U.S. DEPARTMENT OF ENERGY
Office of Science

Technical Meeting on Plasma Physics and Technology Aspects of the Tritium Fuel Cycle for Fusion Energy:
Topic 1: Plasma Chamber and Tritium Behavior

Plasma chamber PMI – Linear plasma facilities (TPE, implantation and irradiated materials)

We are trying to address:
"T retention issues associated with high neutron fluence, high T_{wall} permeation not fully addressed in ITER"
with linear plasma device and irradiated materials.

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INL/MIS-22-69644

Outline

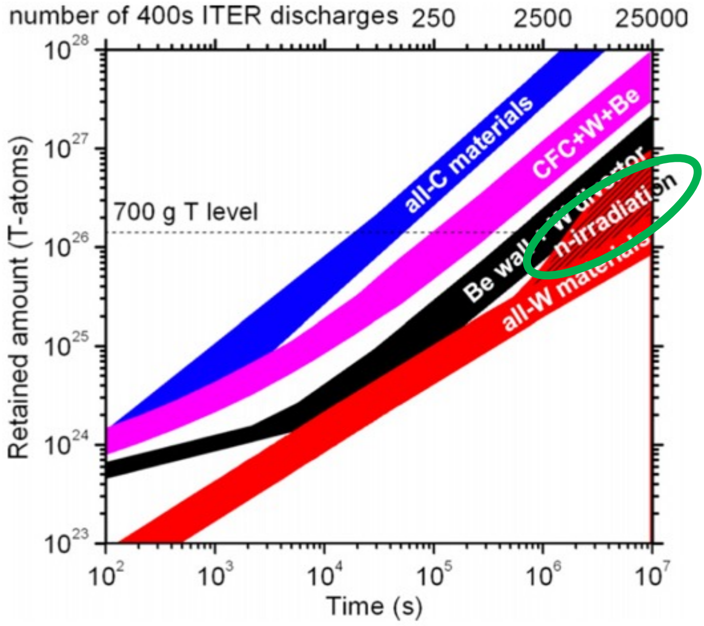
- Motivation
- Experimental apparatus
- Tritium migration and trapping in neutron-irradiated tungsten
- Recent progress on tritium retention with neutron-irradiated tungsten
- Summary

Motivation: Tritium behavior and IAEA

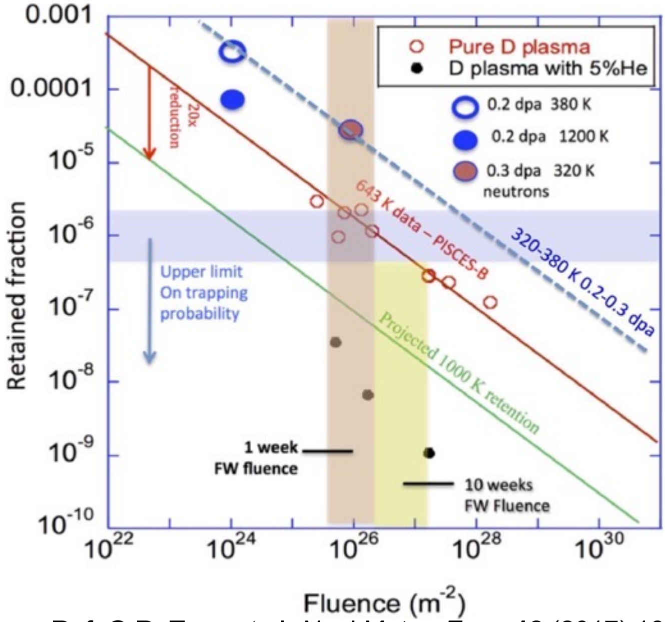
- **Why INL is investigating tritium behavior (tritium retention and permeation):**
  1) **Tritium retention is one of the major concerns for fusion safety**
     - Tritium transport database determines both tritium retention and permeation in safety analysis
     - IAEA CRP on "Hydrogen Permeation in Fusion-Relevant Materials (2020-2025)" (F43025)
  2) **H/D/T transport database in fusion materials with tritium-exposed and low-activation materials**
     - INL's experimental capabilities for tritium-exposed and low-activation (e.g. neutron-irradiated) materials
       - H/D/T permeation systems (SGAP and TGAP) to to measure H/D diffusivity, solubility, surface dissociation/recombination coefficients, and also T diffusivity and solubility
       - Linear plasma device (TPE) to study PMI in neutron-irradiated materials and measure detrapping energy and trap density in low-activation materials
     - IAEA CRP on "Plasma-wall Interaction with Irradiated W and W Alloys in Fusion Devices (2013-2018)" (F43021)
  3) **Development of analytical/computational tools (e.g. MELCOR-TMAP and TMAP) for safety analysis**
     - Integration of TMAP (Tritium Migration Analysis Program) capability in MECLOR-fusion (1.8.6) ➔ MELCOR-TMAP
     - Development of TMAP8 (new version of TMAP is based on MOOSE, which is extremely flexible environment that permits the solution of coupled physics problems of varying size and dimensionality)

Motivation: H(T) retention studies

- **Safety concern and impact to PFC material choice**
  - **Minimize tritium in-vessel inventory in D-T fusion systems**
    - Allowable tritium in-vessel components inventory limit 1kg in in-vessel components [1]:
      - The maximum retention in the in-vessel components is set at 700 g with:
        - 120 g of T trapped on the divertor cryopumps and
        - 180 g of measurement uncertainty
  - **Tritium (T) retention has a major impact on PFC material selection**
    - T retention was a major factor to exclude CFC from ITER [2]
    - Tungsten (W) and W alloys are candidate material for solid PFC option for DEMO & FPP.
- **Challenge for achieving self sustainable T fuel cycle**
  - **Effect on tritium breeding ratio [3]**
    - Probability of permanently trapped tritium in W needs to be below 10^{-6} to reach tritium breeding ratio (TBR) of TBR> 1.05
- **Research gaps in T retention**
  - Incident D/T ion fluence above 10^{28} m^{-2} (up to 10^{31} m^{-2}) e.g $4x10^{26}$ m^{-2} = 1kg(T) m^{-2}
  - Effect of impurity (e.g. helium, nitrogen)
  - Effect of mixed materials in net deposition region
  - Radiation damage above 1 dpa (up to 10-20 dpa) with fusion neutron spectrum



Ref: J. Roth et al., *PPCF* **50** (2008) 103001



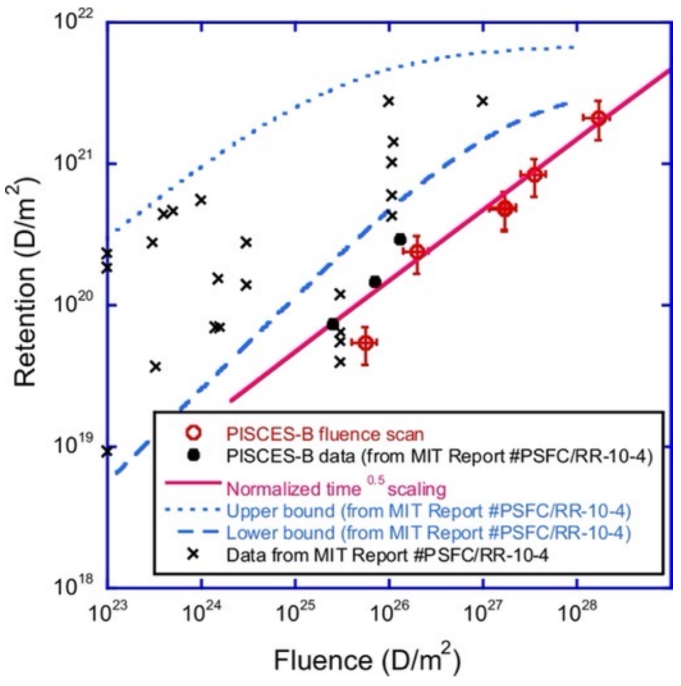
Ref: G.R. Tyan et al, *Nucl.Mater. Ener.* **12** (2017) 164

References:
[1] De Termmeman et al., *Nucl. Mater. Energy* **12** (2017) 167 ,[2] Roth et al., *Plas. Phys. Contr. Fusion* **50** (2008) 103001, [3] G.R. Tyan et al,, *Nucl. Mater. Energy* **12** (2017) 164

Key questions for tritium retention in tungsten

1) T retention behavior at high ion fluence above 10^{26} m^{-2}

- **2010 MIT report (ITPA SOL/divertor group) suggested saturation [1]**
  - Collected D retention data around 500K from 10^{23} to 10^{27} m^{-2} ion fluence
  - Suggested the possibility of **saturation** in retention above 10^{26} m^{-2}
- **2016 PISCES-B study suggests the increase with square root of time [2]**
  - Explored D retention data at 640K from 7x10^{25} to 2x10^{28} m^{-2} ion fluence
  - Suggested that D retention **increases as the square root of time (~$t^{0.5}$)**



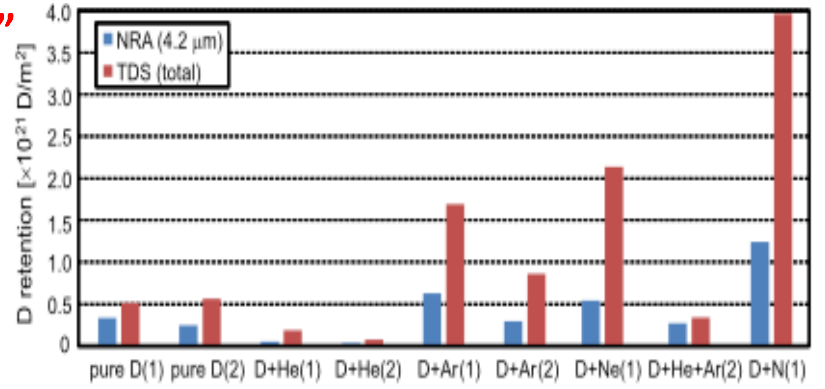
Ref: R. Doerner et al,, *NME* **9** (2016) 89

2) Effect of neutron induced radiation damage

- Tritium migrates into bulk PFC and can be trapped in radiation damages created by 14MeV neutrons
- Trapping effects in radiation damage plays a major role in tritium transport in tungsten due to low tritium solubility
- Concern for "**deep T migration and trapping in neutron-irradiated tungsten**"

3) Effect of impurities (e.g. He, N, Ar etc.)

- **Formation of He bubble layers reduced D retention**
  - 5% He-D plasma **reduces** D retention to detection limit of 10^{18} m^{-2} [2]
  - (1-5%)He - D plasma **reduces** D retention by 2 order of magnitudes [3]
- **N and Ar impurities can lead to higher D retention**
  - Nitrogen enriched layer, in contrast, serves as a desorption barrier for D atoms and **increases** D retention [4]



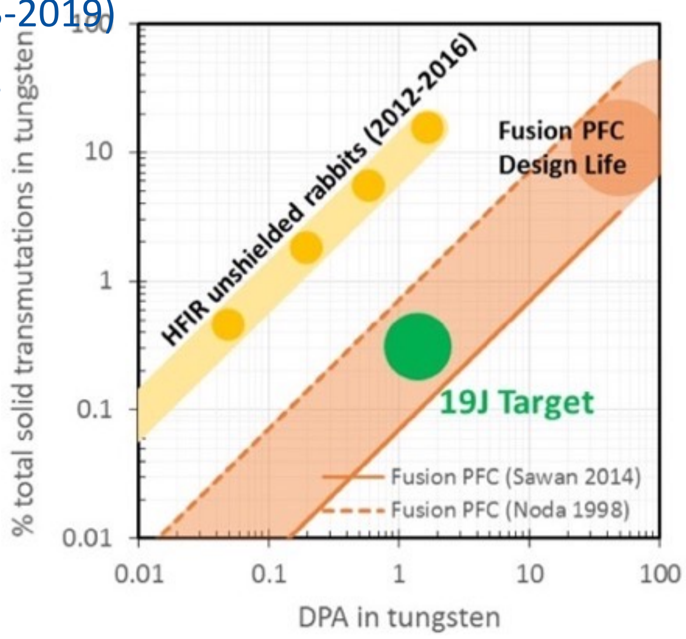
Ref: A. Kreter et al,, *NF* **59** (2019) 086029

Reference:
[1] B. Lipshultz et al., MIT report (2010) PSFC/RR-10-4,
[2] R. Doerner et al,, *NME* **9** (2016) 89
[3] Y. Nobuta et al., Fus. Sci. Technol. 77 (2021) 76
[4] A. Kreter et al., Nucl. Fusion 59 (2019) 086029

Research approach: tritium retention with neutron irradiated PFC

- **Leverages US-Japan collaboration to perform neutron irradiation**
  - Fission neuron is suitted to study irradiation effects in tungsten
    - Tungsten has low He production rate (~0.1 appm/dpa) & activation products has short half-life, $t_{1/2}$= (70-80) days
  - Use of available fission reactor (e.g. High Flux Isotope Reactor - HFIR).
    - Relatively high-flux fast neutron (>0.1 MeV) available to simulate neutron damage with similar dpa rate (10^{-7} dpa/s) to fusion
    - High-flux thermal neutron accelerates solid transmutation and increases activation
  - Use of thermal neutron-shielding (e.g. cadmium or gadolinium)
    - To minimize thermal neutron and simulate fast fusion neutron spectrum.
    - Successful thermal neutron shielded irradiation (RB-19J) carried out under PHENIX (2013-2019)
  - Use of linear plasma device to study sequential neutron-plasma irradiation.
    - TPE can handle tritium and neutron-irradiated material.
- **Experimental approach with small samples (6mm OD 0.5mm thick discs)**
  - Neutron-irradiation at HFIR, ORNL
  - D/He plasma exposure at TPE, INL
  - D/He thermal desorption spectroscopy,
  - Nuclear Reaction Analysis for D depth profile
- **Modeling approach**
  - (Low fidelity) 1D reaction-diffusion modeling with TMAP (Tritium Migration Analysis Program)
  - Simulate T implantation and desorption behavior in tungsten to derive E_{bin} and n_{trap}

Ref: Y. Katoh et al, PHENIX SCM



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Neutron irradiation at HFIR

- **Experimental condition in HFIR, ORNL**
  - Out-of-pile (rabbit) irradiation or in-pile (e.g. RB*) irradiation with inert gas
  - Displacement damage rate (~10^{-7} dpa/s) in fission, similar in fusion reactor
    - 1 dpa/year (at RB*) and up to 5 dpa/year (at flux trap) in HFIR
    - ~24.5 days/cycle, ~6 cycles/year in HFIR
- **Challenges in fission reactor irradiation**
  - Accelerated solid transmutation rate in mixed spectrum reactor (e.g. HFIR)
    - Higher solid transmutation rate [1]
      - Re production rate of ~8.4% in first 1 dpa in HFIR flux trap [1]
      - Re production rate of ~1.1% in first 2 dpa in W PFC divertor [2]
    - Effect of solid transmutation (Re and Os) in H retention
      - Re is known to reduce H retention in W, might be underestimating H retention
      - Changes in elemental composition affects thermo-mechanical properties and H transport properties (diffusivity and solubility)
  - Potential surface contamination and effect on H diffusion
    - During handling materials at hot cell
    - Inner-diffusion of surrounding material at elevated temperature at long irradiation
  - Sequential irradiation (e.g. neutron irradiation ➔ plasma exposure)

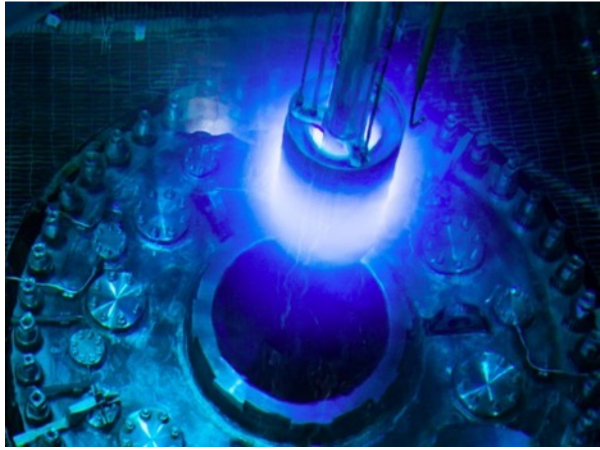
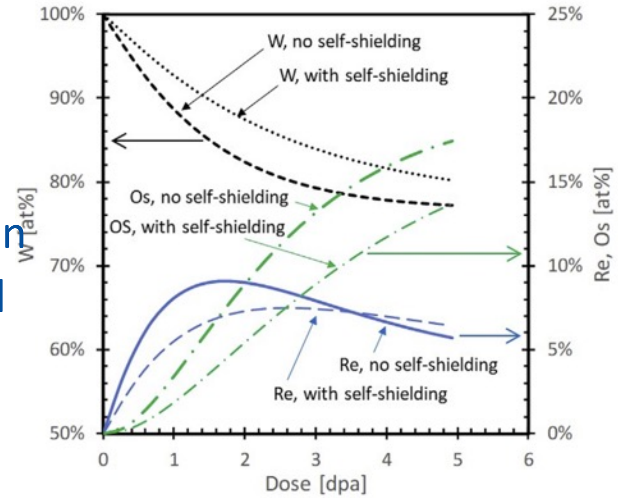


Table 1
Solid transmutations rates in tungsten (W) during fusion reactor service or HFIR irradiation experiments. Note that the transmutation rates are not constant over time but are shown as average values over the period indicated.

	W FW DCLL Blanket [26]	W PFC Divertor [27]	W in HFIR Flux Trap
Period	First 20 dpa	First 2 FPY	First 1 dpa
Hf	2 ppm/dpa		0
Ta	200 ppm/dpa	0.1–0.2%	0
Re	470 ppm/dpa	0.3–1.1%	8.4%/dpa
Os	14 ppm/dpa		3.2%/dpa
Total	690 ppm/dpa		11.6%/dpa



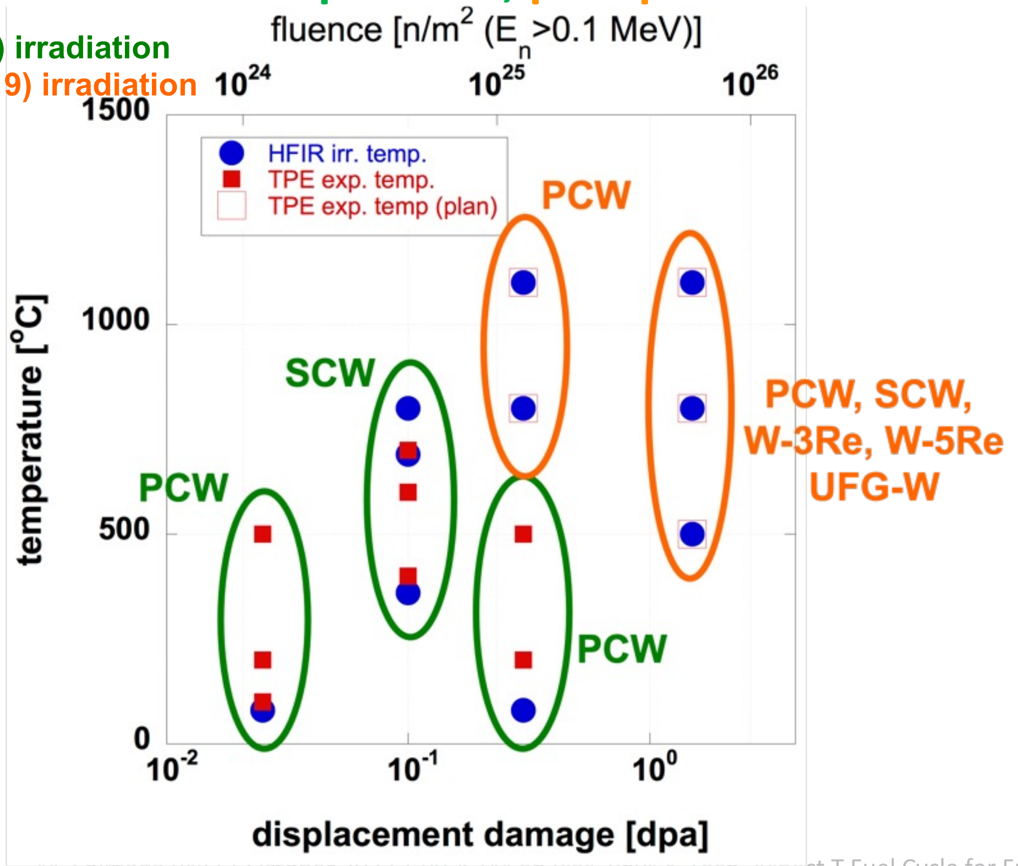
Ref: Y. Katoh et al,, *JNM* **520** (2019) 193

Reference: [1] Y. Katoh et al,, *J. Nucl. Mater.* **520** (2019) 193, [2] M.R. Gilbert et al., *Nucl. Fusion* **57** (2017) 1

Materials and irradiation condition of < 1 dpa neutron-irradiated tungsten

- **Material and heat treatment**
  - PCW: Polycrystalline, Stress-relieved (1 h@900 °C in H_2 + 0.5 h@900 °C in vacuum) 99.99 wt.% (A.L.M.T., Japan)
  - SCW: Single-crystal [110] (0.5 h@900 °C in vacuum) 99.99 wt.%
  - W-3Re: 3%Re/W (prepared by Tohoku U.) and W-5Re: 5%Re/W (prepared by U. of Toyama)
  - UFG-W: Ultra fine grained (1.1% TiC)
- **Microstructure: loop & void, precipitate & void**

TITAN (2007-2013) irradiation
PHENIX (2013-2019) irradiation



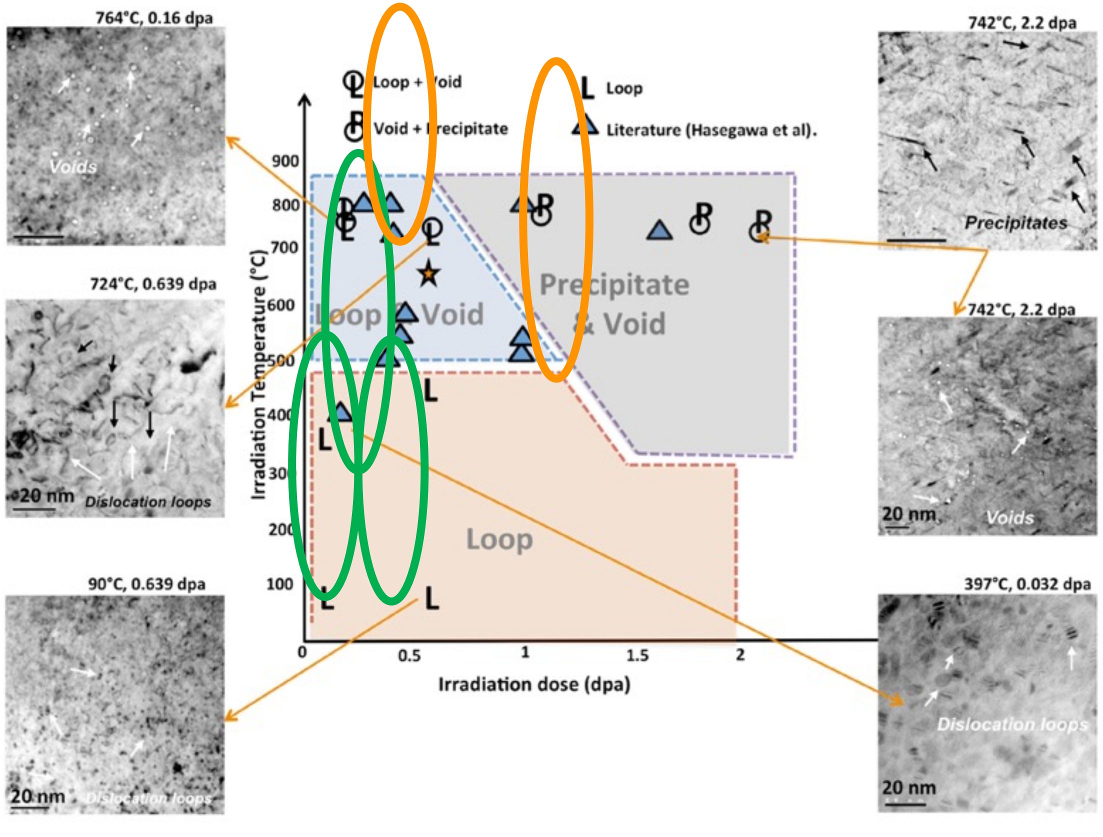
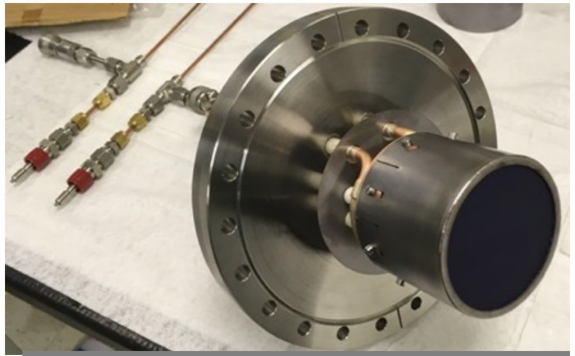


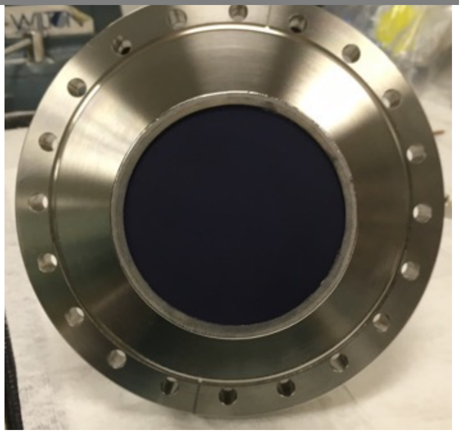
Figure 9. Irradiation phase diagram corresponding to neutron irradiated W in HFIR [90].

Ref.
J. Marian et al., *Nucl. Fusion* **57** (2017) 092008
X. Hu et al., J. Nucl. Mater. **480** (2016) 235
A. Hasegawa et al,, *Fus. Eng. Des.* **89** (2014) 1568

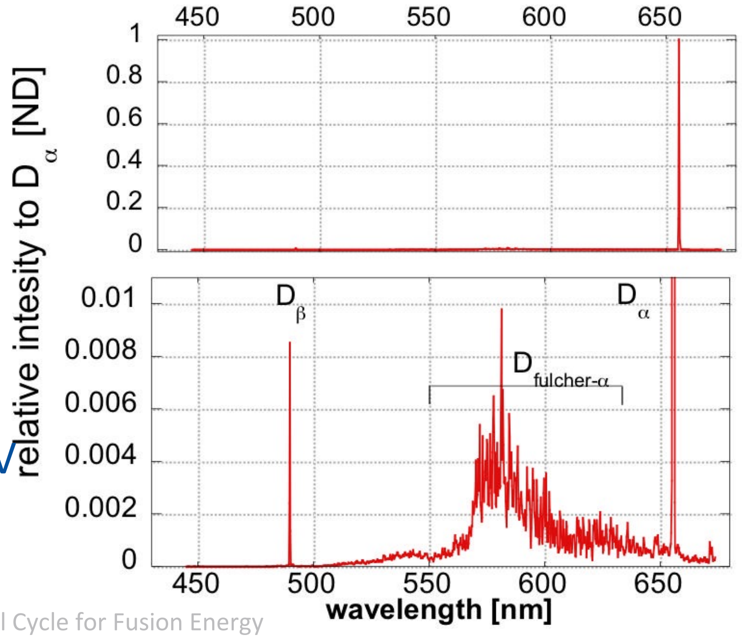
Plasma exposure at Tritium Plasma Experiment (TPE), INL

- TPE is located at Safety and Tritium Applied Research (STAR) facility, INL
  - STAR facility is (DOE less than Hazard Category 3) radiological facility
  - **Tritium handling** up to 1.6 gram (15,390 Ci)
  - **Radioactive material handling** up to < 1 mSv/hr (100 mrem/hr) at 30 cm
  - Use of a inert-gas glovebox and/or ventilated enclosure for T and rad. containment
- TPE is a unique linear plasma device in the followings:
  - **Tritium plasma (< 500 Ci per discharge),**
  - Divertor-relevant high-flux plasma (>10^{22} $m^{-2}s^{-1}$) ,
  - **Moderately radioactive (< 1 mSr/hr @ 30 cm) materials**
- **Installation of new plasma source (2017)**
  - New design based on UCSD PISCES-A source
    - Minimized impurities (e.g. C) in plasma
    - Increased plasma performance and availability
- **Typical D plasma conditions for this study**
  - T_e = 6-8 eV
  - n_e = (0.5-1.2)x10^{18} m^{-3}
  - Γ_i = (0.5-1.2)x10^{22} m^{-3}
  - E_{D+} = (95-100) eV by biasing the sample to -100V





New plasma source



Optical Spectroscopy

Outline

- Motivation
- Experimental apparatus
- **Tritium migration and trapping in neutron-irradiated tungsten**
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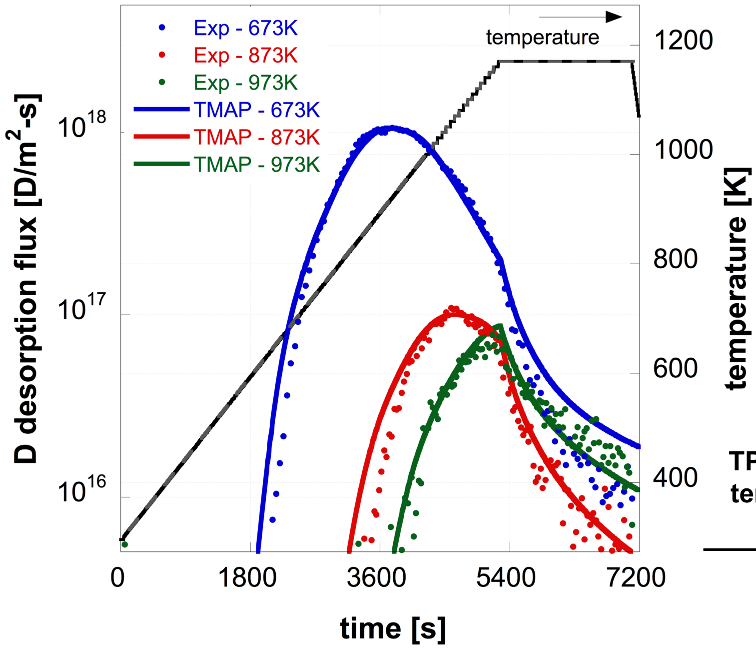
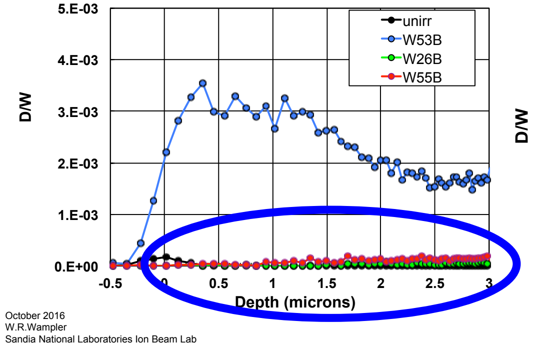
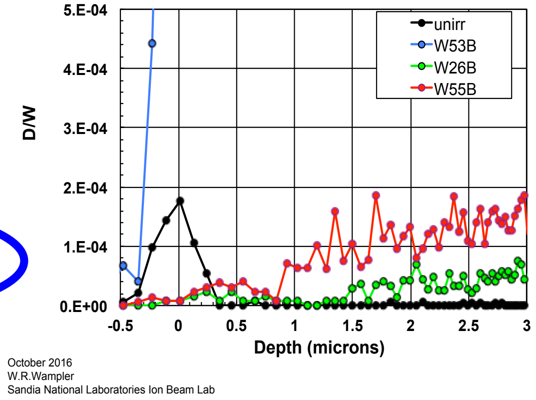
TMAP simulation on thermal desorption from 0.1dpa SCW

- **2 samples (A and B) were exposed to similar ion fluence of 5.0 x 10^{25} D m^{-2} at 3 temp.**
  - A-samples (TPE-TDS study) :TDS (10 K/min to 1173K) were performed at INL within 24 hours from D plasma exposure
  - B-samples (TPE-NRA study): NRA (up to 3 micron) were performed at SNL-NM within 30 days from D plasma exposure
- **Simulate with the assumption of uniform trap concentration with NRA measured D/W ratio**
- **Similar incident flux reduction factor (10^{-1} - 10^{-2}) is required to fit experimental TDS spectrum**
  - PISCES study uses 10^{-4} for incident flux of 10^{22} $m^{-2}s^{-1}$ ➔ Model overpredicts T retention (x10-10^4) without this factor.

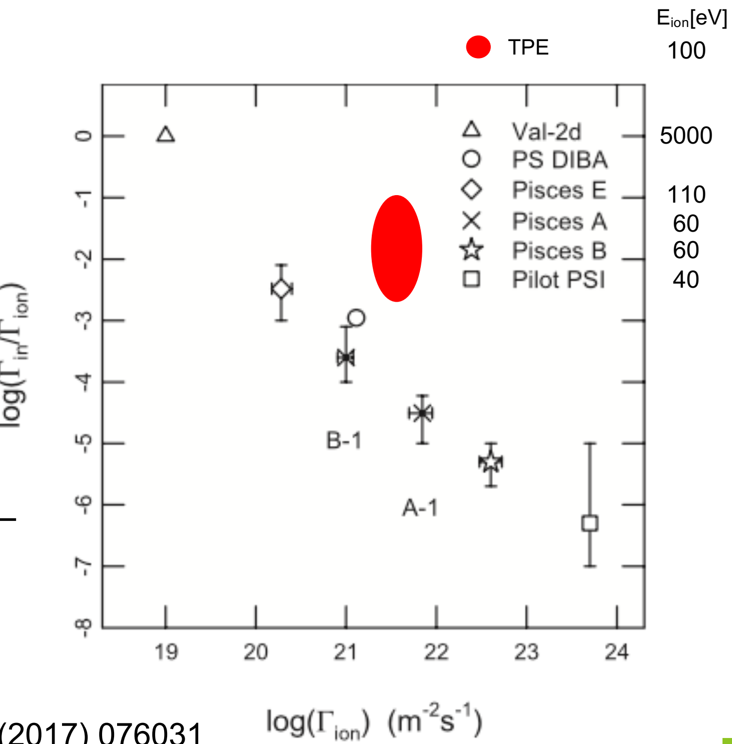






TPE exp. temp.[K]	exp. D. conc. at 3E-6 m [D/W]	reflection coefficien: R [ND]	detraping energy: E_{det} [eV]	Trap conc.: n_{trap} [T/W]	sim. D. conc. at > 3E-6 m [D/W]
673	~2.0E-03	0.90	1.80	2.0E-03	2.0E-03
873	~ 2.0E-04	0.99	2.30	2.0E-04	2.0E-04
973	~ 1.0E-04	0.99	2.60	2.0E-04	2.0E-04

Ref: M. Shimada et al,, *Fus. Eng. Des.* **136** (2018) 1161



Ref: M. Baldwin et al,, *Nucl. Fusion* **57** (2017) 076031

High binding energy in neutron-induced radiation damage

- **Definitions**
  - Detrapping energy: E_{det} [eV] = E_{bin} + E_d
  - Biding energy: E_{bin} [eV]
  - Activation energy for H diffusion in W: E_d [eV]
    - E_d = 0.39 eV

- **O.V. Ogorodnikova, J. Appl. Phys. 2015**
  - Dislocations: $0.4 \le E_{bin}$ [eV] ≤ 0.8
  - Vacancy: $0.8 \le E_{bin}$ [eV] ≤ 1.4
  - Vacancy cluster: $1.4 \le E_{bin}$ [eV] ≤ 2.2

Sample ID	TPE exp. temp.[K]	detraping energy: E_{det} [eV]	binding energy: E_{bin} [eV]	possible trap site
W53 (0.1dpa)	673	1.80	1.41	Vacancy and/or vacancy cluster
W55 (0.1dpa)	873	2.30	1.91	vacancy cluster
W26 (0.1dpa)	973	2.60	2.21	vacancy cluster ?

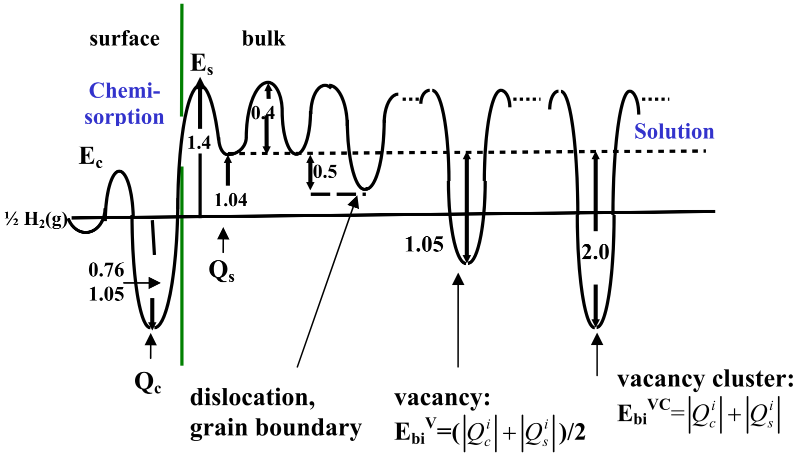


FIG. 6. Schematic potential energy diagram for hydrogen chemisorption, solution, and trapping at different trapping sites.

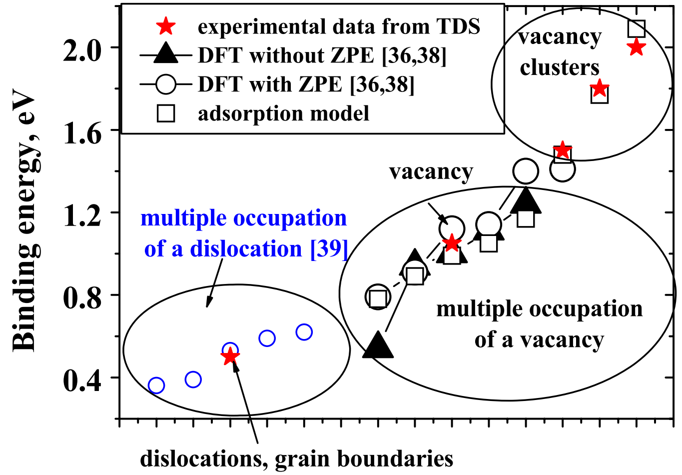


FIG. 10. Fine spectrum of H binding energy in polycrystalline W according to DFT calculations without and with ZPE (Refs. 36, 38, and 39) and according to "adsorption model" in comparison with the rate equation modelling of experimental TDS data in assumption of single binding energy of deuterium with each defect.[1,4,12–14]

Ref: O.V. Ogorodnikova, *J. Appl. Phys.* **118** (2015) 074902

Deep tritium migration and trapping

- **Deep tritium migration depth can be calculated the difference between TDS and NRA**
  - (0.025-0.3) dpa polycrystalline tungsten at 773K in ion fluence of 10^{26} m^{-2} [1]
    - T migration depth: **50 µm** for 0.025 dpa and 35 µm for 0.3 dpa
  - 0.1 dpa single crystal tungsten in ion fluence of 5.0×10^{25} m^{-2}[2]
    - T migration depth: 15 µm at 673K, 51 µm for 873K, and **170 µm** at 973K

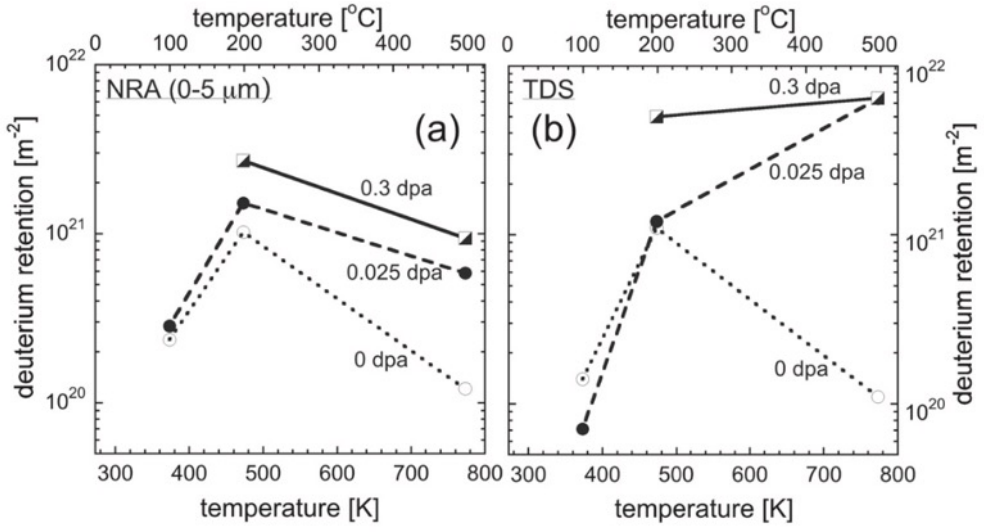
References:

[1] M. Shimada et al., *Nucl. Fusion* **55** (2015) 013008

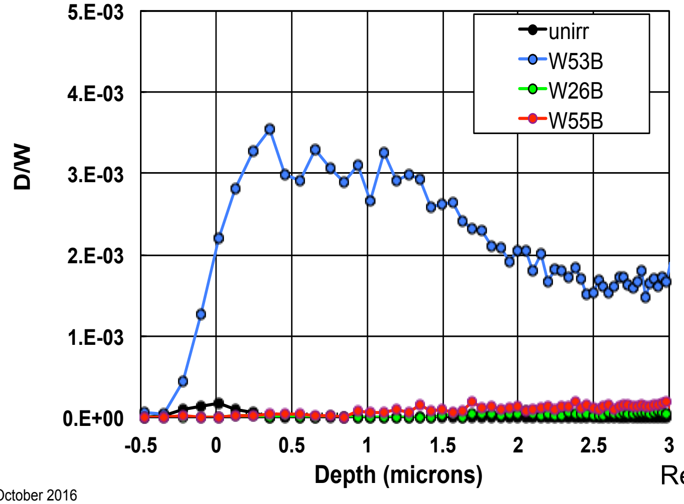
[2] M. Shimada et al., *Fus. Eng. Des.* **136** (2018) 1161

[3] M. Yajima et al., *Phys. Scr.* **96** (2021) 124042

- **Deep tritium migration was measured up to 11 µm by IPP-Garching**
  - 0.016 dpa polycrystalline tungsten at 563K in ion fluence of 1.0×10^{25} m^{-2} [3]
    - T migration depth: 17 µm at 573K, and **32 µm** at 773K
- **T migration depth increases with increasing plasma exposure temperature**

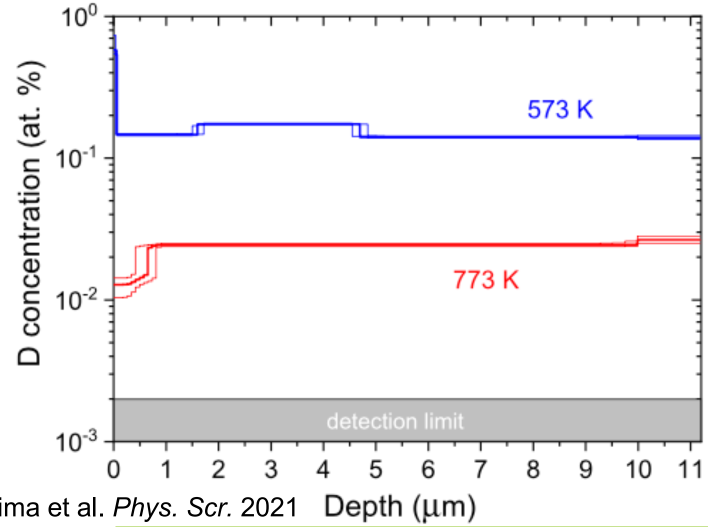


Ref: M. Shimada et al. *Nucl.Fusion* 2015



October 2016
W.R.Wampler
Sandia National Laboratories Ion Beam Lab

Ref: NRA by SNL-NM



Ref: M. Yajima et al. *Phys. Scr.* 2021

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Limited data available with neutron irradiated W for D ion fluence above 10^{26} m^{-2}

- **Incident ion fluence dependence in pure D_2 plasma around 500K**
  - D retention in (0.016-0.3) dpa neutron-irradiated W follows the upper bound trend from MIT report [1-7]
  - Limited data available with neutron irradiated W for high ion fluence above 10^{26} m^{-2}
    - Highest incident D ion fluence reported is 1.2×10^{26} m^{-2} in TPE study [2]
    - Highest incident D ion fluence reported is 1.3×10^{26} m^{-2} in CDPS (Compact Divertor Plasma Simulator) study [4]
- **CDPS study suggests the increase with square root of time [5]**
    - Measured D retention at 563K from 8×10^{24} to 1.3×10^{26} m^{-2} ion fluence
    - Suggested that D retention **increases as the square root of time ($\sim t^{0.5}$)**
- **Observations**
  - Even low dose (<0.3 dpa) radiation damage increases D retention
  - D retention may increase with $t^{0.5}$, but more data is needed.
- **Future work**
  - Incident D ion fluence dependence above 10^{26} m^{-2}
  - Explore high radiation damage (>> 1 dpa)

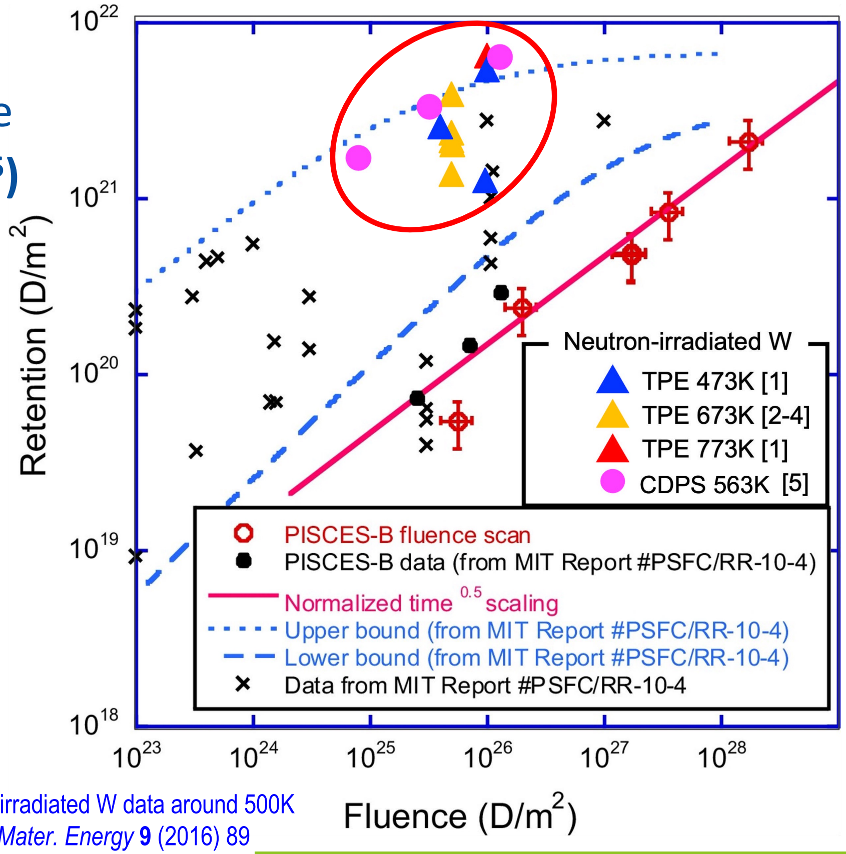


Figure: Overlay of neutron-irradiated W data around 500K on R. Doerner et al., *Nucl. Mater. Energy* **9** (2016) 89

References:

[1] M. Shimada et al., *Nucl. Fusion* **55** (2015) 013008

[2] M. Shimada et al., *Fus. Eng. Des.* **136** (2018) 1161

[3] M. Kobayashi et al., *Fus. Eng. Des.* **146** (2019) 1624

[4] Y. Nobuta et al., *Fus. Sci. Technol.* **77** (2021) 76

[5] M. Yajima et al., *Nucl. Mater. Energy* **21** (2019) 100699

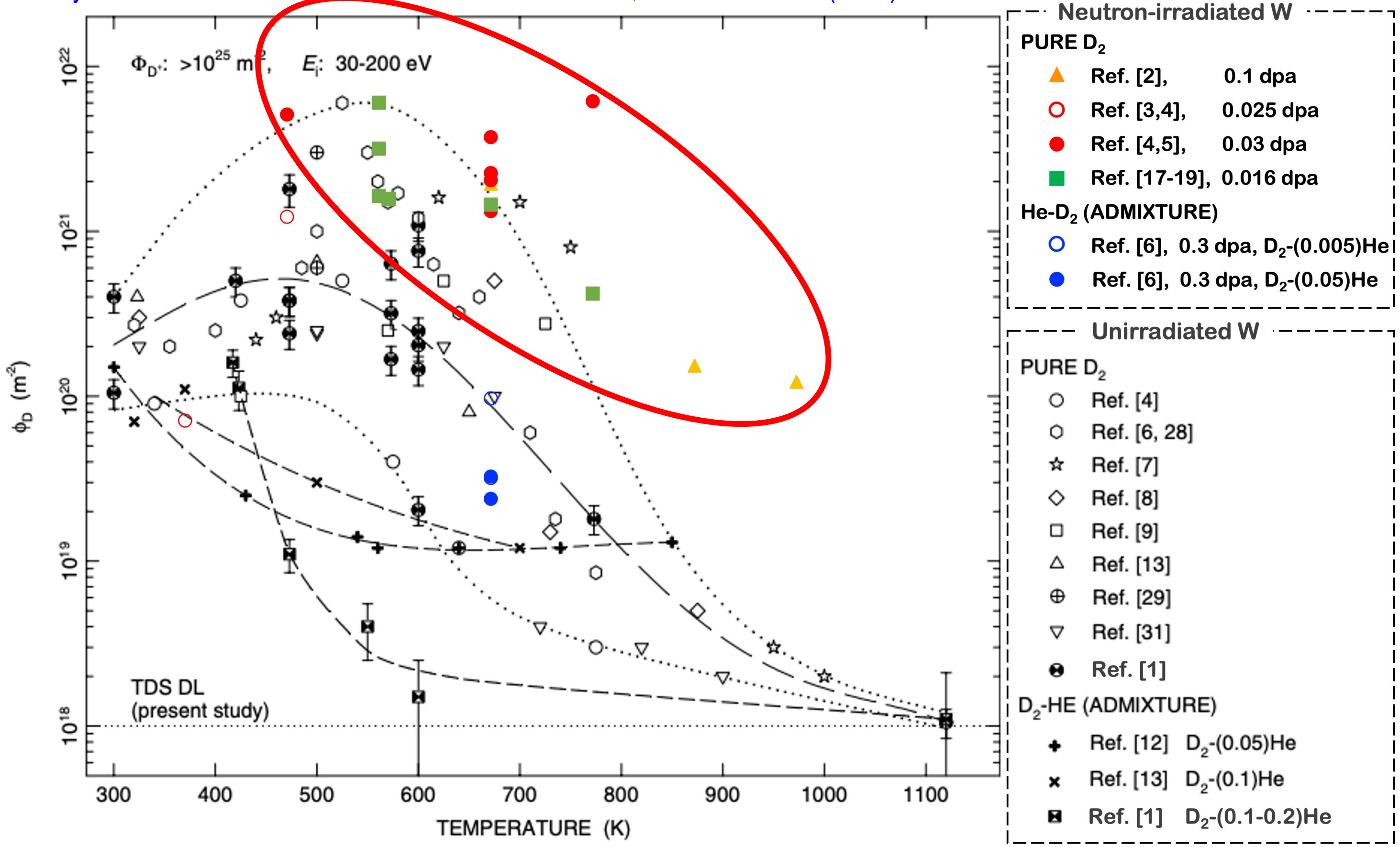
[6] R. Doerner et al., Nucl. Mater. Energy 9 (2016) 89

[7] B. Lipshultz et al., 2010 MIT repport PSFC/RR-10-4

D retention at elevated temperature > 773K due to radiation damages

- **Temperature dependence in pure D_2 and D-He plasma in the fluence of $(0.5\text{-}1.2)\times10^{26}m^{-2}$**
  - D retention in (0.016-0.3)dpa n-W is x(10-100) higher <u>at elevated temperature of >773K</u>
  - Due to high detrapping energy **(>1.8eV)** associated with radiation induced trapping site
- **Mixed D_2-(1-5%)He plasma reduces D retention by 2 order of magnitudes in n-W at 673K**
  - Formation of He bubble layers reduced D retention

Figure: Overlay of neutron-irradiated W data on M. Baldwin et al., *Nucl. Fusion* **51** (2011) 103021



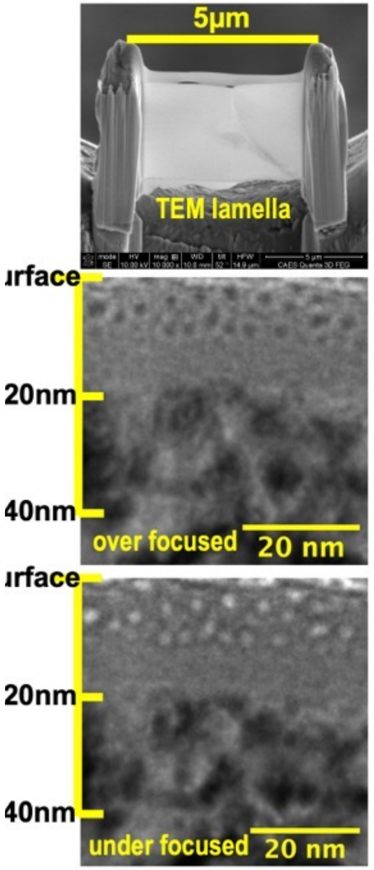


Figure: TEM of 0.3 dpa D_2-(0.05)He [6]

Reference:

[1] M. Baldwin et al., *Nucl. Fusion* **51** (2011) 103021
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[13] O.V. Ogorodnikova et al., *J. Appl. Phys.* **109** (2011) 013309
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[15] G.-N. Luo et al., *Fus. Eng. Des.* **81** (2006) 957
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[17] M. Yajima et al., *Nucl. Mater. Energy* **21** (2019) 100699
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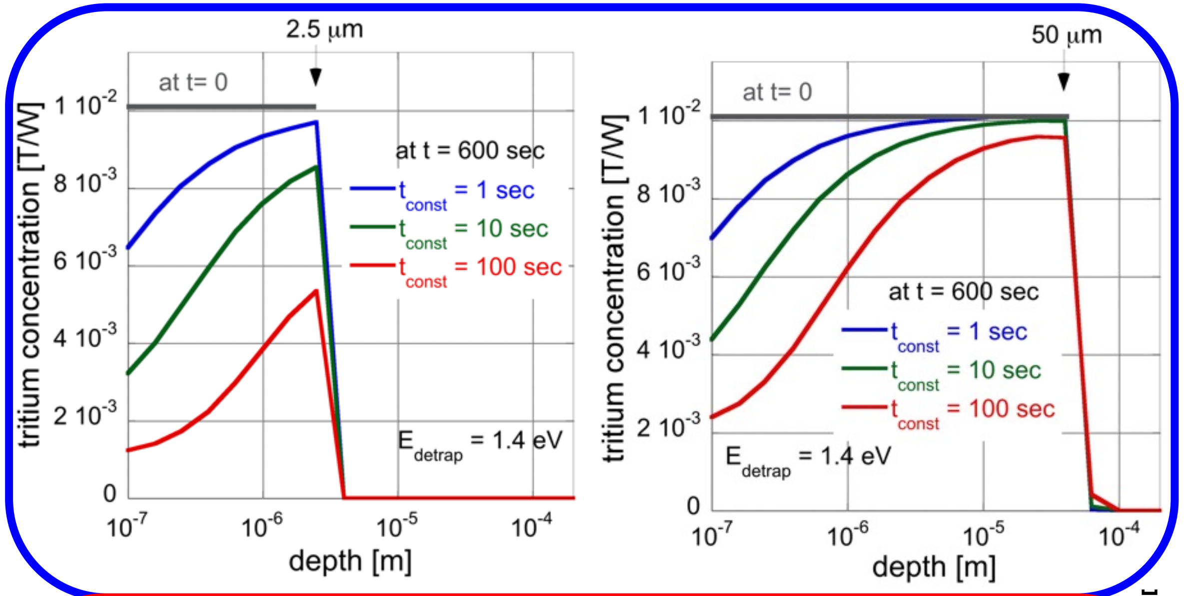
Mechanism of discrepancy in T retention at elevated temperature

1. Cooling rate

- **Effect of cooling down**

TMAP at E_{detrap}=1.4 eV

ion-damaged (2.5 μm) **neutron-irradiated (>50 μm)**



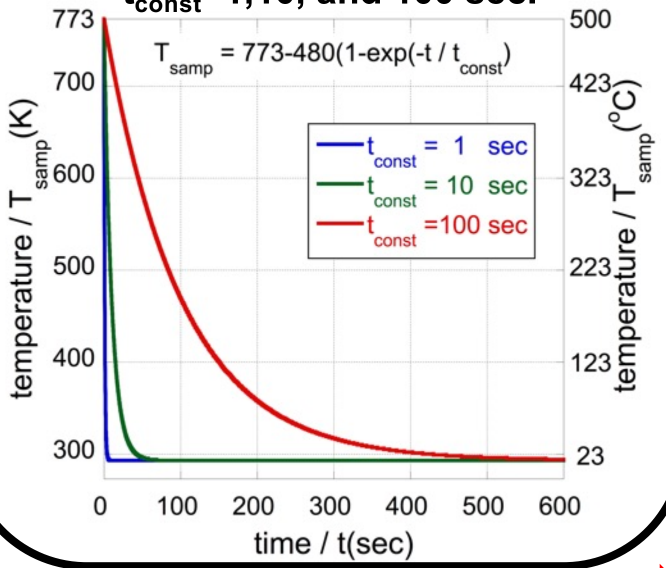
Temperature profile:

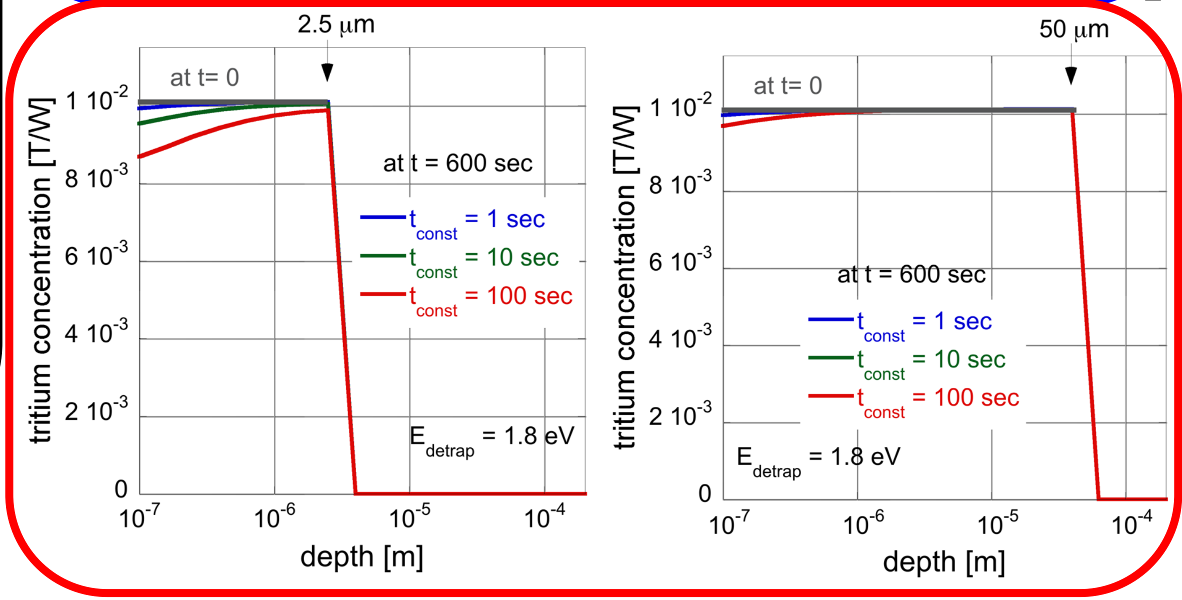
$$T(t) = T_{initial} - (T_{initial} - T_{final})^* [1-\exp(-t/t_{const})]$$

$T_{initial}$ = 773 K, T_{final} = 300 K

t_{const}: time constant

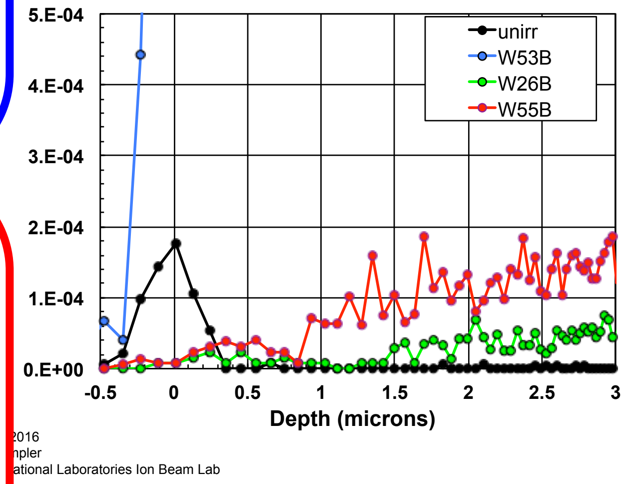
t_{const}=1,10, and 100 sec.





TMAP at E_{detrap}=1.8 eV

D depth profile from 0.1 dpa SCW



Outline

- Motivation
- Experimental apparatus
- Tritium migration and trapping in neutron-irradiated tungsten
- Recent progress on tritium retention with neutron-irradiated tungsten
- **Summary**

Summary of talk

- **Deep tritium migration and trapping was experimentally measured**
  - INL's previous studies had indicated the possibility of deep (>>10 µm) tritium migration in bulk W
  - Recently, deep tritium migration was measured up to 11 µm by NRA with 0.016 dpa polycrystalline W
- **Tritium retention studies with neutron-irradiated tungsten shows that:**
  - T retention can remain relatively high at elevated temperature > 773K due to high binding energy (E_{bin} > 1.4eV) associated with neutron-induced radiation damages and deep T migration at elevated temp.
  - Only limited data is currently available with neutron irradiated W for high ion fluence above 10^{26} m^{-2}
- **Future work with neutron-irradiated PFC will focus on:**
  - Effect of impurities (e.g. He, N, Ar, O etc.) on surface chemistry and tritium migration and trapping
  - Incident D ion fluence dependence above 10^{26} m^{-2}
  - Explore high radiation damage (>> 1 dpa)
  - Plan to restart D/He plasma experiment with RB* neutron-irradiated W and W alloys in January 2013
    - We received ~ 300 neutron-irradiated samples from HFIR, ORNL, will have >30 samples from IAEA CRP.
    - Dose rate from activated products reduced to << 10mrem/hr (0.1 Sv/hr) at 30cm
- **Implications for future (all tungsten) plasma chamber PMI in DEMO and FPP**
  - Higher radiation damage in blanket FW than that in divertor due to closer to the plasma core
  - Synergetic effect with CX neutrals poses additional concerns for T retention