大阪大学
OSAKA UNIVERSITY

Towards an improved understanding of hydrogen transport in tungsten

H.T. Lee

Graduate school of engineering, Osaka University, Japan

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Acknowledgements

Group

Prof. Y. Ueda
Dr. K. Ibano

Students
Y. Hamaji
M. Oya
Peng Han Yee
H. Endo
M. Ishida
H. Katsuma
T. Soeda
K. Yakushji

Collaborators (Japan)
Research Center for Ultra-High Voltage Electron Microscopy
E. Taguchi, T. Nagase

Institute of Laser Engineering, Osaka University
N. Tanaka, H. Nishimura

RIKEN SPring-8 Center
H. Yamaoka

Doshisha University
M. Sasao (Prof. emeritus – Tohoku University), Y. Tawada

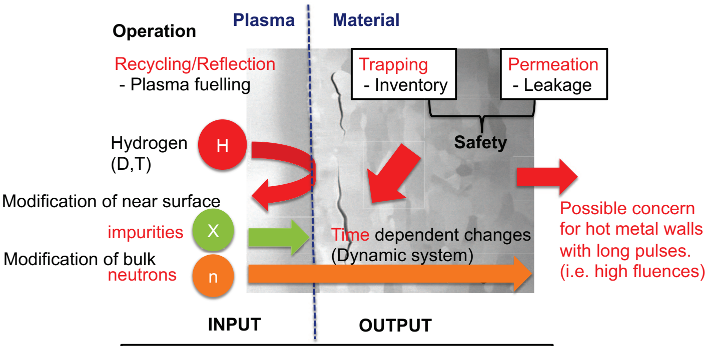
National Institute for Fusion Science
Y. Kisaki, A. M. Ito, Y. Oda

Collaborators (EU)
Max Planck Institute for Plasmaphysics, Germany
T. Schwarz-Selinger, A. Manhard, L .Gao, G. Meisl. T. Hoeschen

Dutch Institute for Fundamental Energy Research, Netherlands
G. De Temmerman*, Thomas Morgan, L. Cheng**

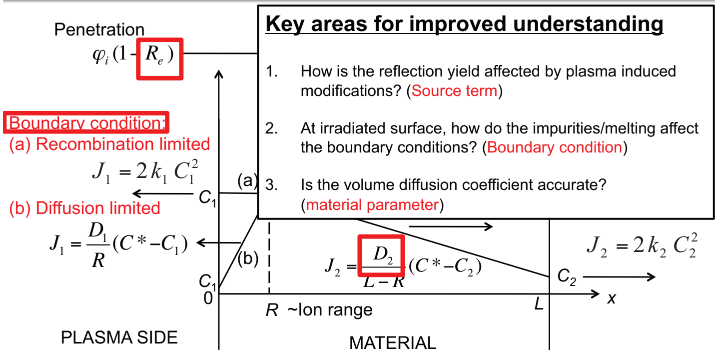
* Now at ITER Organization
** Beihang University, China

Accurate transport model should be able to predict recycling, trapping, and permeation processes



In general, temperature (and its variation) is the most important parameter for the above processes.

Standard hydrogen transport model under steady state conditions using empirical parameters



Key areas for improved understanding

1. How is the reflection yield affected by plasma induced modifications? (Source term)
2. At irradiated surface, how do the impurities/melting affect the boundary conditions? (Boundary condition)
3. Is the volume diffusion coefficient accurate? (material parameter)

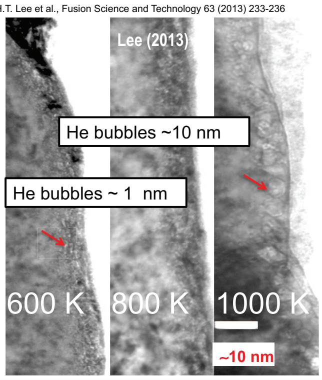
Hydrogen transport in solids (inter-relationship between plasma-material parameters)
B.L. Doyle, J. Nucl. Mater 111/112 (1982) 628.
I. Ali-Khan, K.J. Dietz, F.G. Waelbroeck, P. Wienhold, J. Nucl. Mater. 76-77 (1978) 33.

Key areas for data needs/improved understanding

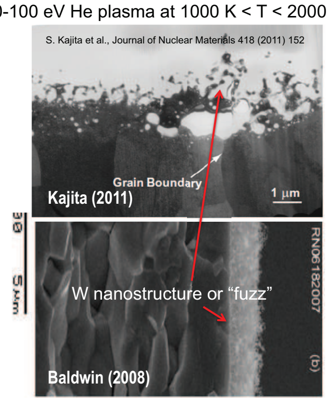
1. How is the reflection yield affected by plasma impurity modifications? (very little data)
2. At irradiated surface, how do the impurities/melting affect the boundary conditions?
3. Is the bulk diffusion coefficient accurate?

Two regions of He induced W near surface modification

1) First-wall like conditions
(keV He ion implantation at T < 1200 K)
H.T. Lee et al., Fusion Science and Technology 63 (2013) 233-236

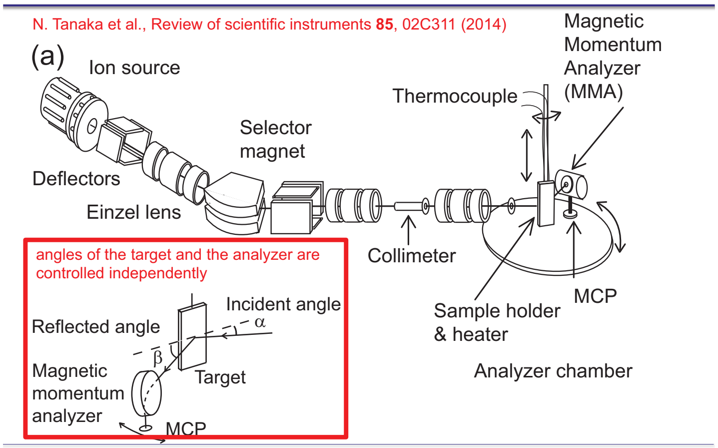


2) Divertor-like conditions
(20-100 eV He plasma at 1000 K < T < 2000 K)
S. Kajita et al., Journal of Nuclear Materials 418 (2011) 152



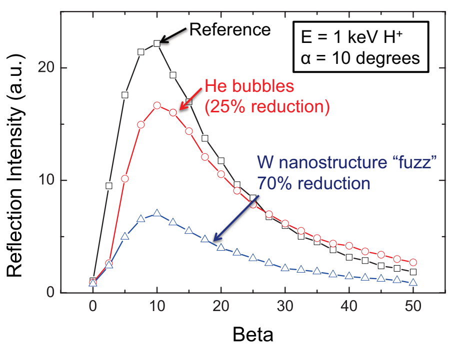
Schematic of reflection experiments

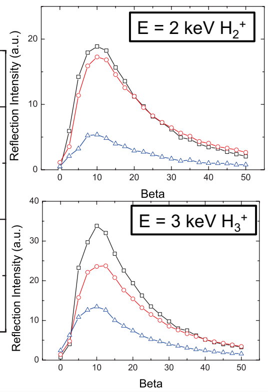
N. Tanaka et al., Review of scientific instruments **85**, 02C311 (2014)



Such data are important for defining source terms in modeling (both edge/material)

Analysis ongoing by comparison to BCA codes/ further experiments planned:
1) Incident species (He)
2) Surface state (D, N, Ne implanted surfaces)

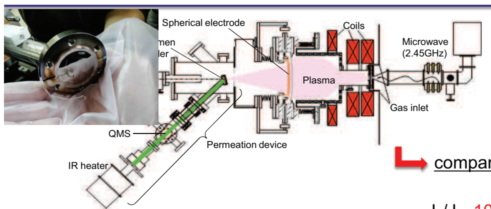




Key areas for data needs/improved understanding

1. How is the reflection yield affected by plasma induced modifications? (very little data)
2. At irradiated surface, how do the impurities/melting affect the boundary conditions?
3. Is the bulk diffusion coefficient accurate?

Experimental approach – Ion driven permeation



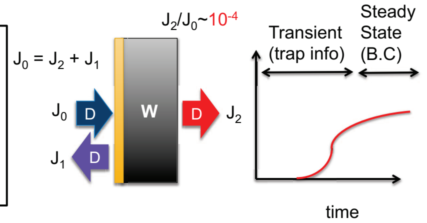
Ion beam apparatus

- Γ=O(10^{20-21} /m²s)
- E>O(100 eV)
- T < 1000 K

comparable to first wall conditions

Main advantages of permeation experiments:

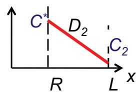
1) Steady state measurements (when one stage is rate limiting)
2) Indirect measurement of solute concentration

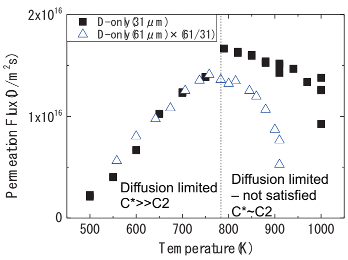


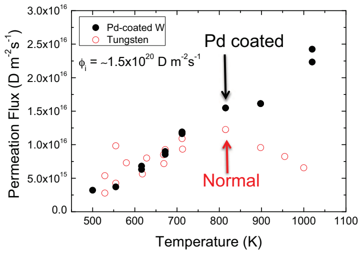
The challenge in interpreting permeation experiments is to correctly identify the rate limiting stage and to extract relevant parameters

- Thickness dependence confirms bulk diffusion is rate limiting and R<<L:

$$J_2 = \frac{D_2}{L}(C^* - C_2)$$







M. Ishida, H.T. Lee et al, J. Nucl. Mater. (2014), in press (doi:10.1016/j.jnucmat.2014.11.123)

Under D-irradiation, the diffusion limited boundary condition is valid provided the front diffusion coefficient is temperature independent

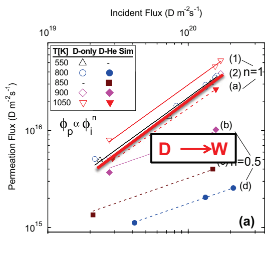
Permeation flux depend linearly on incident flux:

rule out recombination limited release.

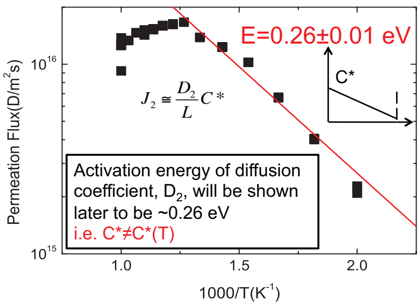
$$\varphi_i(1-R_e) = J_1 = \frac{D_1}{R}C^*$$

$$\boxed{C^* \propto \varphi_i(1-R_e)}$$

- If diffusion limited - diffusion coefficient must be temperature independent. (~4×10^{-10} m²/s if R ~ 10 nm - reasonable)

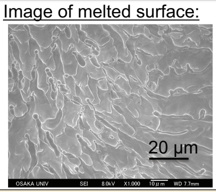


H.T. Lee et al., J. of Nucl. Mater., 415 (2011) S696

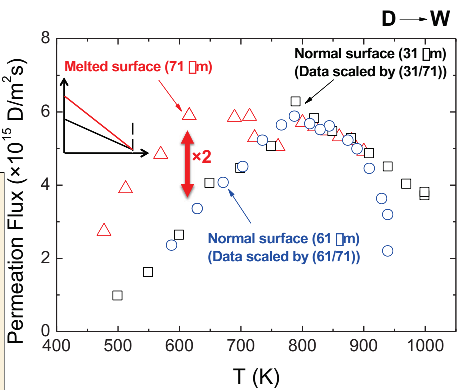


Macroscopic effects (melting, surface roughness increase) result in modest increase in near surface concentration

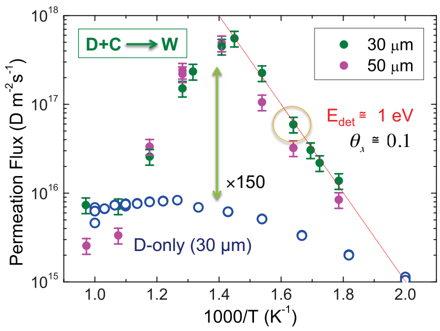
Image of melted surface:



- Macroscopic surface roughness increases (×4)
- T ≥ 700 K, an increase in steady state flux for melted surface (×2). (i.e corresponding increase in solute C*)
- Decrease in D1 but still T independent.



Permeation experiment indicate no incident flux dependency at T = 600 K

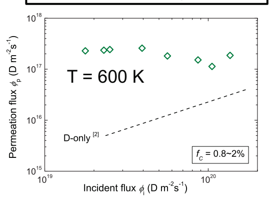


It is suggested that this θ_x may be the "dynamic" retention observed during irradiation and will certainly influence the outgassing behavior.

If trapped concentration controls the solute concentration:

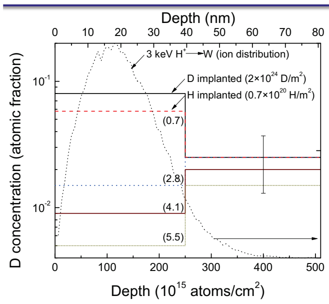
$$J_2 = \frac{D_o NZ}{L}\left(\frac{\theta_x}{(1-\theta_x)}\right)\exp\left(\frac{-E_{det}}{kT}\right)$$

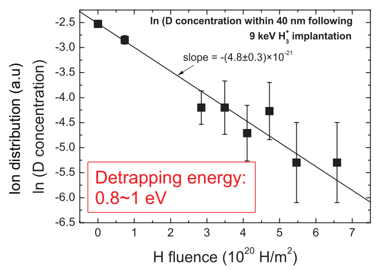
No incident flux dependence

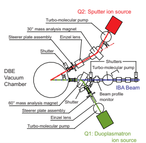


H.Y. Peng, H.T. Lee et al., J. of Nucl. Mater., 438 (2013) S1063

Ion-induced traps show detrapping energies of 0.8~1 eV and saturation density 8 at.%







In-situ experiment
9keV D_3^+, H_3^+

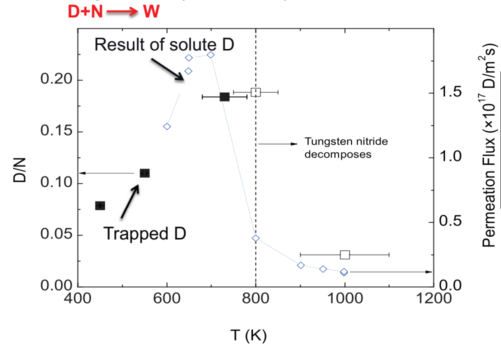
1) D ⟶W (300 K)
2) H ⟶W (300 K)

- Kinetics studied by isothermal D-H isotope exchange.
- Near surface release is first order – (i.e. limited by detrapping)

H.T. Lee and T. Schwarz-Selinger, Nucl. Instr. Meth B.(2014) under review

With nitrogen impurities the analogous concept of trapped concentration appear to control the solute concentration

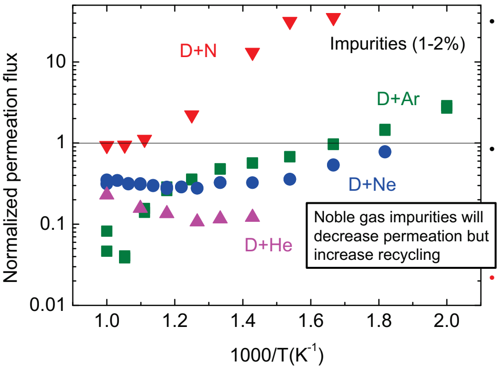
Magnum PSI (~10²⁴ D/m²s)
D+N ⟶ W



- The picture is more complex with N impurities likely due to the changing nature of trapped D/N with temperature

H.T. Lee, G. De Temmerman, L. Gao, T. Schwarz-Selinger, G. Meisl, T. Hoeschen, and Y. Ueda, J. of Nucl. Mater., in press (2014)

Difference in volatile vs noble gas impurities



- At present, we do not have enough data to test which boundary conditions hold for the noble gases.
- Even so, such scaled data is valuable for design studies (e.g. tritium permeation barriers)
- Sputtering and any synergistic effects need to be clarified.

Helium: H.T. Lee et al., J. of Nucl. Mater., 415 (2011) S696
Nitrogen: H.T. Lee et al., Phys. Script. T159 (2014) 014021
Neon, Argon: M. Ishida, H.T. Lee et al, J. Nucl. Mater. (2014), in press (doi:10.1016/j.jnucmat.2014.11.123)

Key areas for data needs/improved understanding

1. How is the reflection yield affected by plasma induced modifications? (very little data)
2. At irradiated surface, how do the impurities/melting affect the boundary conditions?
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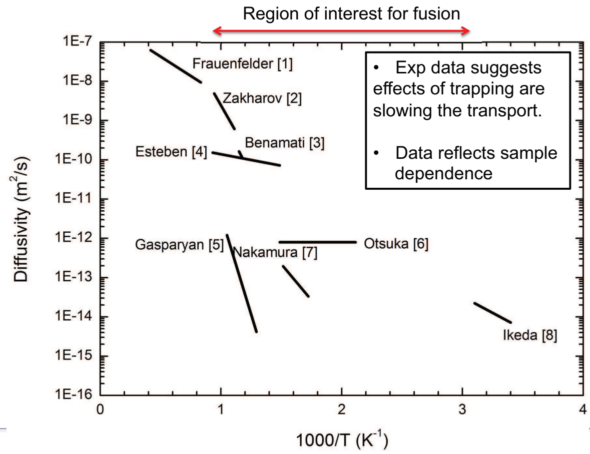
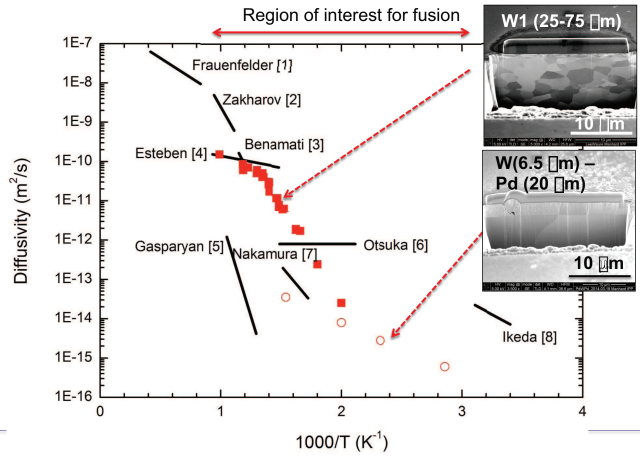
Significant scatter in data in temperature region of most interest in fusion (300-1000 K)

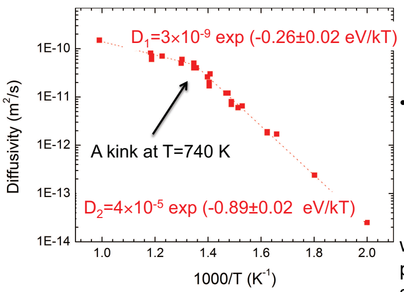


Bridged the experimental gap (300-1000 K)



High temperature data indicate difference in activation energy for diffusion in comparison to accepted values

Frauenfelder's values (experimental)
D=4.1×10^{-7} exp (-0.39 eV/kT)
DFT values
0.39 eV – Johnson, 0.2-26 eV – Heinola



- Change in slope at T=740 K

Interpreted to be from change in no trapping to sparse trap occupation

- When traps sparsely occupied

$$D_{eff} = D_L(1+\alpha)^{-1}$$

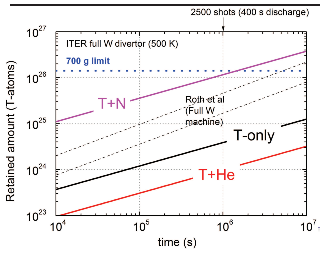
$$\alpha = (6.3 \pm 0.1)\times 10^{-5} \exp\left(\frac{0.64 eV}{kT}\right)$$

where α contains the trap parameters (natural traps for our specimen).

Comparison to recommended values

Phys. Scr. **T145** (2011) 014031 **Table 1.** Data for the transport parameters of hydrogen in W J Roth and K Schmid

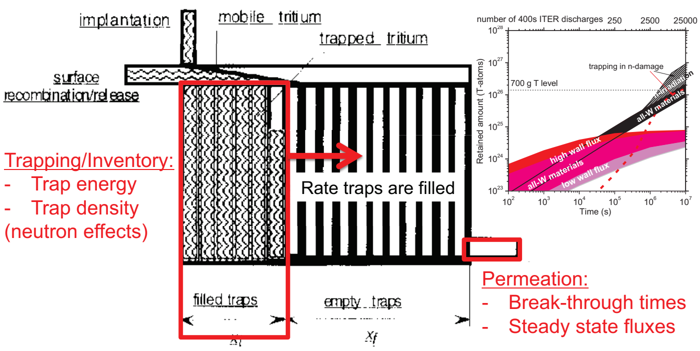
Recommended values [14]	Activation energy (eV)	Pre-exponential factor	Trap concentration (at.%)	Reference
Diffusion coefficient	−0.39 (0.26)	4.1×10^{-7} (m^2 s^{-1})	(>3×10^{-9})	Frauenfelder [2]
Recombination rate coefficient	1.16	3.2×10^{-15} (m^4 s^{-1})	(Varies by impurity May not matter for C/N impurities)	Anderl [8]
	−2.06	$3 \times 10^{-25}/T^{1/2}$ (m^4 s^{-1})		Pick and Sonnenberg [9]
Solubility	−1.04 (1.17)	9×10^{-3} (H/W atm$^{1/2}$)	(7.5×10^1)	Frauenfelder [2]
Intrinsic traps	0.85 (0.9)		(6×10^{-5}) 4×10^{-4} to 10^{-2}	Ogorodnikova [10]
Ion-induced traps	1.45 (0.8)		(8) Up to 10	Ogorodnikova [10]
n-induced traps	1.8–2.2		Up to 1	Tyburska *et al* [11]



	Divertor
Sample thickness (mm)	8
Total area (m^2)	50
Surface temperature (K)	500
D^+ incident flux (m^{-2}s^{-1})	2.0x10^{23}
D^+ incident energy (eV)	48
Diffusion coefficient (front, back) (x 10^{-11} m^{2}s^{-1})	(3.44, 3.44)
Trapping density (at.%)	1
Trapping energy (eV)	1.8

Hydrodynamic analogue of hydrogen transport in solids when trapping is active

Transport: 1-D diffusion trapping model



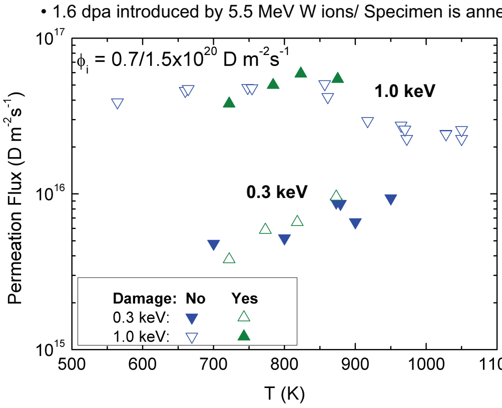
Trapping/Inventory:
- Trap energy
- Trap density (neutron effects)

Permeation:
- Break-through times
- Steady state fluxes

G. Federici, D.F. Holland, R. Matera, Journal of Nuclear Materials. 233-237 (1996) 741–746.

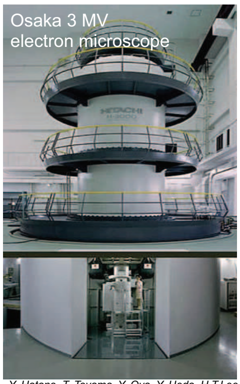
W self-damaged experiments

- 1.6 dpa introduced by 5.5 MeV W ions/ Specimen is annealed at 850 K.



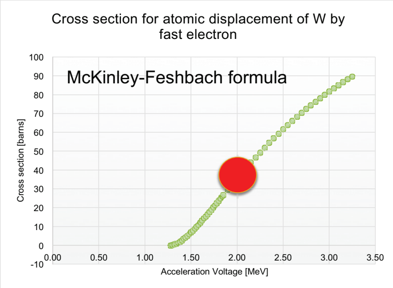
- Traps introduced play no role in steady state permeation consistent with classic diffusion and trapping theory.
- since trapping in n-damaged sites depend on fluence (dpa) if the system reaches steady state prior to such effect ongoing n-damage will not alter permeation.

Neutron effects – fast electrons as a means to study fundamental irradiation damage

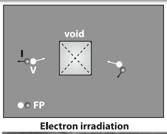


Y. Hatano, T. Toyama, Y. Oya, Y. Ueda, H.T.Lee IAEA Plasma-Wall Interaction with Irradiated Tungsten and Tungsten Alloys in Fusion Devices (2013-2018)

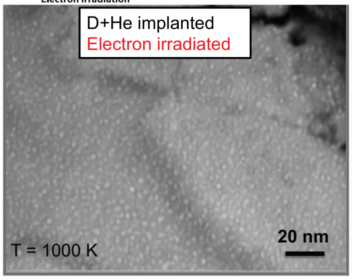
<u>High cross sections and focusable beam allows:</u>
- Fluence studies: high dpa (displacement per atom)
  e.g. 1 dpa ~ 100 s; 10 dpa ~ 1000 s
- Flux studies: dose rate (importance of defect transport kinetics)

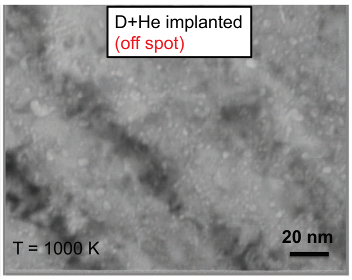


Examining the dynamics of He bubble growth under irradiation conditions



- A unique aspect of the DT fusion environment is **substantial** production of gaseous transmutants such as He and H.
- How are the growth/re-combination rates of He bubbles affected in the presence of He/H/displacement damage?





Summary

- Improved modeling of hydrogen transport in tungsten requires improved source, boundary, diffusion, and trapping parameters (impurities and neutrons effects)
- Laboratory reflection, permeation, TEM experiments can provide such basic data:
  - He modified layers can result in 25-75% reduction in reflection yield.
  - Surface melting and roughening results in increased near surface concentration (factor of two) but is modest in comparison to impurities.
  - C,N impurities: orders of magnitude increase in permeation flux (Temperature) with a corresponding decrease in the recycling flux. Trapped concentration controls solute concentration.
  - Noble gas impurities (He, Ne, Ar) generally decrease the permeation flux, with corresponding increase in recycling flux. The effects of sputtering need to be clarified.
  - The activation energy of diffusion appears to be ~0.26 eV.
  - MeV TEM experiments is proposed as a tool to study fluence (dpa) and flux (dpa/s) effects of irradiation damage.