Deuterium permeation in tungsten by mixed ion irradiation

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10th International Symposium on Fusion Nuclear Technology (ISFNT)
September 11th - 16th, 2011 in Portland, USA

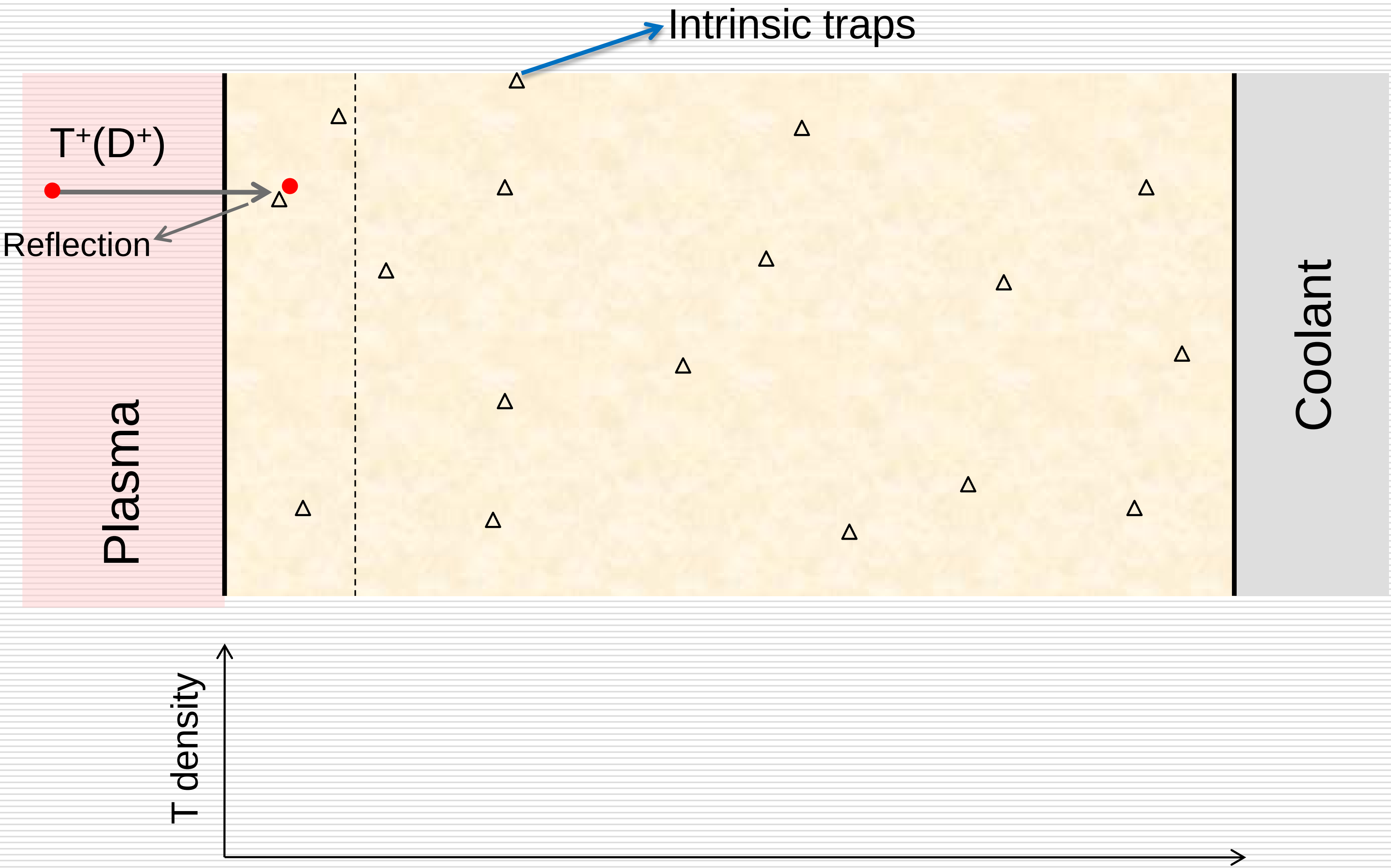
Outline

- Background and Purpose
  - General explanation of tritium behavior in wall materials
- Experimental details
  - Permeation apparatus
  - W specimens
- Pure D ion driven permeation
  - Microstructure (annealing temperature) dependence
- He/D mixed ion driven permeation
- C/D mixed ion driven permeation
- Discussion and Summary

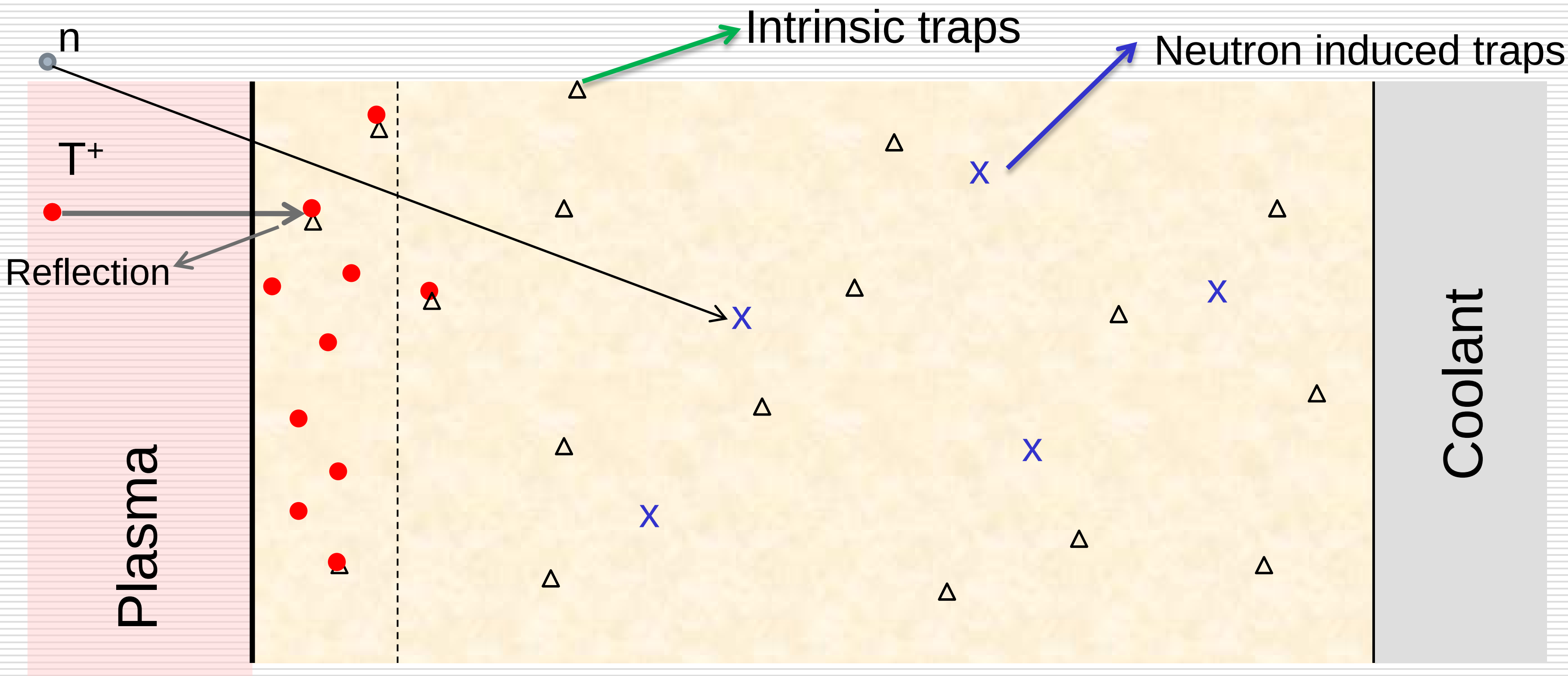
Introduction

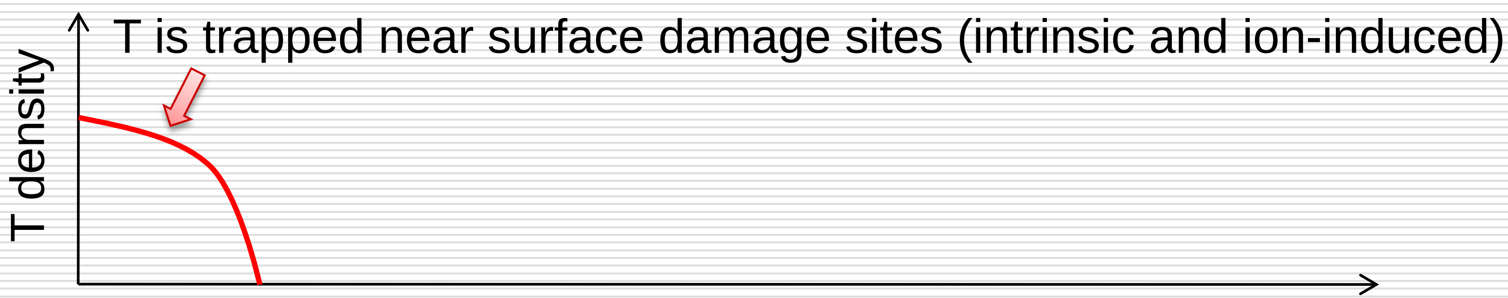
- T retention and permeation to coolant in blankets need to be studied for evaluation of T economy and safety.
- Tungsten is a leading candidate of armor materials of blankets.
- Tungsten has low diffusivity and solubility of T, and relatively high trapping energies of defects.
- Therefore, tungsten armor on reduced activation materials of blankets (Ferritic steel, V alloy) can be permeation reducer to coolant.
- However, still basic parameters to determine permeation such as <u>effective diffusivity</u>, <u>recombination coefficient</u> etc. are not well determined.
- In addition, <u>effects of mixed ion irradiation (D+He,C)</u> are not known well either.

Tritium behavior in a first wall

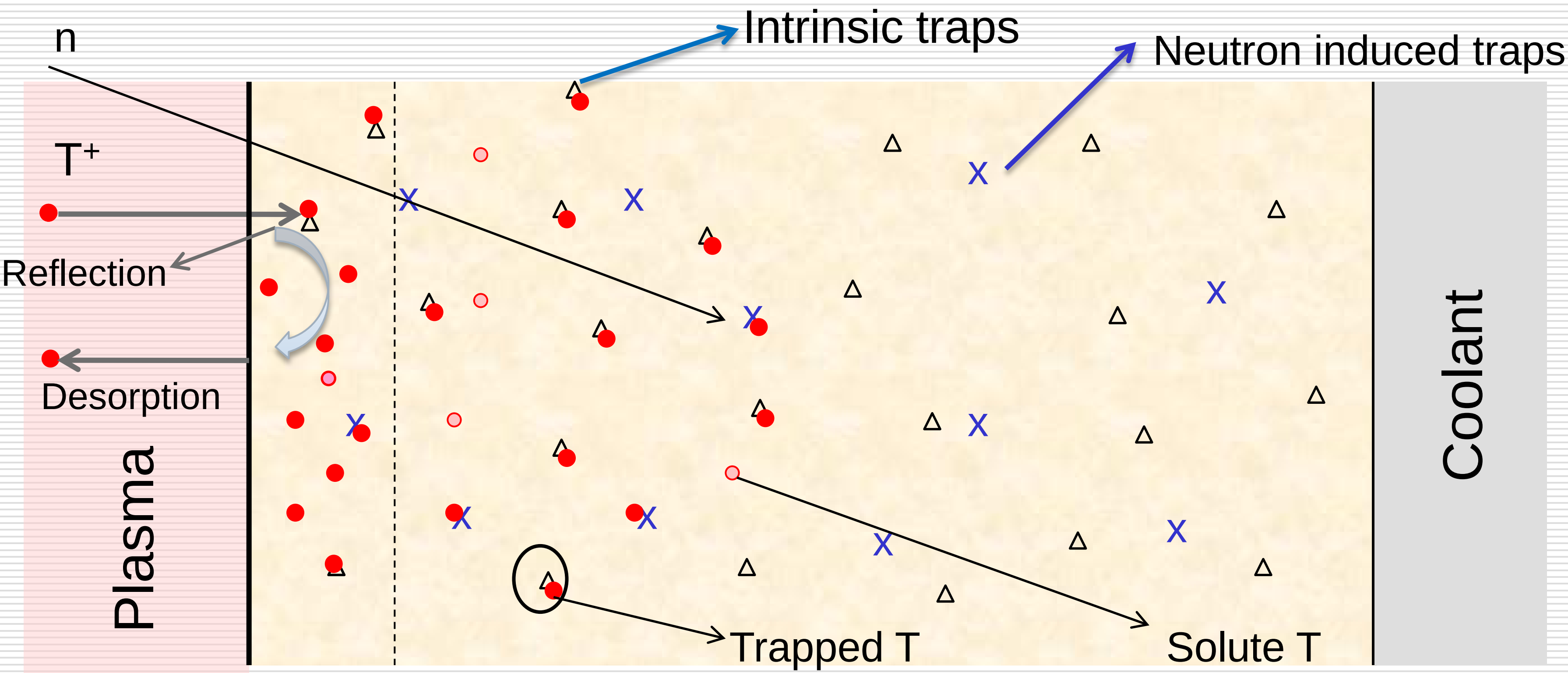


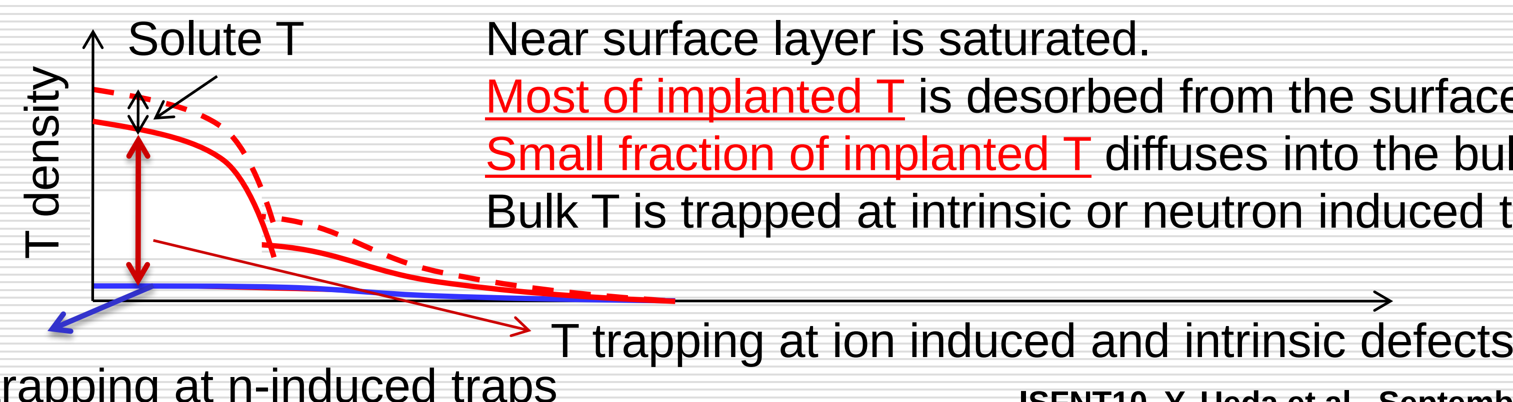
Tritium behavior in a first wall





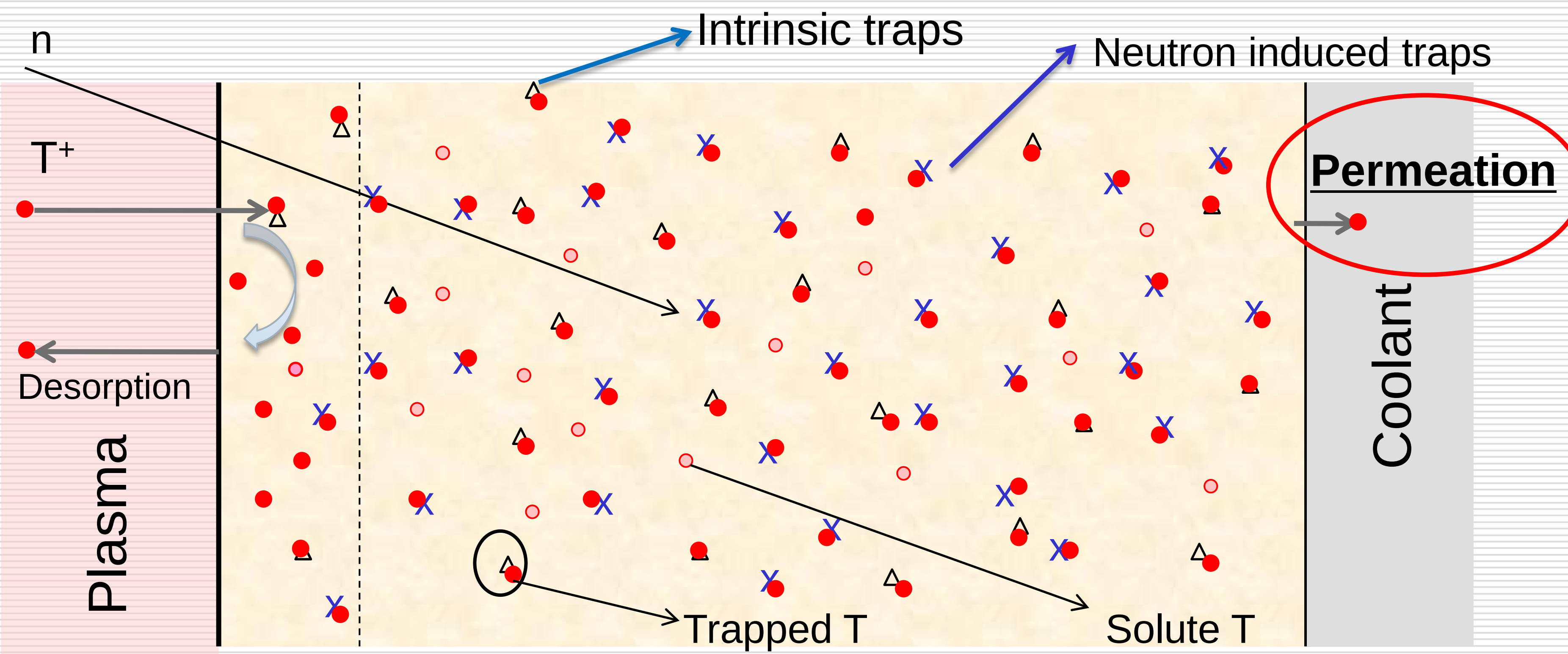
Tritium behavior in a first wall

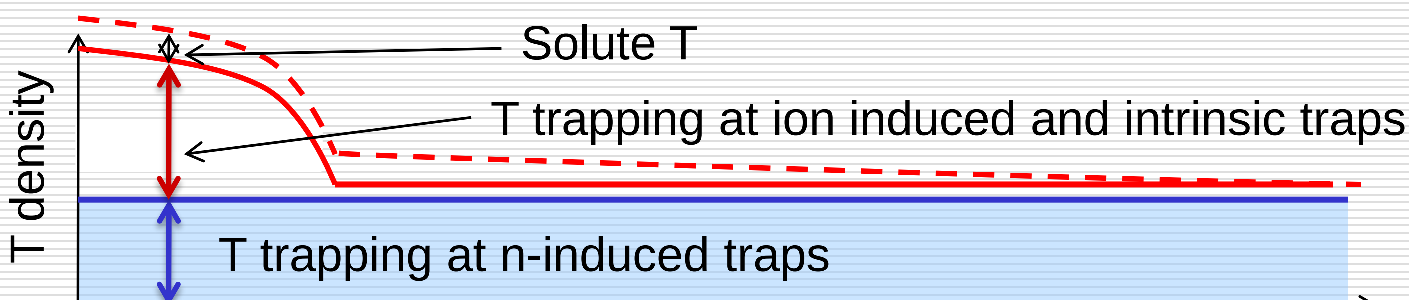




Near surface layer is saturated.
Most of implanted T is desorbed from the surface.
Small fraction of implanted T diffuses into the bulk.
Bulk T is trapped at intrinsic or neutron induced traps.

Tritium behavior in a first wall





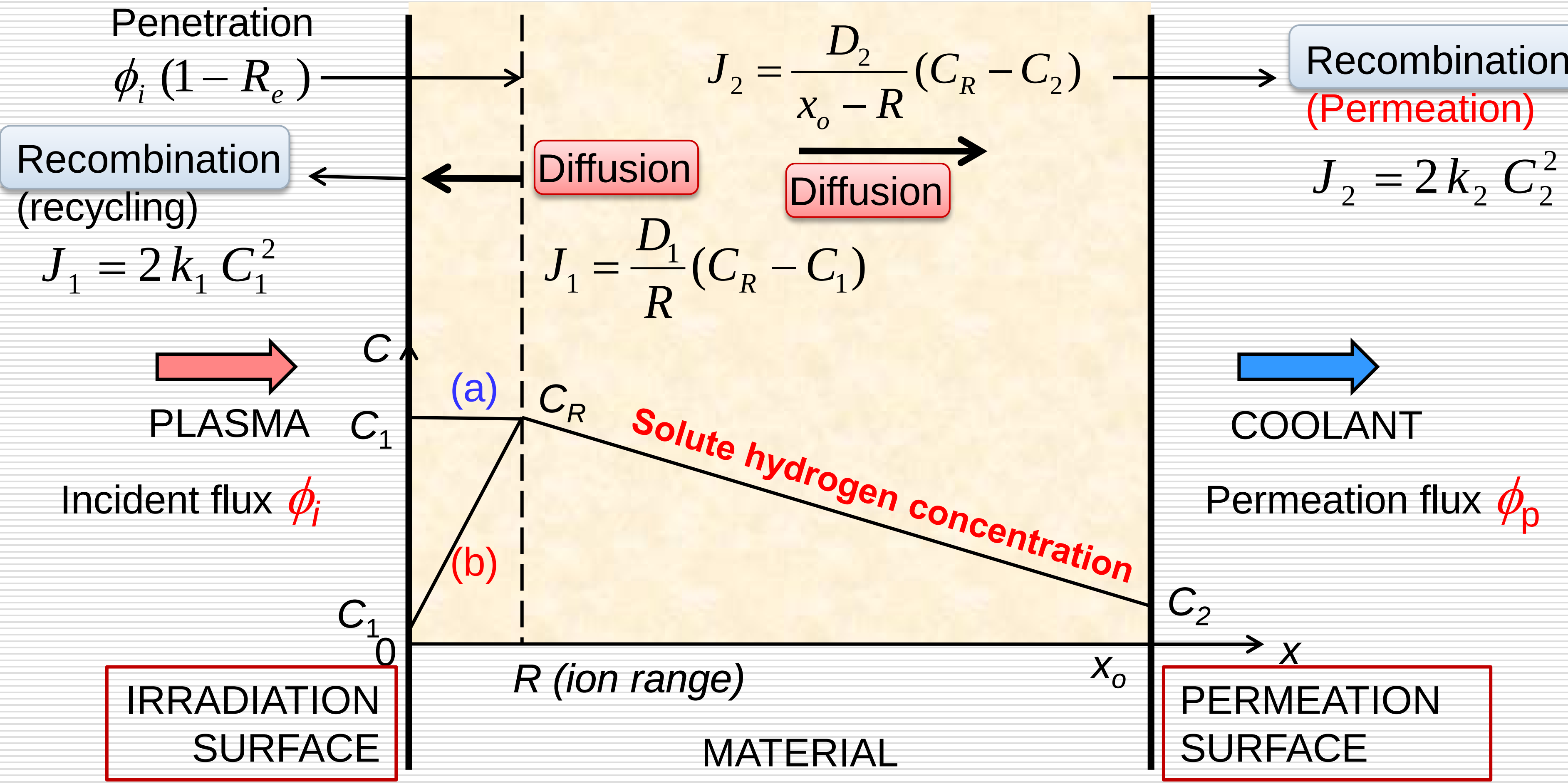
As temperature increased, trapped T decrease,
but still traps could affect effective diffusivity

ISFNT10, Y. Ueda et al., September, 2011

Necessary information for T behavior

- Surface damage effects (How they affect T retention and release)
  - Recoil damage effect
  - Effects of bubbles and blisters (due to oversaturation etc.)
  - Impurity mixing effects (He, wall impurities (C, Be etc.)
- Effective diffusion mechanism (How they affect T transport)
  - Trap sites effect
  - Microstructure effect (intergranular diffusion, grain boundary diffusion)
- Neutron induced traps (How they affect T retention)
  - Trapping energies
  - Production rate (as a function of dpa)
  - Saturation level with respect to dpa

Ion driven permeation model (Brice & Doyle)



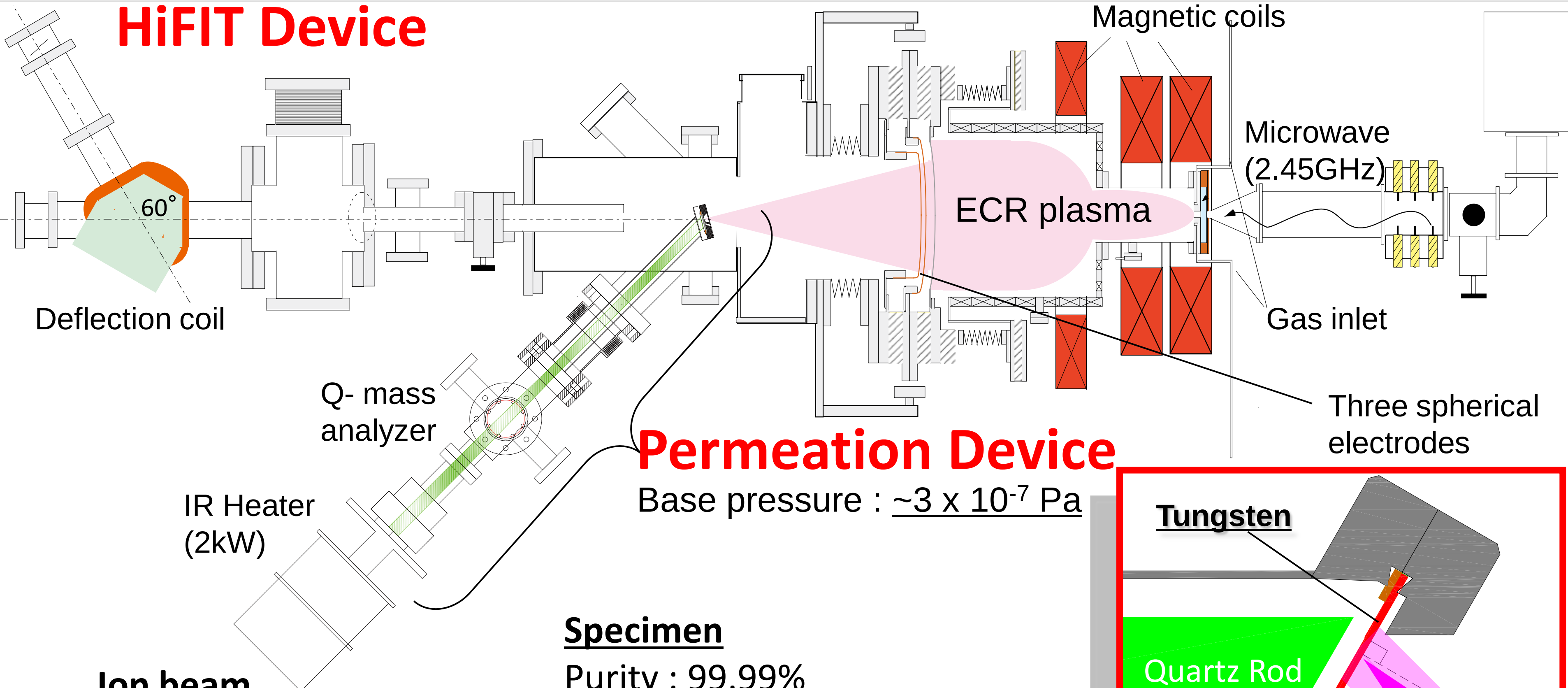
(a) Recombination limited condition : $\phi_p = \frac{D_2}{x_0\sqrt{k_1}}\sqrt{\alpha\phi_i}$

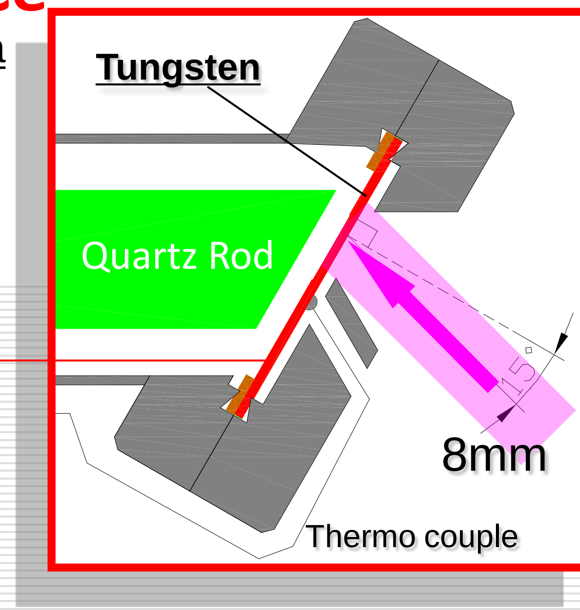
(b) Diffusion limited condition : $\phi_p = \frac{RD_2}{x_0 D_1}\alpha\phi_i$

Brice D.K. and Doyle B.L., J. Nucl. Mater. 120 (1984) 230-244.

Experimental setup

Experimental device





Ion beam

Energy : 0.15 keV-3 keV

flux(D) : ~10^{20} $m^{-2}s^{-1}$

Species : D_3^+, D_2^+, D^+

Impurity(C) : 0.1~0.2%

Specimen

Purity : 99.99%

Annealed: 1573 K for 1h

Thickness: 75 µm, 30 µm

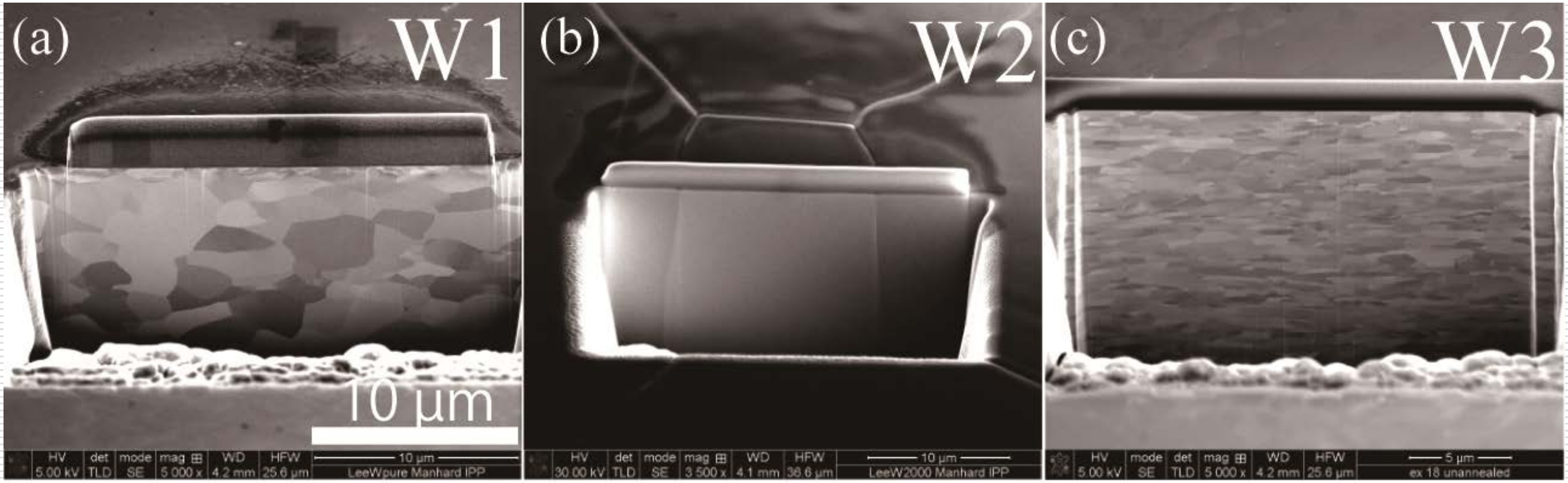
Roughness: R_a=0.01µm

Temperature : ≤ 1050 K

Blanket first wall conditions

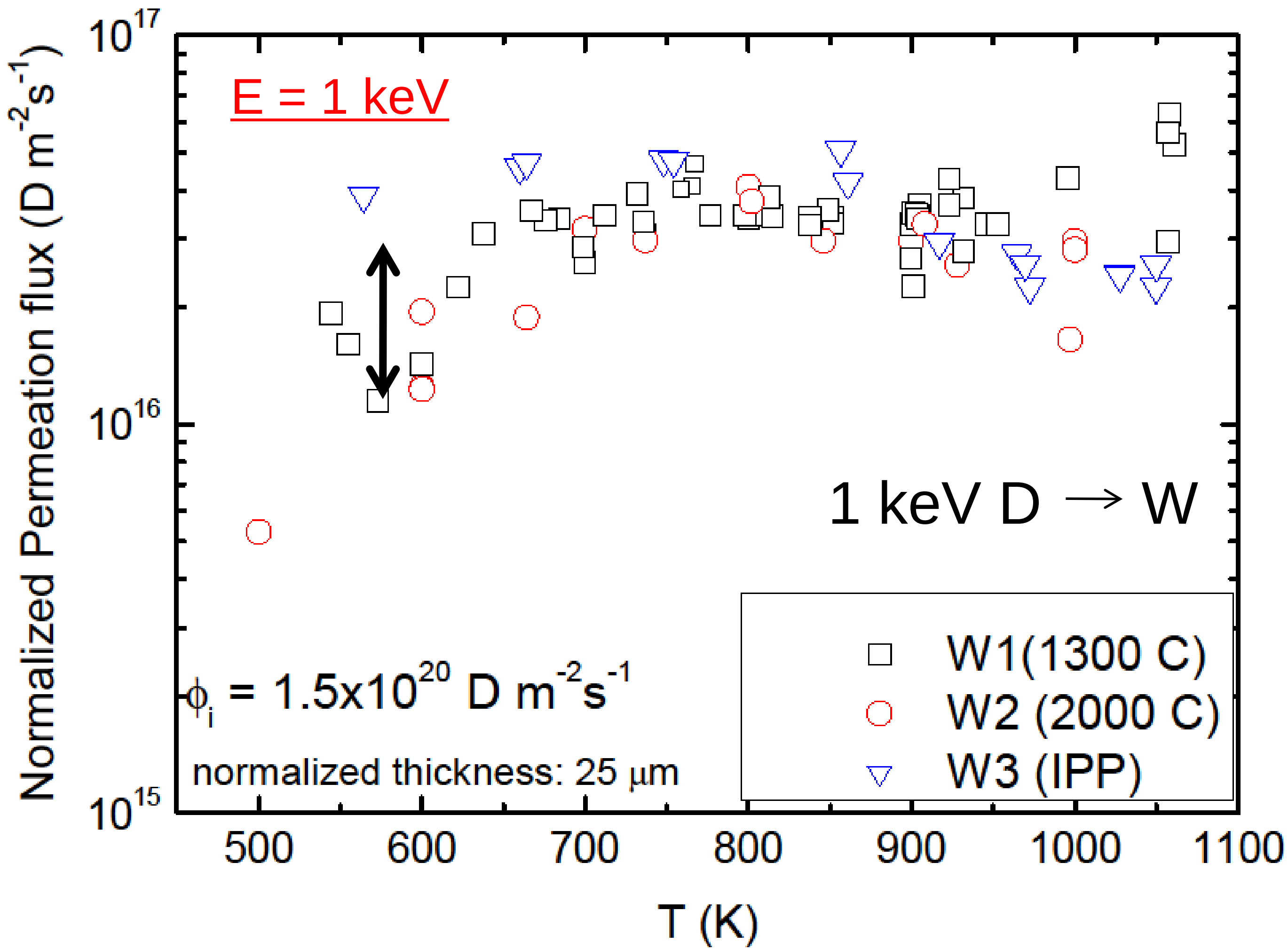
Tungsten specimens

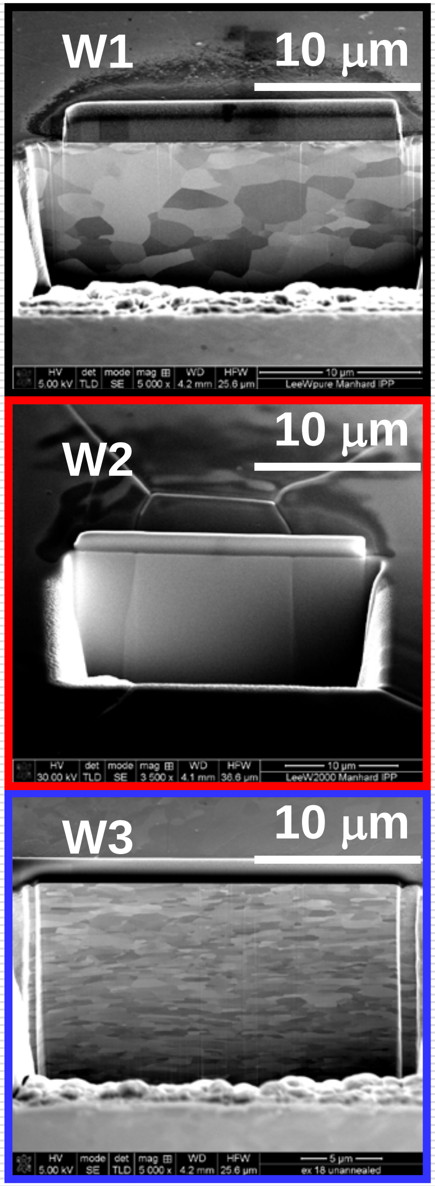
- W1: Sintered polycrystalline W → annealed at 1573 K.
  - Standard sample in our experiments (medium grains and low density defects)
- W2 : Sintered polycrystalline W → annealed at 2273 K.
  - Large grains and almost no defects
- W3 : Sintered polycrystalline W → annealed at 1203 K.



Microstructure dependence—SS permeation--

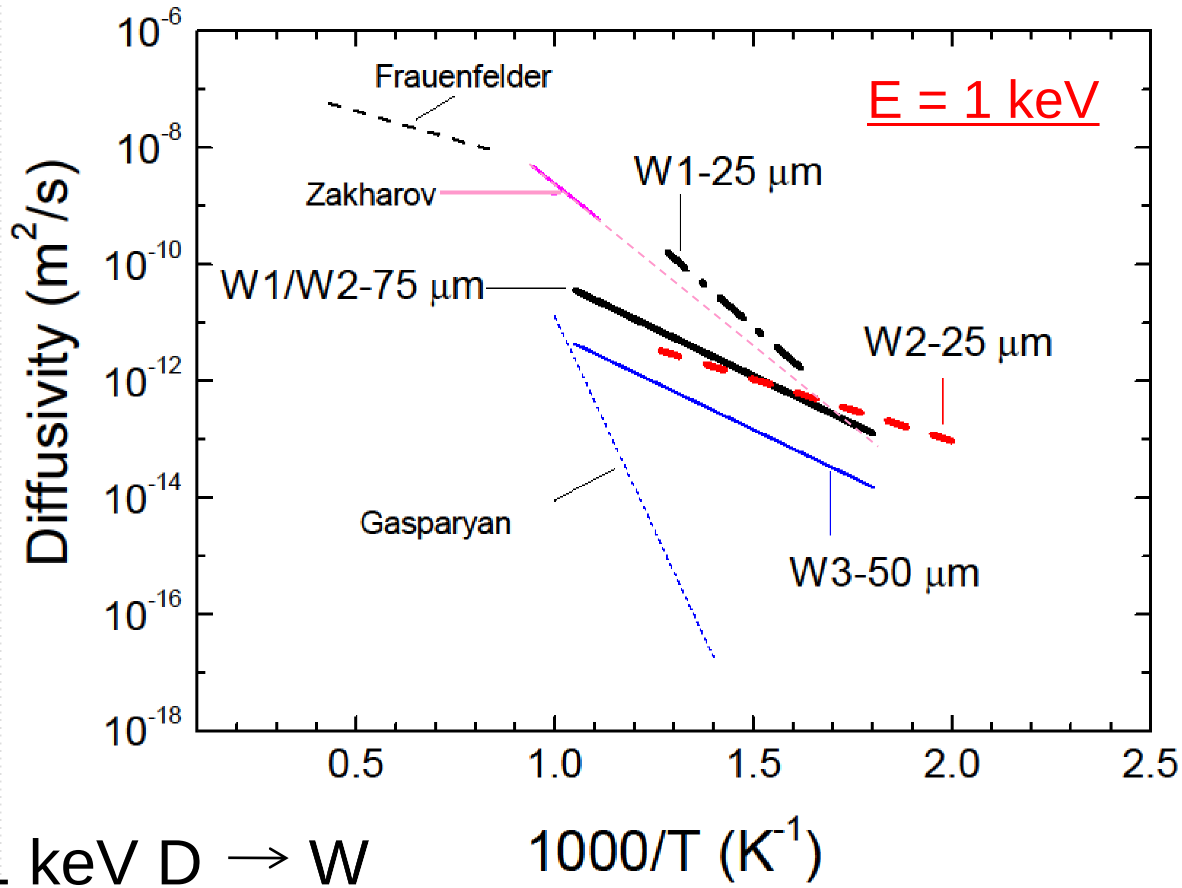
- Weak grain boundary dependence (factor of two)
- Peak in permeation flux observed T ≃ 800 K





H. T. Lee et al., Phys. Scripta., accepted

Microstructure dependence -Effective diffusivity-



1 keV D → W

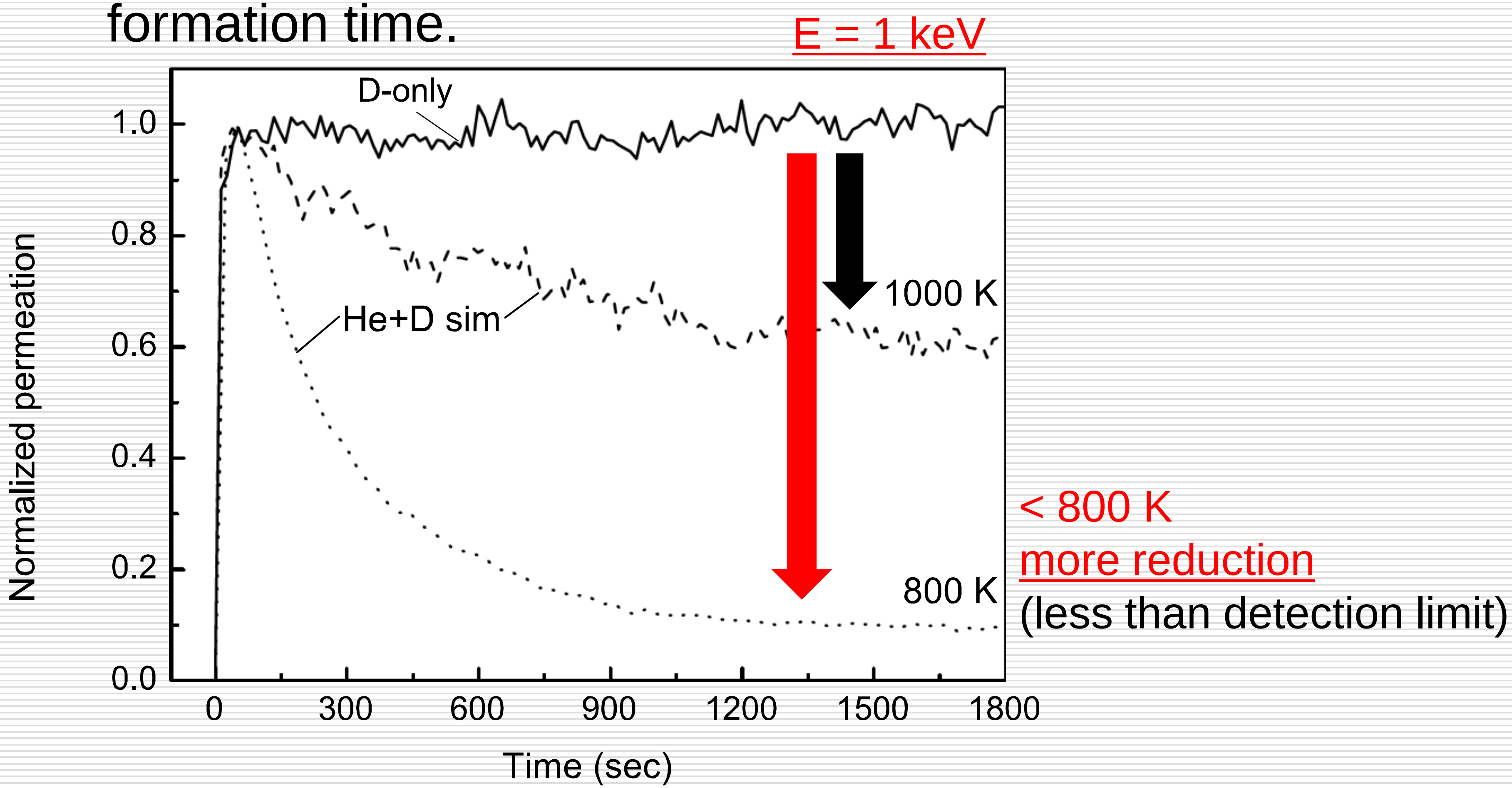
- Effective diffusivity values determined from lag time measurements.
- There are some dependences on thickness and microstructure.
- Effective diffusivities are close to Zakharov's, but much less than Frauenfelders.
  - This could be the effect of some traps.
- Need more investigation

R. Frauenfelder, J. Vac. Sci. Techn. 6 (1969) 388.
A.P. Zakharov, E.I. Evko. Fiz. Khim. Mekh. Mater. 9 (1973) 29.
Yu.M. Gasparyan et al., J. Nucl. Mater. 390 (2009) 606-609.

He/D mixed ion driven permeation

He/D mixed ion driven permeation

- Addition of He (2%) greatly reduces permeation flux.
- Saturation time almost corresponds to He bubble formation time.

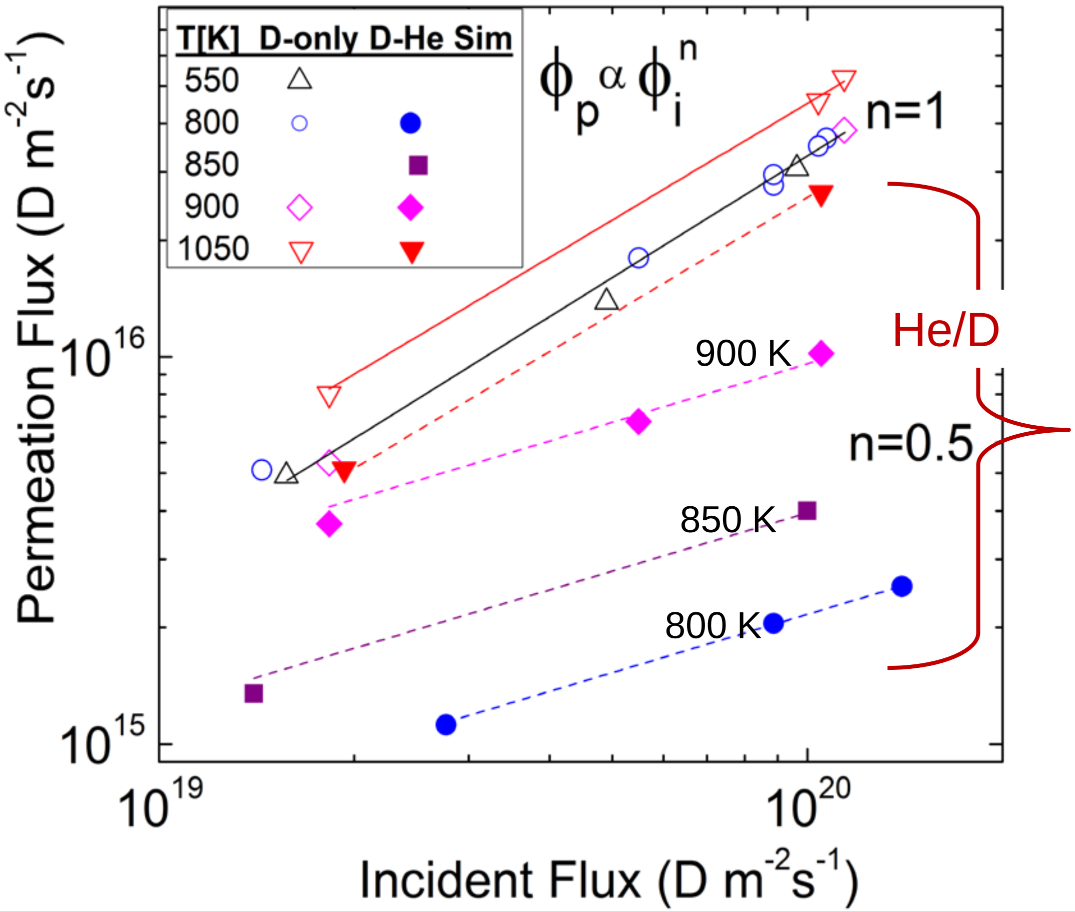


H. T. Lee et al., J. Nucl. Mater. (accepted)

Incident flux dependence

- $\phi_p \sim \phi_i$ (D only irradiation)
- $\phi_p \sim \phi_i^{1/2}$ (D/He irradiation)
  - ϕ_p : Permeation flux
  - ϕ_i : Incident flux
- Change of flux dependence suggests D release from the front surface could change from diffusion limited (D) to recombination limited (D/He).
- Two possibilities
  - Diffusion increased.
  - Recombination decreased.

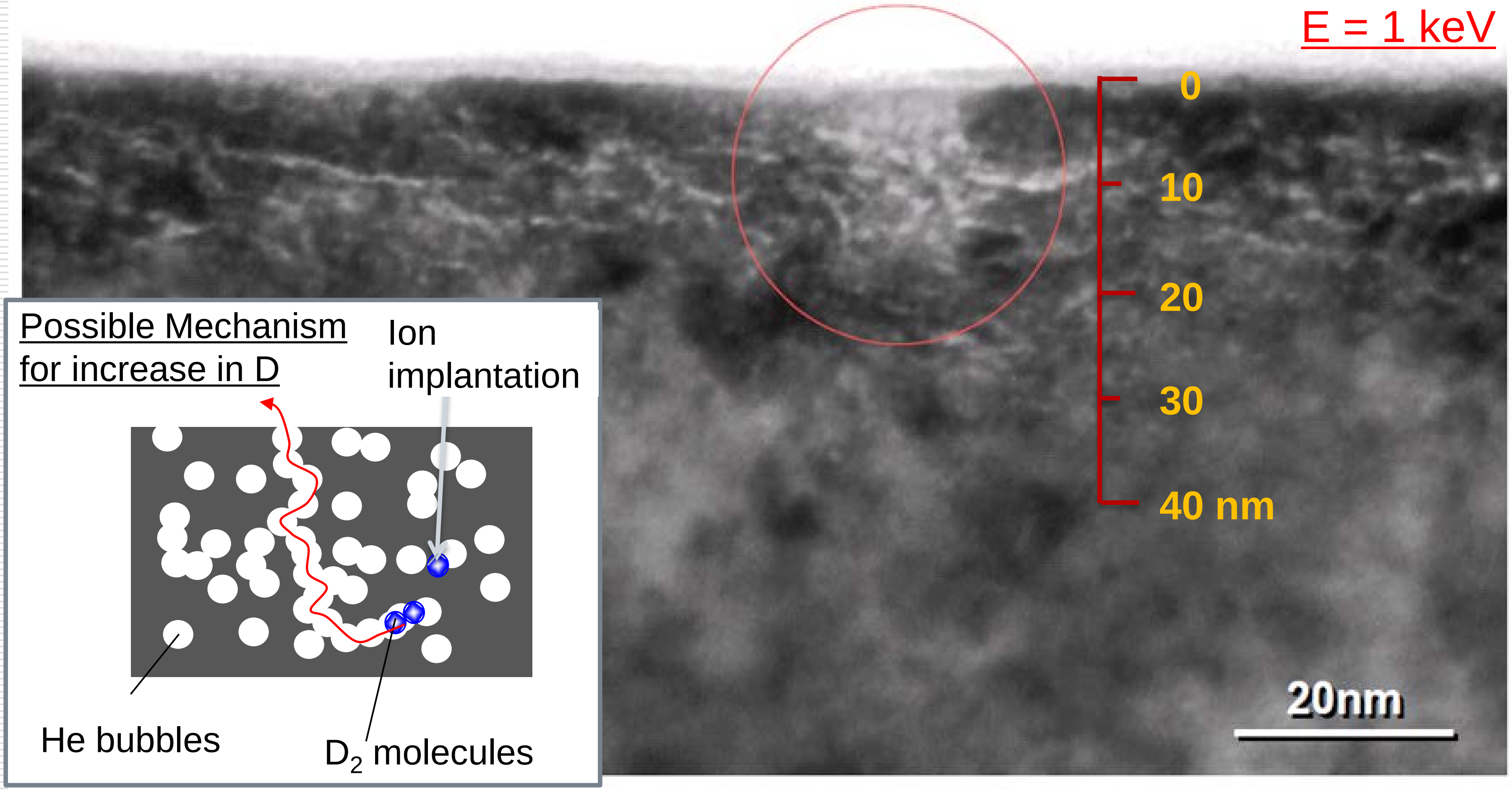
E = 1 keV He:2%



Permeation flux ϕ_p vs. Incident flux ϕ_i

Depth profile of He bubble layer

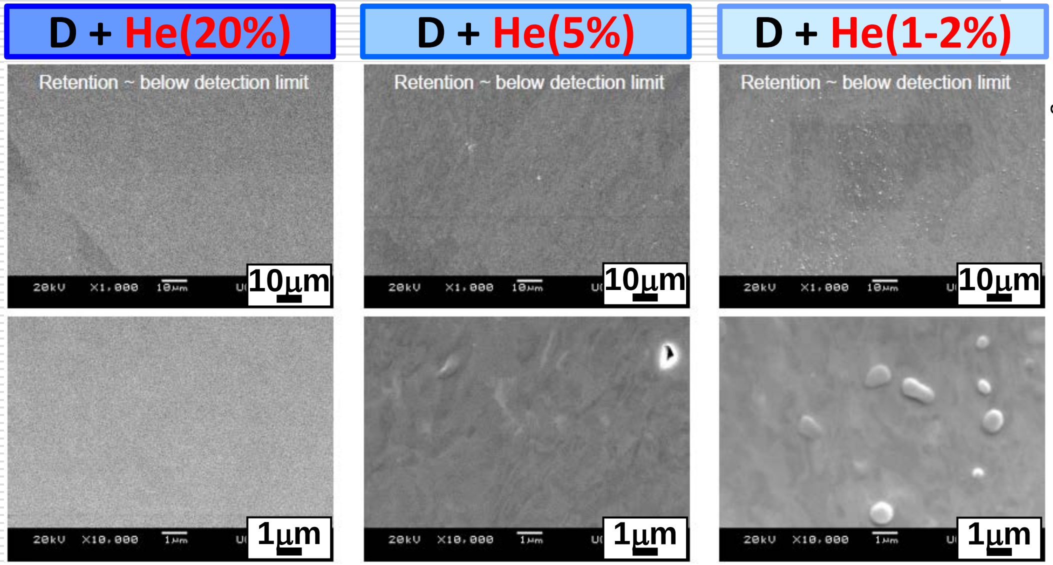
- He bubble layer was observed up to the depth of about 30 μm.
- Thickness of He bubble layer was larger than ion range（~10 μm).
- He bubbles are interconnected to form



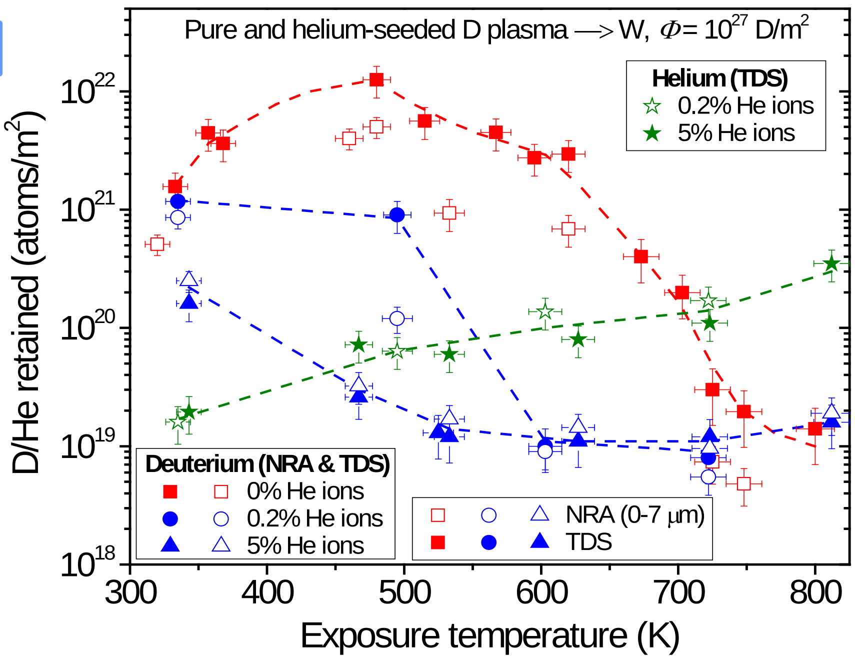
He/D effects on retention and blistering

- Blistering disappeared (or reduced) for He/D irradiation.
- D retention greatly reduced for He/D irradiation.

PISCES results (573 K)



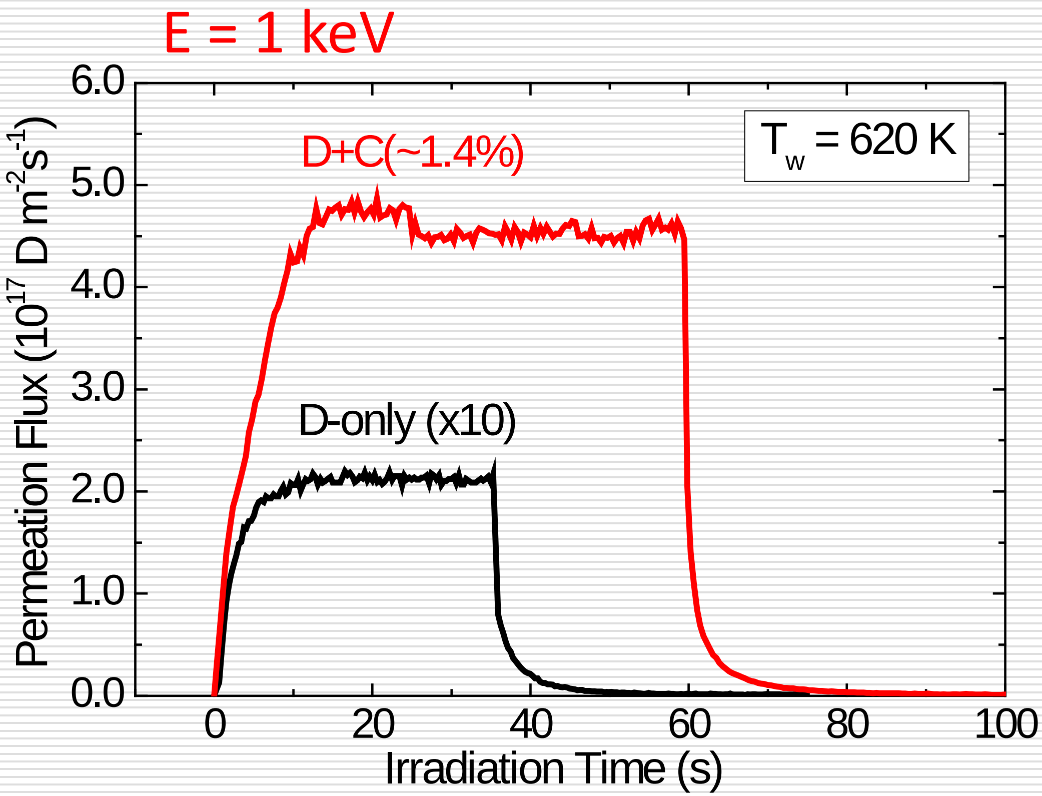
M. Miyamoto et al., Nucl. Fusion 49 (2009) 065035 (7pp).



V. Alimov et al., 12th ITPA (SOL/DIV)

C/D mixed ion driven permeation

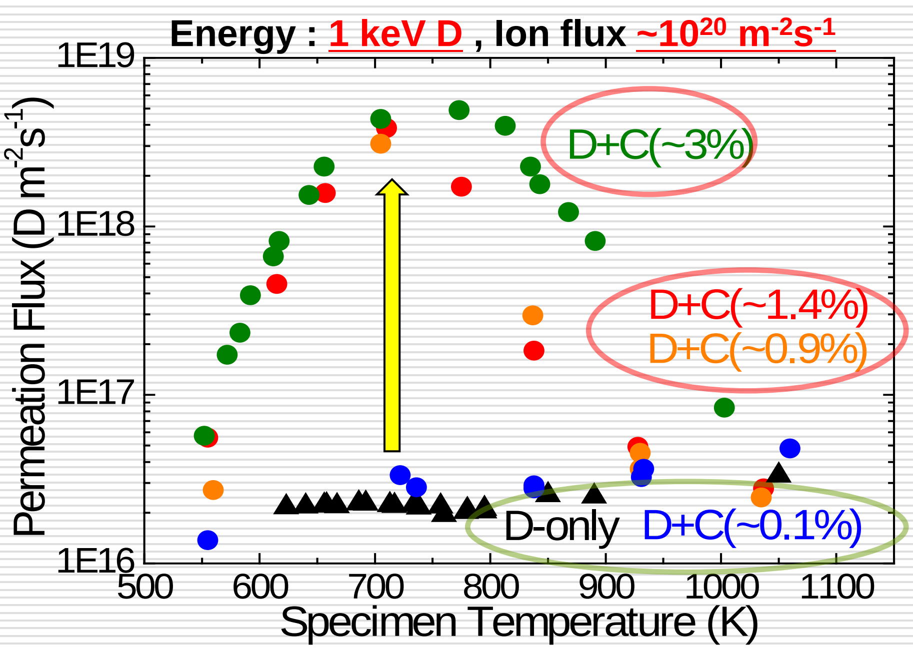
C/D mixed ion driven permeation



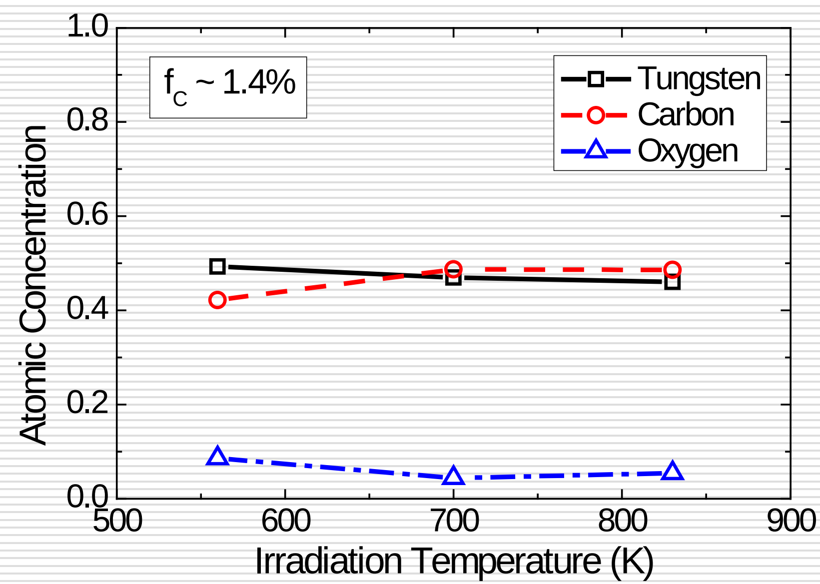
- The steady state D permeation flux for simultaneous D+C case is 20 times larger than that of D-only case.
- The lag time for simultaneous D+C case is 1.5 times larger than that of D-only case

Temperature & concentration dependence

- D permeation greatly increased even with C (>0.9%) in ion beam.
- Strong temperature dependence.
- Surface elemental composition shows little dependence on temperature (C:1.4%). → only temperature affect change of parameters
- Maximum increment factor in the present exp. : ~250



Temperature dependence of permeation

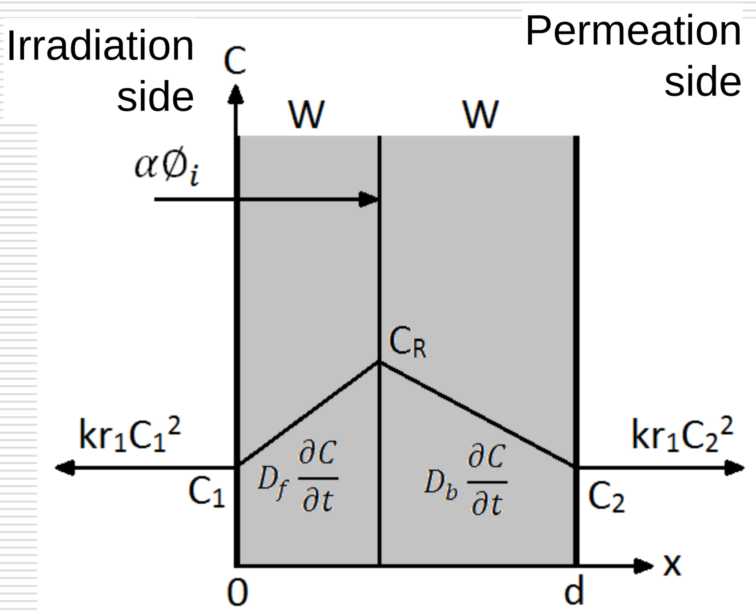


Near surface atomic composition

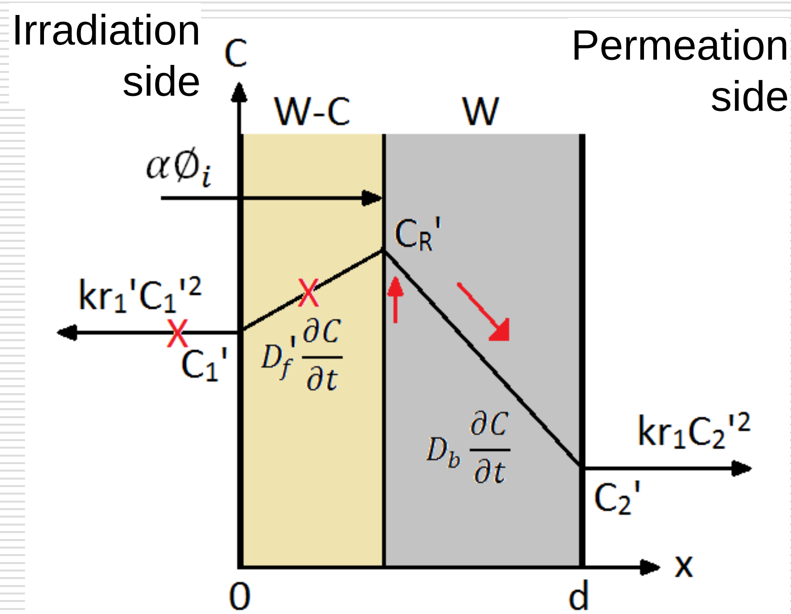
H.Y. Peng, Phys. Scripta., accepted

How C/W mixed layer affect permeation

- Surface carbon concentration was determined by the balance between re-erosion and implantation of C ions.
- Either significant reduction in surface recombination (more than 10^{-4}) or reduction in diffusion (10^{-2}) could cause significant increase in permeation.



Pure D irradiation

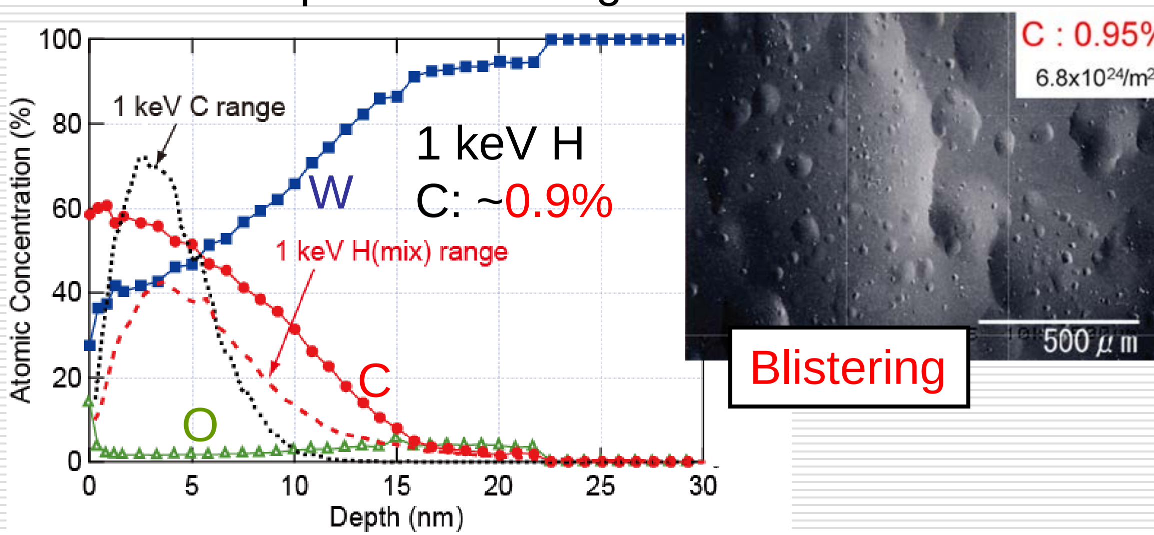


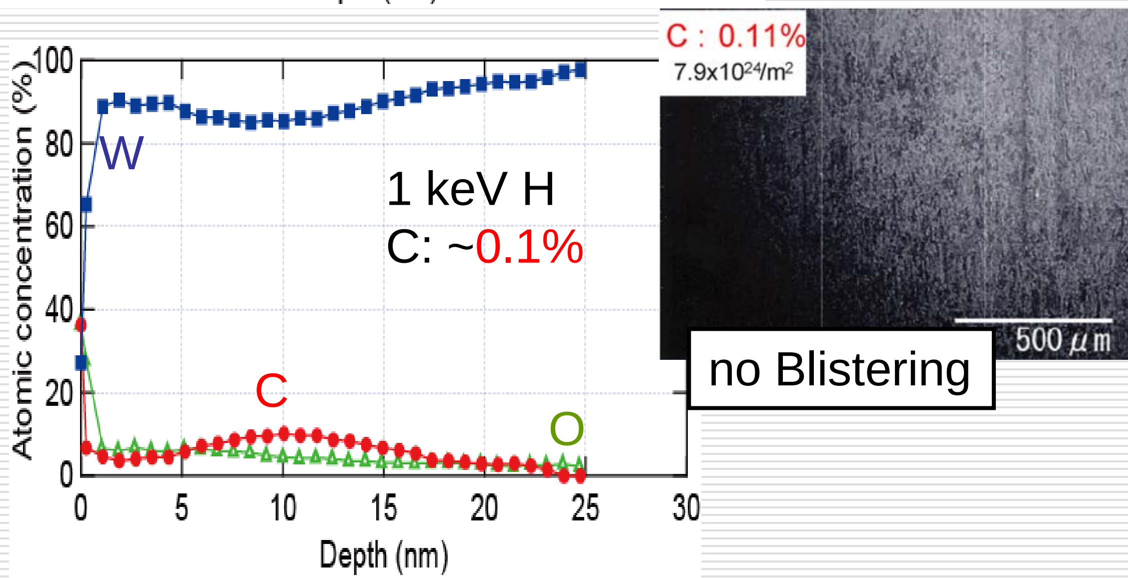
C/D mixed irradiation

Effect of C/W mixed layer on blistering

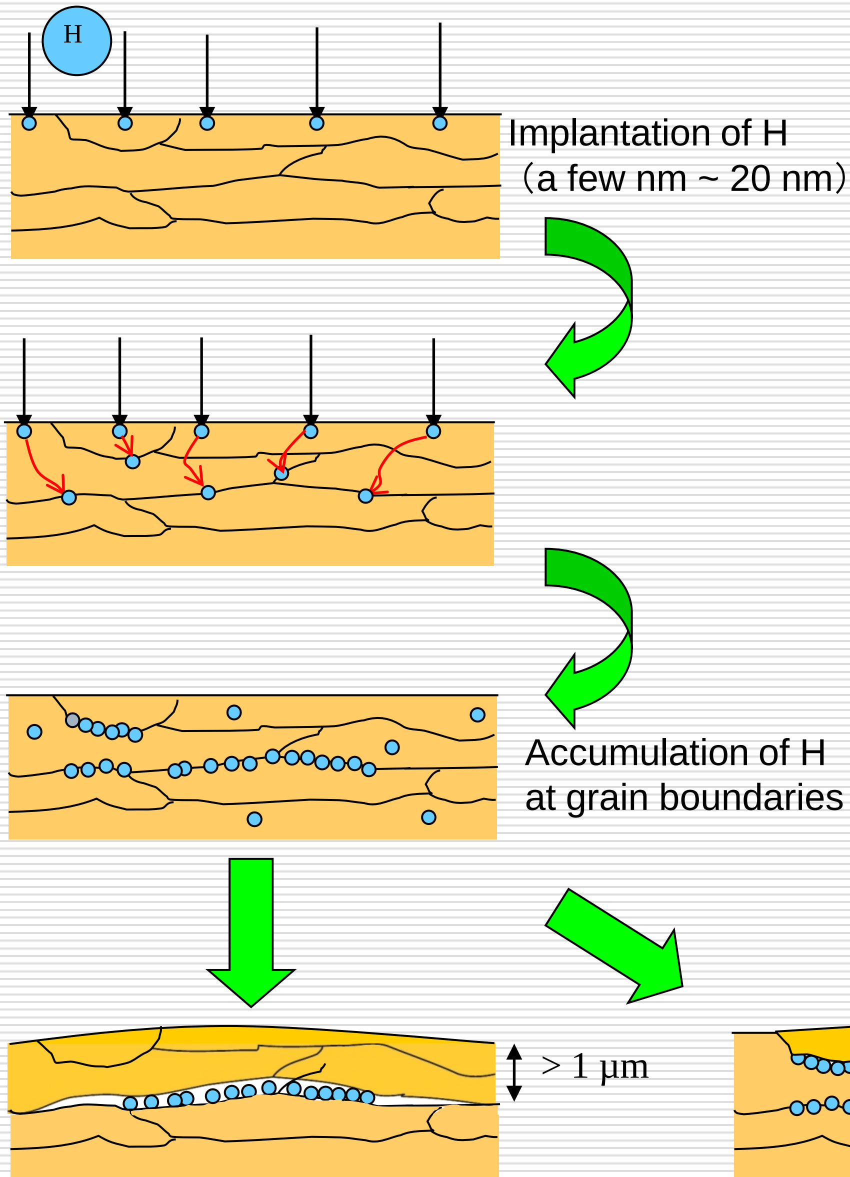
- For high carbon concentration in the beam (C:0.9%), blister appeared.
- C/W mixed layer effectively increases T diffusion into the bulk over certain fluence.

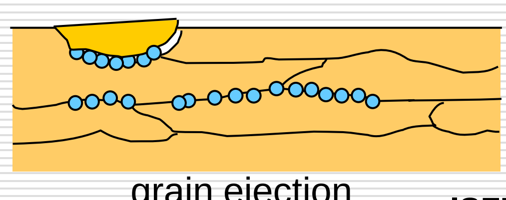
Atomic composition in tungsten

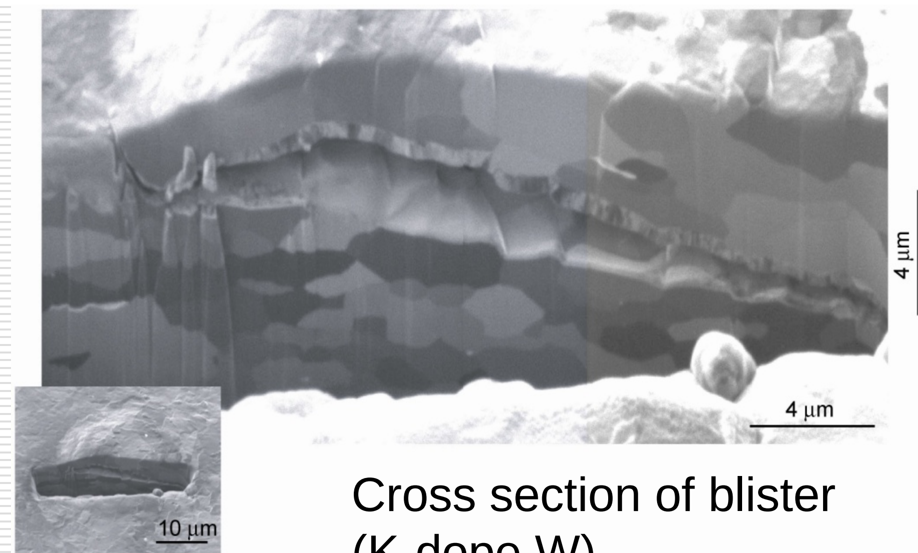




Mechanism for blistering







Cross section of blister
(K-dope W)

Diffusion of D into the bulk is necessary for blistering.

Enhanced bulk diffusion by the C/W layer causes blistering.

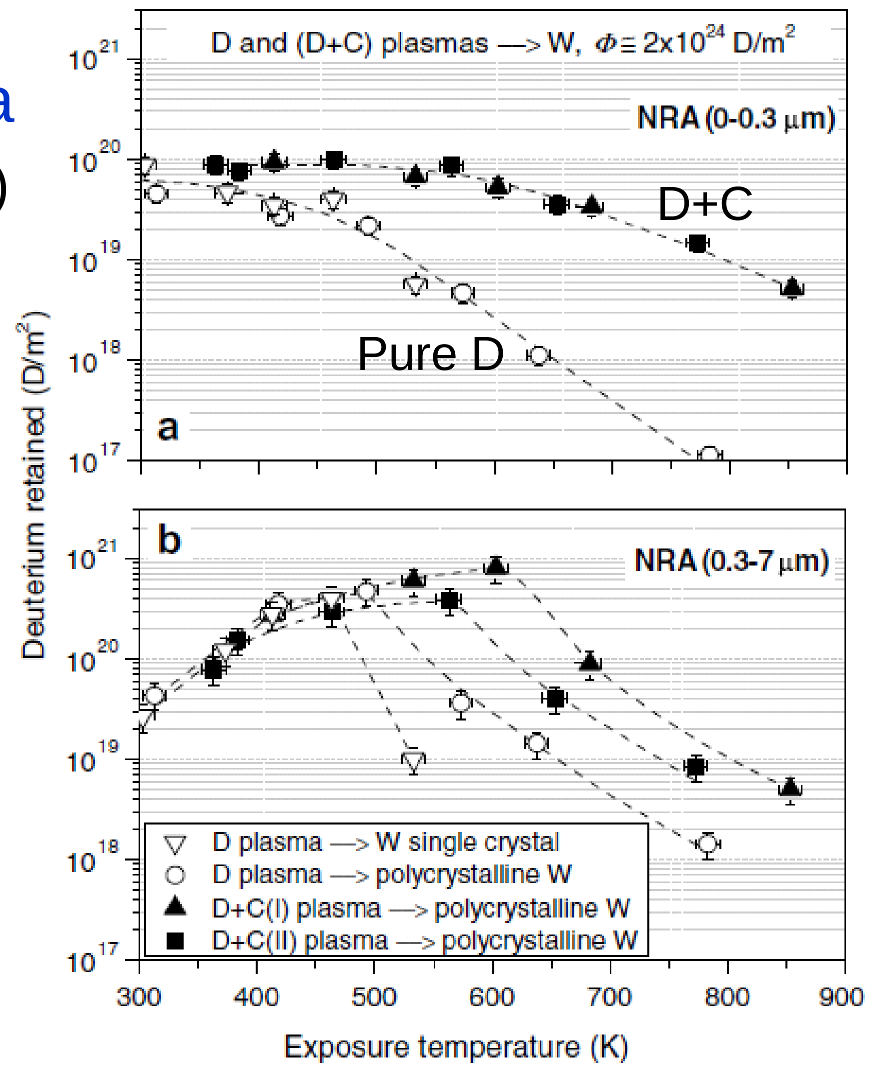
An opposite effect was observed for He/D.

ISFNT10, Y. Ueda et al., September, 2011

D/C mixed plasma exposure raised retention

- Planar DC magnetron plasma
  - Energy : ~200 eV (D_2^+ mainly)
  - Flux : 1 x 10^{21} $m^{-2}s^{-1}$
  - 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.

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

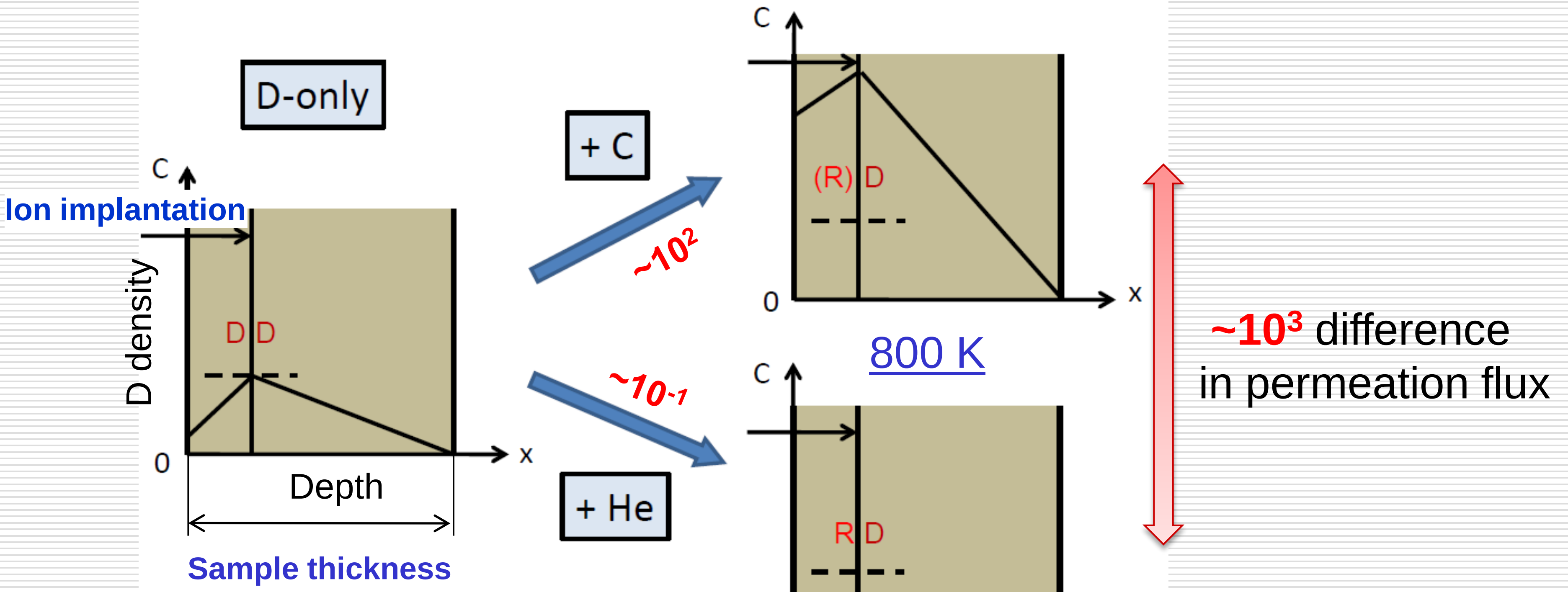


D retention in W exposed to pure D plasma (☐▽) and D+C plasma (▲■)

Comments on T behavior in blankets

- Surface phenomena greatly affect whole T behavior in blankets.
- Permeation barrier increases T concentration in materials
  - To reduce T permeation by putting permeation barrier on the permeation side, solute T density increase.→ Does it affect material performance?
    - 10^{-5} D/W for C/D irradiation (1.4%C, 800 K) (Assumption: Zakharov's effective diffusivity)
    - Trapped T density (equilibrium with solute T) increases with the increase in solute T.

Summary on D behavior for mixed irradiation



D: diffusion limited
R: recombination limited

- **Mechanism of permeation reduction (He/D) and enhancement (C/D)**
  - **Addition of He→ Effective diffusion near surface area increased.**
    Probably due to dense He bubble structure
  - **Addition of C→ Recombination or diffusion reduced : under investigation**

Concluding summary

- Mixed ion irradiation (He/D and C/D) into tungsten significantly changes permeation.
  - Addition of He : decrease (10^{-1} at 800 K, and more at <800 K)
    - Correlation with blister suppression
    - Correlation with decrease in retention
  - Addition of C : increase by two orders
    - Correlation with blister enhancement
    - Correlation with increase in retention

  Preferable for 1st wall

  - Strong temperature dependence
    - He or C effect significant at ~800 K
      → close to blanket surface temperature (FS based)
  - Indicating importance to include material mixing effects in T modeling in wall materials
- Microstructure dependence on permeation
  - No significant effects of microstructure (W1, W2, W3) on steady-state permeation