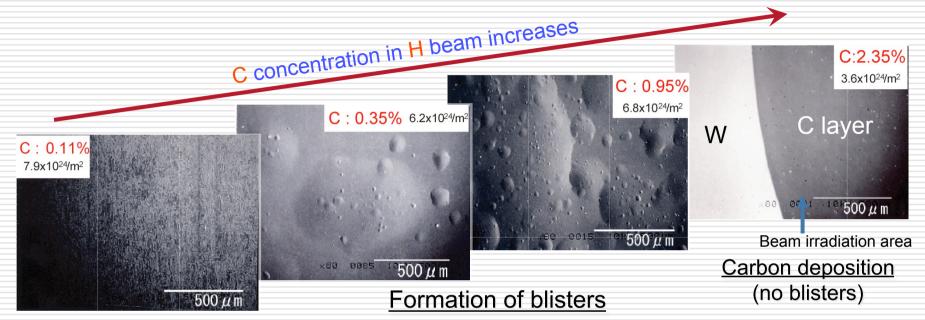


Shallow implantation

Deep implantation

Retention/blistering by simultaneous C/ He/D exposure

Enhancement of blistering by carbon impurity



No blisters

Small amount of carbon (less than 1%) in ion beam can enhance blister formation on W.

Experimental conditions

Beam Energy: 1keV H₃+, Flux: (3-4)x10²⁰ Hm⁻²s⁻¹

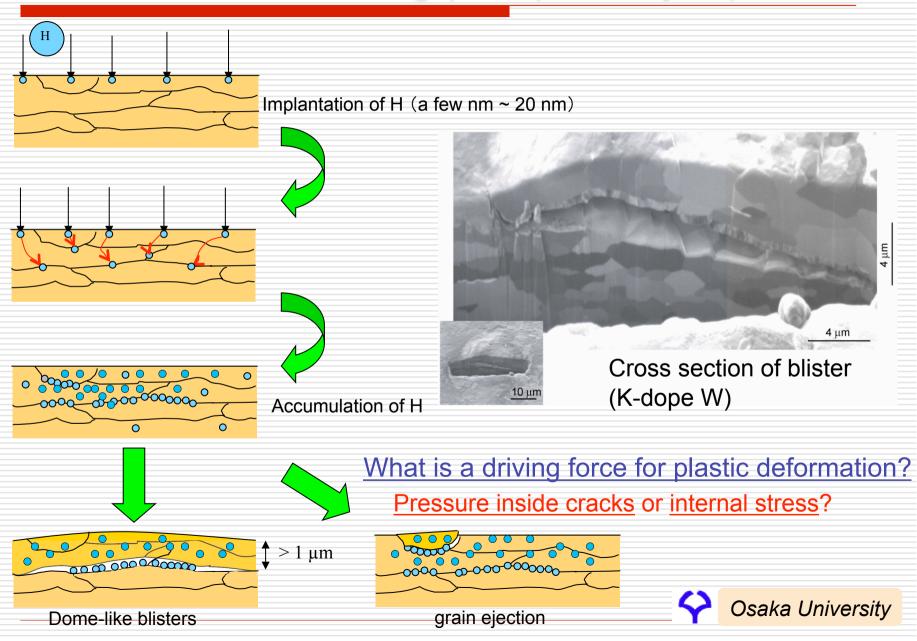
Temperature: 653 K

Sample: pure W with mirror polished

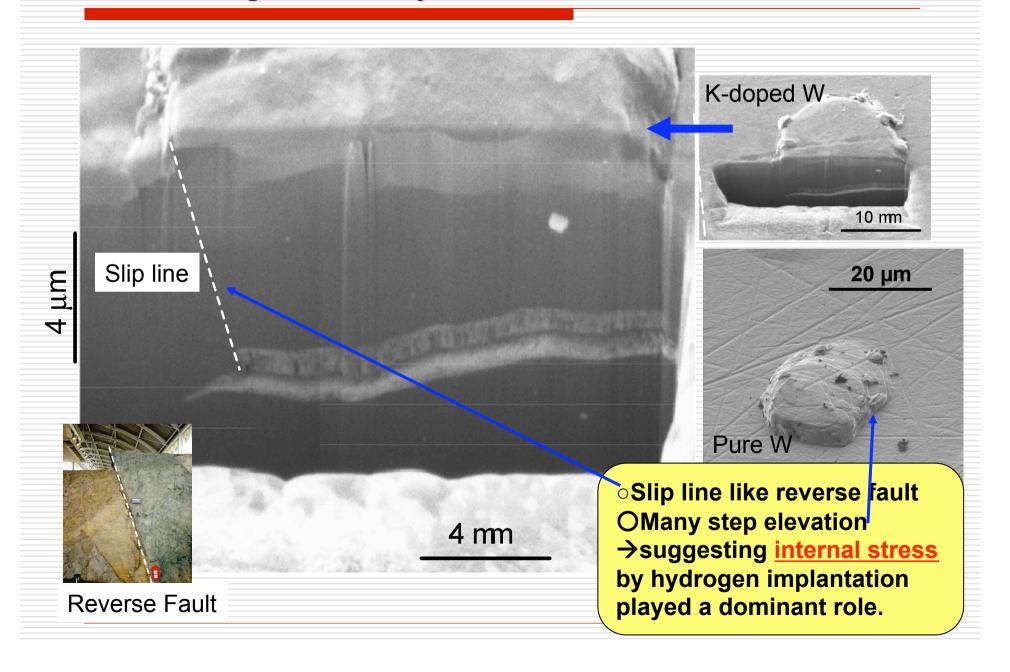


Osaka University

Mechanism for blistering (K-doped Poly-W)

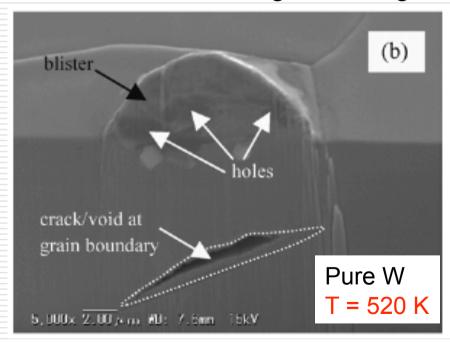


Blistering of Recrystallized W



Blisters without gaps

- Blisters without gaps (or small gaps) are reported lately.
- Formation mechanism is not known.
 - Abnormal diffusion of W?
 - Giant swelling due to high flux D/H irradiation?

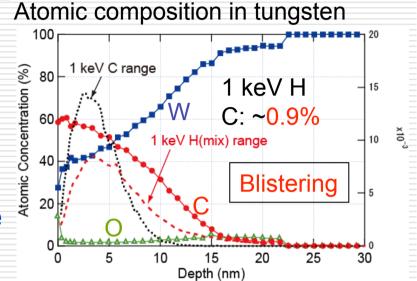


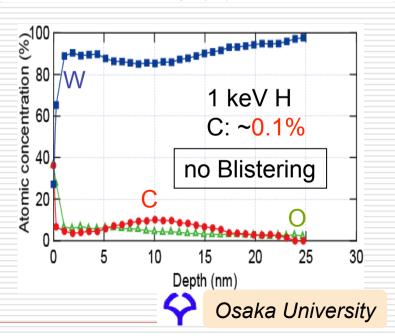




W and C mixing layer reduced desorption

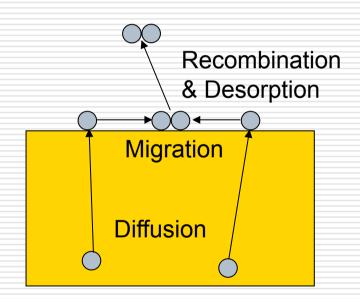
- C depth distribution
 - Absolutely calibrated by NRA
 - broader than ion implantation range
 - Recoil implantation by H
- ☐ High C (~0.9% in the beam) case
 - WC layer reduced desorption of H
 - Enhance bulk diffusion of H
 - Enhance blister formation
- □ Low C (~0.1% in the beam) case
 - Low surface C concentration
 - no significant reduction of recombination



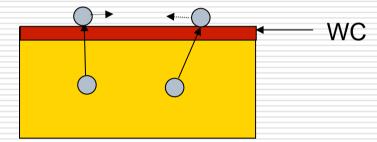


How does WC layer affect H behavior?

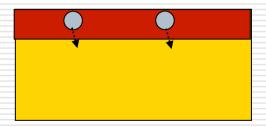
- □ Low H-recombination rate on WC surface
 - Suppress surface migration of H atoms
- Low H-diffusion coefficient in WC



Hydrogen Desorption (<1000 °C)



Surface WC-rich layer suppresses hydrogen recombination and desorption



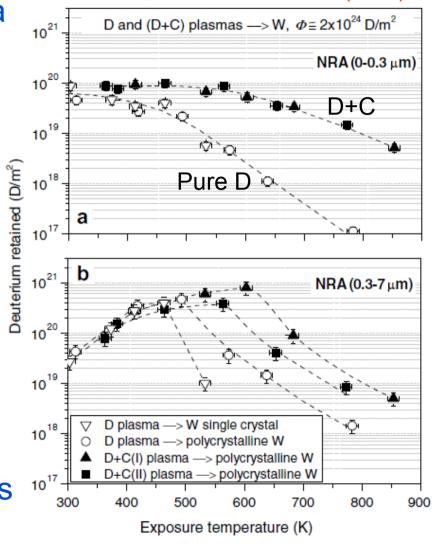
Bulk WC layer reduces hydrogen diffusion

D & C mixed plasma exposure to W

Planar DC magnetron plasma

- Energy : ~200 eV (D₂+ mainly)
- Flux: 1 x 10²¹ m⁻²s⁻¹
- C plate on cathode surface to provides C into plasma
- For D+C, D retention near surface (a) and bulk (b) increased at elevated temp.
- ☐ For D+C, fraction of C on W surface is higher.
- □ Possibly, surface C+W mixed layer (C existed as carbidic and graphitic phases) reduces release of D from surface.

V. Alimov et al., J. Nucl. Mater. 375 (2008) 192.



D retention in W exposed to pure D plasma ($\square \nabla$) and D+C plasma ($\blacktriangle \blacksquare$)

From 300-700 K, thin and thick layers of Be suppresses blister formation.

M. Baldwin et al. PSI 18(2008)

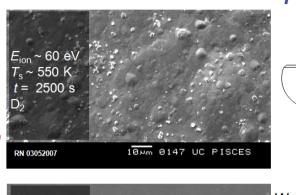
PISCES

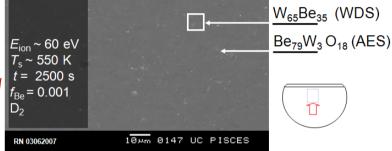
Blistering & exfoliation of blister caps is a concern for certain varieties of W.

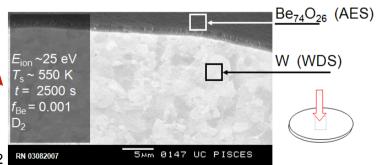
Increased retention is associated with the trapping of hydrogen in blisters. E.g. K Tokunaga et al. J. Nucl. Mater. (2004) 337-339, 887.

 At 550 K a blistered surface is prevalent after exposure to D₂ plasma.

 A thin layer of Be as little as a few 10's of nm, or thicker, is found to suppress blister formation.







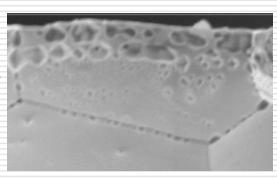
D⁺ ion fluence $\sim 1 \times 10^{26}$ m⁻²

PISCES



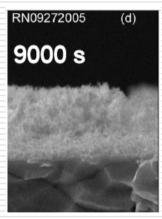
He effects on W

- ☐ High temperature (> ~1,600 K)
 - Large He bubbles formation with recrystallization
 - Degradation of mechanical and thermal properties
 - Dust formation (enhanced erosion)
- ☐ Medium temperature (> ~1,100 K)
 - Nano-structure formation
 - Dust formation (enhanced erosion)
 - Initiation of arcing
- □ Low temperature (< ~900 K)</p>
 - Small He bubble formation (a few nm)
 - Significantly affects D/T retention and diffusion



NAGDIS (Nagoya Univ.)

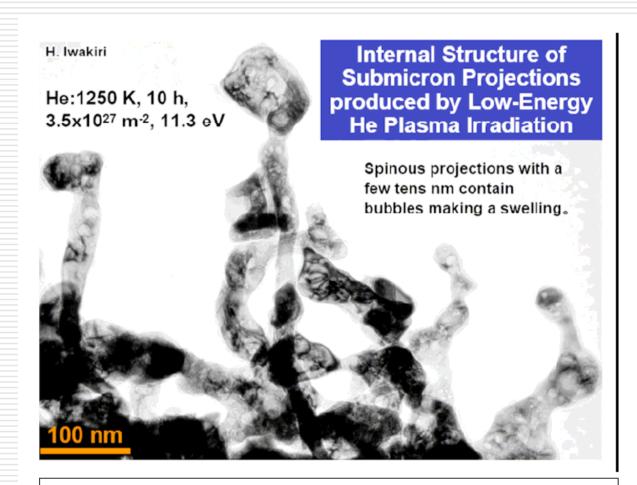
T ~ 1,600 K



PISCES (UCSD)

T ~ 1,120 K

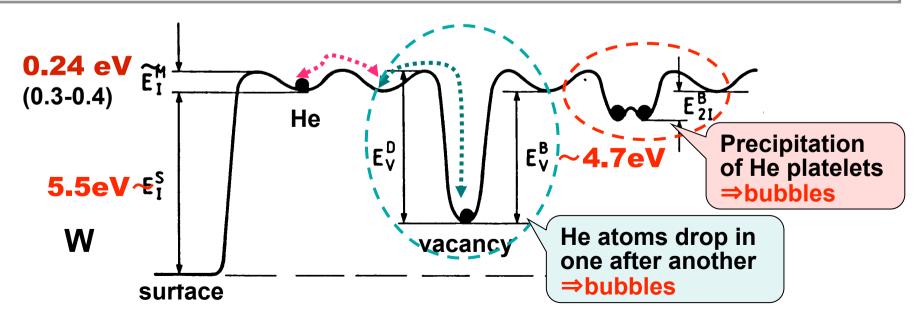
Submicron structure on W (T~1250 K)



From Prof. Takamura presentation at ITPA sol./div meeting, Toronto, Nov. 2006.

Y. Yoshida (Kyushu U) 18th PSI (2008)

- Very low solubility.
- Very fast thermal migration via interstitial sites (very high mobility even at R. Temp.)
- Very deep trapping in a vacancy (Large E_V^B)
- Comfortable positions of He in W lattice: empty sites such as vacancy, bubble, grain boundaries, dislocations etc. ≡ closed electron shell structure
- He enhances the formation of voids (bubbles) and dislocation loops even above 1000°C → hardening, embitterment
- He atoms can aggregate by themselves → He atoms can form clusters once get in the lattice (E>E^S_I) → no need displacement damage

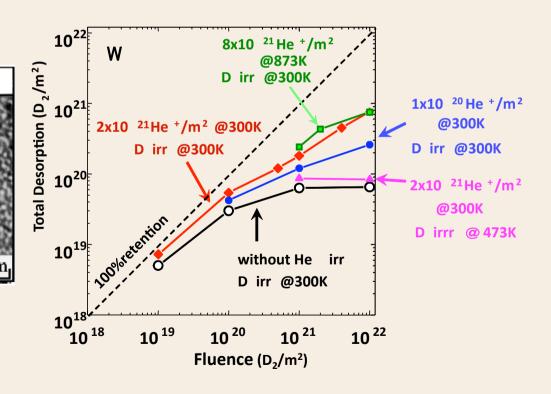


He effect on retention

- Sequential irradiation of He and D.
- Formation of He bubbles enhances D retention very much.
- He bubbles become traps of D.
 - (H. Iwakiri et al., J. Nucl. Mater. 307-311 (2002) 135-138)

1.0x10²⁰He⁺/m² 2.0x10²¹He⁺/m²

8keV He $^+\rightarrow$ W (300K)



Blister formation under H & He (&C) irradiation

753 K

653 K

473 K

- Small amount of He affected blistering
 - He: ~0.1% has strong effects
- □ Suppression of blisters at T>653 K
 - 0.1% He did not change surface mixing layer much.

He: ~0.1% 1 keV H. C:~0.85% Carbon (as WC) Depth (nm) He: 0% Atomic Concentration [%] Tungsten 1 keV H. C:~0.85% Carbon (total) Carbon (as WC) 25 Depth [nm]

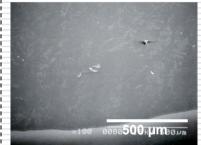
Energy : **1 keV** (**H**₃⁺, **H**₂⁺, **H**⁺)

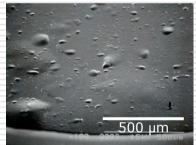
Carbon : ~0.8%

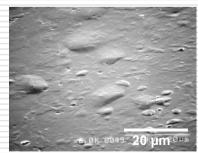
Fluence : ~7.5 x 10²⁴ m⁻²

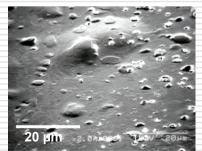












He: 0.1%

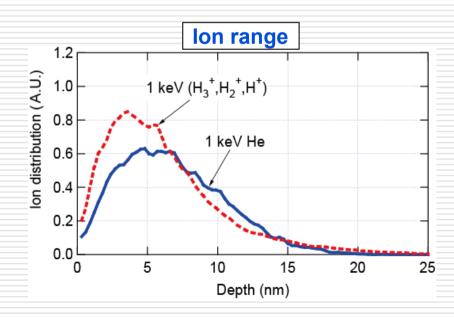
He: 0%

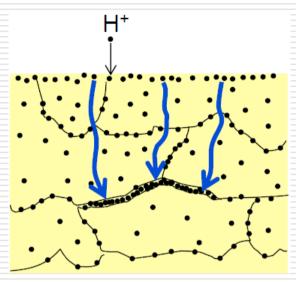


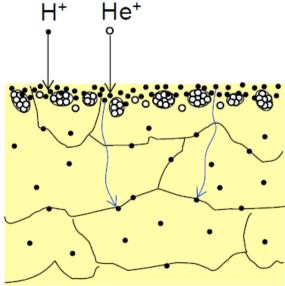
Osaka University

He bubble could affect H diffusion

- □ 1 keV He has slightly longer range than 1 keV H (mixed).
 - He bubbles could be formed around the end of ion ranges.
 - He bubbles in W and C mixed layer.
- ☐ He bubbles could block H diffusion into the bulk.









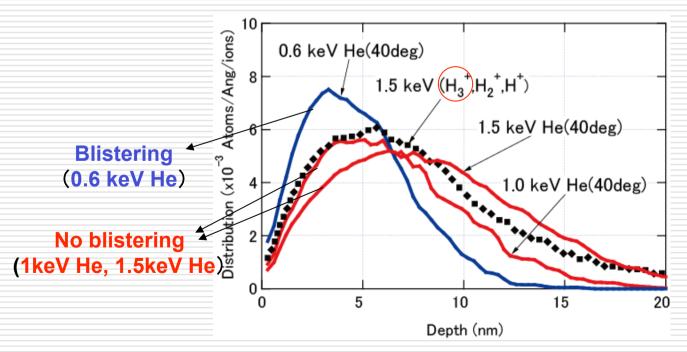
Effects of He energy on blistering

Main Ion Beam(1.5 keV: H+C:0.8%)

- (a) no He ion beam → Blistering
- (b) 2^{nd} He beam :0.05%(0.6 keV) \rightarrow Blistering
- (c) 2nd He beam :0.05%(1.0 keV)* >no Blistering

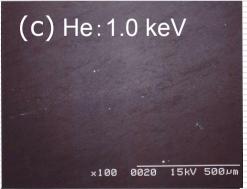
2nd He beam :0.05%(1.5 keV) *

*angle of incidence ~ 40 deg







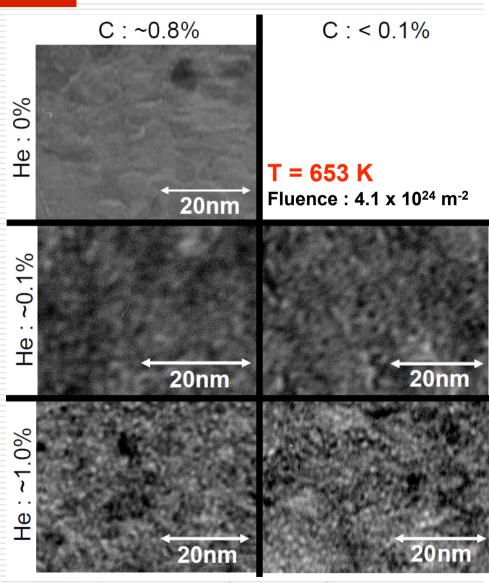




Ion range in tungsten

TEM observation of He bubbles

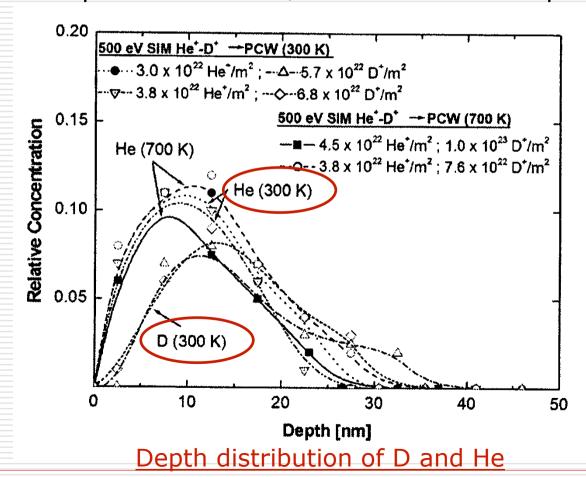
- ☐ He:1.0%, ~2 nm⁰ He bubbles
- ☐ He:0.1%, 1~2 nm[†] He bubbles
 - He fluence : 4.1 x 10²¹m⁻².
- □ Bubble size and bubble number density had weak dependence on He % and C%.
- ☐ He bubbles were formed in WC layer for C:~0.8%.



TEM observation of near surface structure

Simultaneous He/D (Toronto)

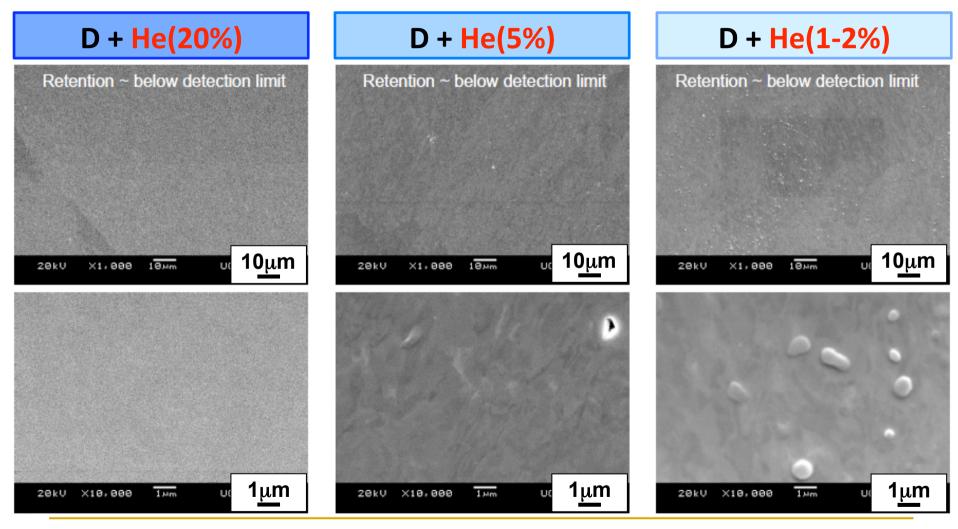
- □ 500 eV D & 500 eV He
- H. Lee, J. Nucl. Mater. 363-365 (2007) 898
- At 300 K, D did not diffuse into the bulk.
 - For pure D irradiation, D diffused much deeper at 300 K.



Suppression of Blistering by He

PISCES (UCSD) $E_i \sim \frac{55\pm15 \text{ eV}}{\Gamma_i} \sim 10^{22} \text{ ions/m}^2$

■W (SR), 5x10²⁵ D/m², 573K



M. Miyamoto et al., Nucl. Fusion 49 (2009) 065035 (7pp).

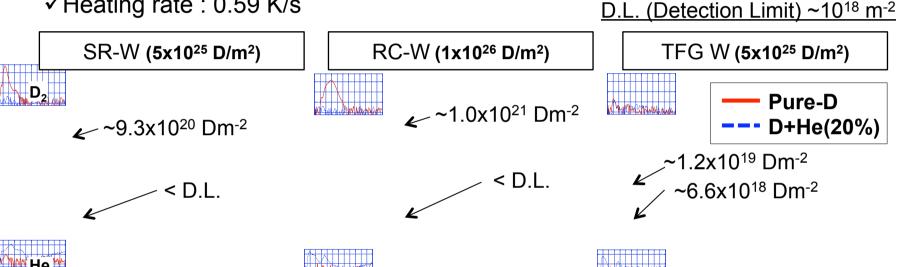
Blisters disappeared above 5% He.

D retention

Partial pressure [torr]

■TDS after plasma exposure

- √ High resolution Q-Mass (D₂ and He can be separated).
- √ Heating rate: 0.59 K/s

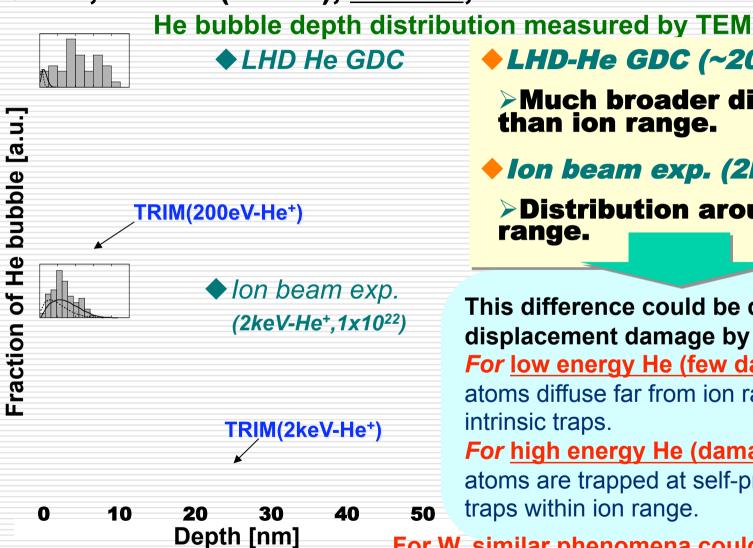


No D retention from W exposed to D+He mixed plasma

Similar results have been obtained in other research group (V. Alimov et al., 12th ITPA (SOL/DIV) meeting (2009)

Depth distribution of He bubbles in SUS.

♦ LHD, He GDC(200eV), <u>SUS304</u>, 65 hours



- **♦ LHD-He GDC (~200 eV)**
 - Much broader distribution than ion range.
- ♦ Ion beam exp. (2keV-He⁺)
 - Distribution around ion range.

This difference could be due to displacement damage by He ions.

For low energy He (few damage): He atoms diffuse far from ion range to find intrinsic traps.

For high energy He (damage): He atoms are trapped at self-produced traps within ion range.

For W, similar phenomena could take place.

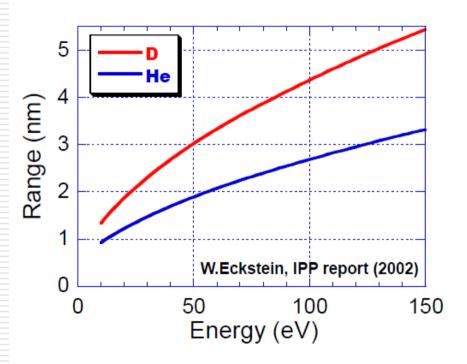
Needs experimental results

M. Miyamoto et al., J.N.M. 329-333 (2004) 742

Is He bubble layer a diffusion barrier?

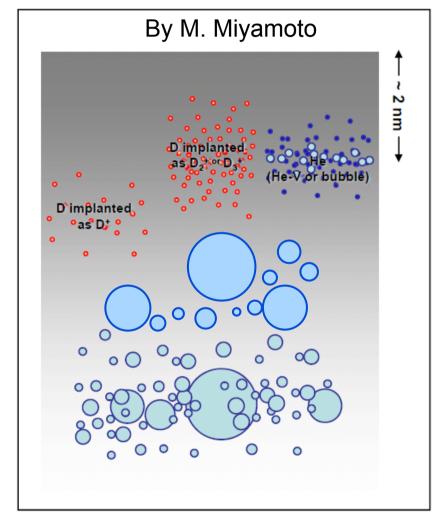
□ For low energy He implantation (less than recoil threshold), He bubble layer could extend deeper and

become the barrier.



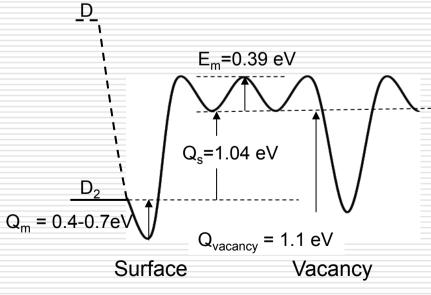
♦lon range (@70eV)

lon	He⁺	D⁺	D ₂ +	D ₃ +
Range	2.2nm	3.6nm	2.5nm	2.2nm

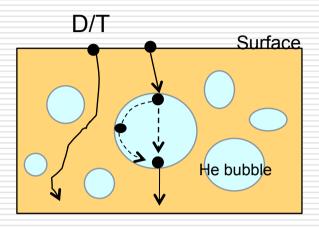


Possible mechanisms

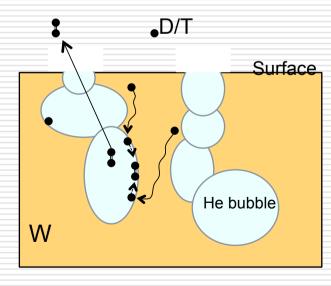
- □ Narrowing diffusion channels by He bubbles
- □ Release of D² through He created pores
- Reduction of diffusion through stress-induced W



Energy diagram of D in W



Diffusion of D/T in W



<u>Desorption of D₂ through pores</u>

Summary

- There still remain unsolved problems in C & W mixing.
 - C erosion from C&W mixed layer and C deposition layer
 - Effects of surface morphology (roughness) on C deposition
 - C atom behavior at elevated temperatures (> 800 °C)
- C & W mixed layer strongly affects D behavior
 - W&C mixed surface layer reduces recombination of D atoms
 - Diffusion of D in W&C mixed layer is reduced compared to pure W
- ☐ He bubble layer strongly affects D retention
 - Initially increase retention by increasing trapping sites
 - But, under high fluence condition, He bubbles greatly reduce retention as they work as diffusion barrier.
- We do not have enough knowledge on material mixing to correctly evaluate T retention in ITER.