First Research Coordination Meeting (RCM) of the Coordinated Research Project (CRP) on "Plasma-Wall Interaction with Irradiated Tungsten and Tungsten Alloys in Fusion Devices"

Tritium behavior in neutron-irradiated tungsten using TPE divertor plasma simulator and TMAP mass transport code

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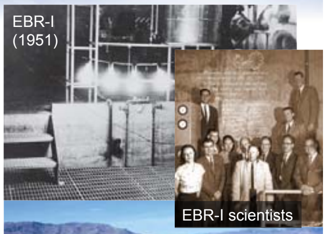
Idaho National Laboratory (INL) and fusion safety

- INL overview:
  - Geographically, the largest lab in 10 multi-program US national laboratories
  - 52 reactors were designed and built in Idaho
  - The world's first usable electricity from nuclear energy generated in EBR-I in 1951
  - The nation's lead laboratory for nuclear energy research and development in the U.S.
- Advanced Test Reactor (ATR)
  - Light water moderated/cooled PWR with Be neutron reflector
  - Max: 250 MW_{th}, "Four Leaf Clover" design, and 120 cm axial length
  - Materials and fuels testing, isotope production (e.g. ^{60}Co)
- Safety and Tritium Applied Research (STAR) Facility
  - Fusion safety and tritium research
  - less than HC3

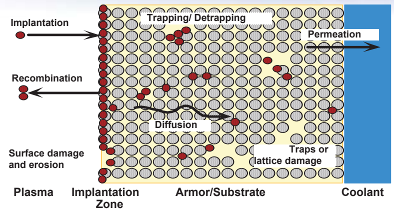


Outlines

1. Motivation for this IAEA CRP (F43021)
   - Motivation
   - INL's proposed research
2. Unique capabilities of Fusion Safety Program, INL
   - Tritium Plasma Experiment (TPE) divertor plasma simulator
   - Tritium Migration Analysis Program (TMAP) mass transport code
   - HFIR neutron-irradiation under US-Japan PHENIX program
   - Other capabilities (NIMIIX and TGAP)
3. Research highlights in "PSI with irradiated tungsten"
   - HFIR irradiation tungsten under US-Japan TITAN program
4. Summary

Safety concern: "in-vessel inventory source term"

Reference: ITER GSSR 2004

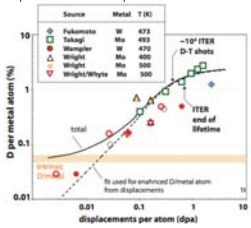


- Challenges in neutron-irradiated plasma facing components (PFCs)
  - The PFCs will subject to intense fusion fast neutrons to 0.7 dpa for W divertor in ITER and > 10 dpa in DEMO/future reactor
  - Radiation damages (vacancy, vacancy-cluster, void etc.) will be created by 14 MeV throughout PFCs thickness, becoming trapping site for tritium
  - Large amount of tritium can be trapped in vacancy-cluster as gas form, leading to bubble formation, and blister formation in metal
  - Tritium behavior in the fusion nuclear environment is not fully understood

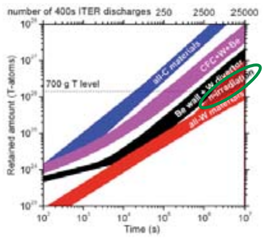
➔ There exists large uncertainty in tritium retention assessment in neutron-irradiated PFCs

Assessment of T retention in neutron-irradiated W

Reference: B.Lipschultz et. al. MIT report 2010



Reference: J. Roth et. al. PPCF 2008



- High energy ion beams have been used to simulate displacement damages by 14 MeV fusion neutron, and recent studies provided us three trends/assumptions:
  1. The trap concentration will most likely saturate at > 1 dpa
  2. Tritium will most likely stay with in a few micro meters from the surface
  3. Small tritium retention from damaged W at high (> 500C) temperature
- If the above three trends/assumptions are true (?), tritium retention in W will be small.

INL's proposed research for this CRP (F43021)

- Title: "Tritium behavior in neutron-irradiated tungsten using TPE divertor plasma simulator and TMAP mass transport code"
- Objectives: to investigate tritium behavior (retention as well as gas-driven and plasma-driven permeation) in neutron-irradiated tungsten utilizing unique capabilities of the INL:
  - Tritium Plasma Experiment (TPE) divertor plasma simulator
  - Tritium Migration Analysis Program (TMAP) mass transport code.
- Under US-Japan PHENIX collaboration, ITER grade tungsten will be irradiated with neutrons at high temperature (300, 600, 1000, and 1200 °C) up to 1.5 dpa at High Flux Isotope Reactor (HFIR) with thermal neutron shielding to better represent the fusion neutron energy spectrum

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   - Motivation
   - INL's proposed research
2. Unique capabilities of Fusion Safety Program, INL
   - Tritium Plasma Experiment (TPE) divertor plasma simulator
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   - HFIR neutron-irradiation under US-Japan PHENIX program
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Tritium Plasma Experiment - TPE

Unique capabilities

- TPE is contained within double enclosure (PermaCon Box and Glovebox)
- TPE is unique in that it combines four specialized elements:
  - (a) the ability to handle tritium (max. T inventory: < 1.5g in STAR)
  - (b) a divertor-relevant high-flux plasma (max. ion flux: $4.0 \times 10^{22}\ m^{-2}s^{-1}$)
  - (c) the ability to handle radioactive materials (STAR limit: < 100 mR/hr = 10 μSv/hr)
  - (d) the ability to handle beryllium
- Plasma-driven tritium permeation capability (under development)

Comparison of plasma parameters among Existing and proposed US Linear Plasma Devices

	PISCES-B (UCSD)	TPE (INL)	MPEX (ORNL)
Deuterium ion flux: Γ_i ($m^{-2}s^{-1}$)	10^{21}–10^{23}	10^{20} – 3.7×10^{22}	$>10^{23}$
Incident ion energy: E_i (eV)	20–300 (bias)	50–200 (bias)	??
Electron temperature: T_e (eV)	4 – 40	5 – 20	3-50
Ion temperature: T_i (eV)	2 – 5	2 - 5	1 - 200
Electron density: n_e (m^{-3})	10^{18}–10^{19}	10^{16} – 3.5×10^{18}	10^{18}-3×10^{19}
Max. heat flux: P_{max} (MW/m²)	5	~1.2	20
Plasma diameter (mm)	75	50	120
Max. specimen size	ϕ ~ 25.4 mm disc	ϕ ~ 50.8 mm disc	100 x 100 mm plate
Pulse length (s)	Steady state	Steady state	Pulse and Steady state
Activated targets	No	Yes	Yes
Tritium	No	Yes	No
Beryllium	Yes	Yes/No*	Yes
Permeation capability	No	Yes**	No
Ion incident angle	Normal	Normal***	Inclined and Normal
Plasma source (cathode)	Reflex arc (LaB_6)	Reflex arc (LaB_6)	Helicon (no cathode)
Year of operation	Since 1988	Since 1989	Proposed phase
Unique capabilities	In-situ surface analysis, transient surface heating, beryllium testing	Tritium use and diagnostics, neutron irradiated materials	Electrodeless plasma (Helicon + ECH + ICH) minimizes plasma contamination by impurity

NOTES:
*: Beryllium has been extensively tested in TPE during it tenure at TSTA, LANL in 90's, but it has not been actively tested in INL.
**: Tritium plasma-driven permeation capability is under development with the SNL/CA collaboration
***: Incident angle can be varied upon target holder design, and the current target holder is designed for normal incidence only.

Tritium Migration Analysis Program - TMAP

- The TMAP calculates the time-dependent response of a system of solid structures or walls (may be a composite layer), and a related gas filled enclosures or rooms by including
  - Movement of gaseous species through structures surfaces, governed by dissociation/recombination, or by solution law such as Sieverts' or Henry's Laws
  - Movement in the structure by Fick's-law of bulk diffusion with the possibility of specie trapping in material defects
  - Thermal response of structures to applied heat or boundary temperatures
  - Chemical reactions within the enclosures
  - User specified convective flow between enclosures
- Equations governing these phenomenon are non-linear and a Newton solver is used to converge the equation set each time-step

TMAP Capabilities

- TMAP does not treat plasma surface physics, such as sputter or sputtered material re-deposition. TMAP's basic equations are:

Surface Movement

Non-equilibrium / Equilibrium

$$J_m = K_d p_m - K_r C_m^2 \qquad C_m = K_s\sqrt{p_{m_2}}$$

Bulk Diffusion

$$\frac{\partial C_m}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial C_m}{\partial x}\right) + S_m - \sum_{i=1}^{K}\frac{\partial C_t^i}{\partial t}$$

Bulk Trapping

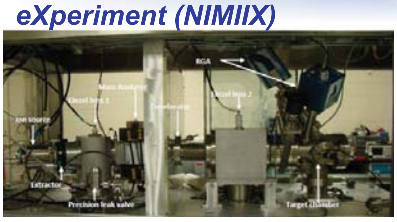
$$\frac{\partial C_t}{\partial t} = \alpha_t f_t C_m - \alpha_r C_t$$

Surface
J_m – Net specie surface flux (m^{-2}-s)
K_d – Molecular disassociation coefficient (m^{2}-s^{-1})
K_r – Atom recombination coefficient (m^{2}-s^{-1})
K_s – Sievert's solubility coefficient (m^{2}-s^{-1})
Bulk
C_m – Mobile specie concentration (m^{-3})
D – Specie diffusion coefficient (m^{2}-s^{-1})
K – Number of traps (-)
C_t – Trapped specie concentration (m^{-3})
α_t – Trapping rate coefficient (s^{-1})
f_t – Probability of landing in a trap site (-)
C_m – Mobile concentration (m^{-3})
α_r – Release rate coefficient (s^{-1})

$$\alpha_t = \frac{D}{\lambda^2};\ f_t = \frac{c_t^o - C_t}{N};\ \alpha_r = \nu_o \exp\left(-\frac{E_t}{kT}\right)$$

λ – jump distance or lattice constant (m)
c_t^o – Trap site concentration (m^{-3})
N – Bulk material atom density (m^{-3})
ν_o – Debye frequency (s^{-1})
E_t – Trap energy (eV)

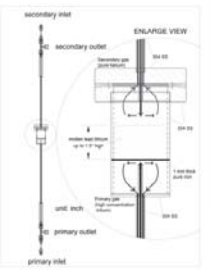
Neutron Irradiated Material Ion Implantation eXperiment (NIMIIX)

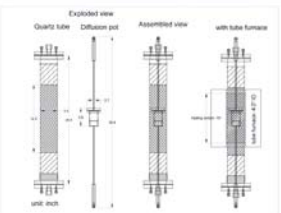


Unique capabilities

- One of very few high-energy ion implantation device that can handle neutron-irradiated materials.
- Moderate (10^{18} – 10^{20} $m^{-2}s^{-1}$) ion flux to study plasma surface interaction in the first wall.
- Specializes in **neutron-irradiated materials**

Tritium GAs Permeation - TGAP





Unique capabilities

- Designed to measure transport properties (e.g. diffusivity, solubility, and permeability) of tritium at realistic blanket conditions (e.g. low tritium partial pressure < 1000 Pa)
- Capable of testing liquid breeder material (e.g. PbLi and FLiBe) and disc shaped metal
- Uniform temperature (+/- 15 C) within the test section utilizing 12" tube furnace

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   - Motivation
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   - Expected outcomes
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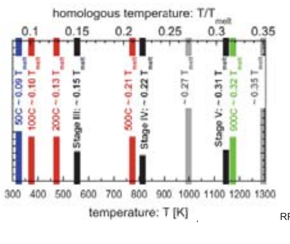
Experimental procedures

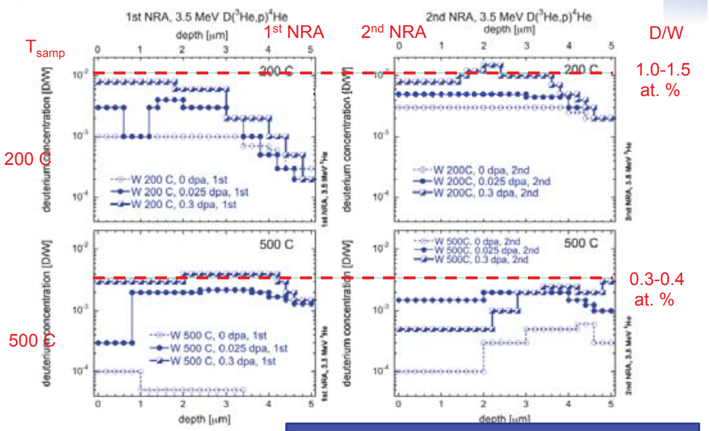
1) W rod fabrication (A.L.M.T. Co., Japan) and sample preparation (slicing to 0.2 mm, polishing, and annealing at 900 °C for 0.5 hour) at Univ. Toyama, Japan
2) Neutron Irradiation at 50-70 °C to 0.025 and 0.3 dpa at the HFIR
3) 1st D plasma exposure at 100, 200, and 500 °C to 5×10^{25} m^{-2} ion fluence at TPE
4) 1st D depth profile measurement via NRA at U. Wisconsin-Madison.
5) 2nd D plasma exposure at 100, 200, and 500 °C to 5×10^{25} m^{-2} ion fluence
   → total ion fluence to 1×10^{26} m^{-2}
6) 2nd D depth profile measurement via NRA at U. Wisconsin-Madison.
7) D retention measurement via TDS 10 C/min to 900 °C



Stage III (0.15 Tm): Vacancy migration
Stage V (0.31 Tm): Vacancy cluster migration

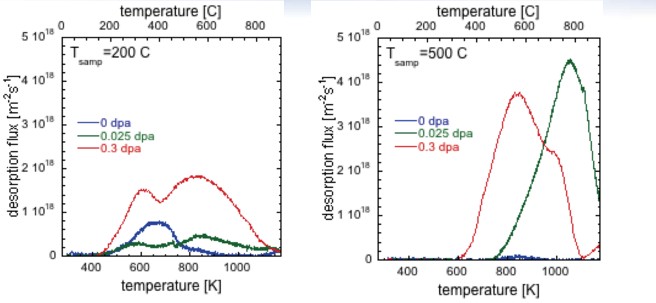
Ref: H. Schultz, Mater. Sci. Eng. '91

Deuterium depth profiles in HFIR neutron-irradiated W



D/W saturated at 1.0~1.5 at. % for 0.3 dpa W

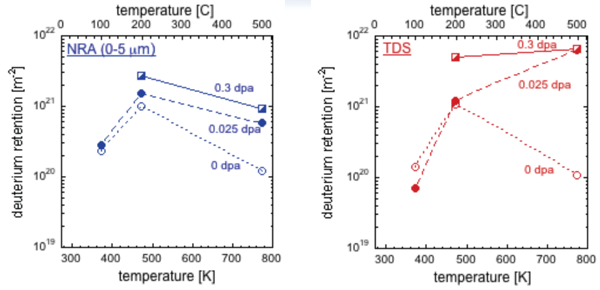
Thermal desorption spectra from HFIR neutron-irradiated W



Experimental procedure:

- **1st TPE (@INL)➔ 1st NRA (@U of Wisc.) ➔ 2nd TPE ➔ 2nd NRA ➔ final TDS**
- Flux: 5×10^{21} m^{-2}s^{-1}, Fluence: (4-5) $\times10^{25}$ m^{-2} each TPE exposure, (8-10) $\times10^{25}$ m^{-2} in total fluence
- Dose: 0, 0.025, and 0.3 dpa
- Plasma exposure temperature: 100, 200, and 500 C
- Ion energy: 100 eV

Comparison of D retention via NRA and TDS



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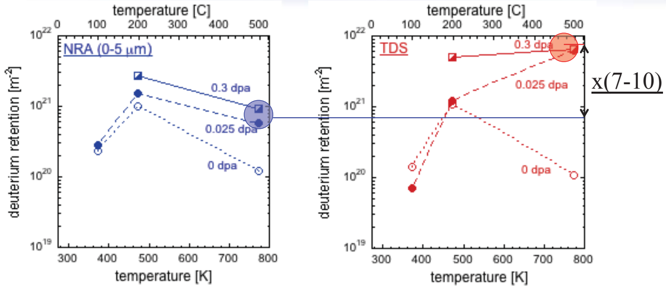
- Plasma exposure temperature: 100, 200, and 500 C
- Ion energy: 100 eV

Deep migration of deuterium into bulk (>> 10 μm)



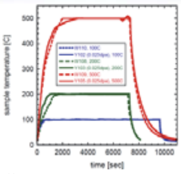
at 500 C

- ✓ NRA shows small (6-9 $\times10^{20}$ m-2) retention ⟷ TDS shows Large (6×10^{21} m-2) retention
- ✓ Discrepancy between TDS and NRA:
  - 0.025 dpa: x10 ➔ > 50μm migration
  - 0.3 dpa: x7 ➔ > 35μm migration

Deep migration of deuterium into bulk (> 35-50 μm from plasma surface) in the ion fluence of 10^{26} m^{-2} range

Application of TMAP to plasma surface interaction

Sample ID	Weight [gram]	Sample size (diameter / thickness) [mm]	HFIR irradiation dose [dpa]	TPE exposure temperature [°C]	1st TPE exposure flux [m^{-2}s^{-1}]	2nd TPE exposure flux [m^{-2}s^{-1}]	1st TPE exposure fluence [m^{-2}]	2nd TPE exposure fluence [m^{-2}]	Cumulative fluence [m^{-2}]
Y102	0.80	6.0 / 0.15	0.025	100	5.3E+21	5.3E+21	5.0E+25	5.0E+25	1.0E+26
Y103	0.89	6.0 / 0.16	0.025	200	7.0E+21	6.4E+21	5.0E+25	4.6E+25	9.7E+25
Y105	0.83	6.0 / 0.15	0.025	500	7.3E+21	9.3E+21	5.3E+25	6.7E+25	1.2E+26
Y107	0.90	6.0 / 0.17	0.3	200	1.1E+22	1.1E+22	5.0E+25	5.0E+25	1.0E+26
Y112	0.91	6.0 / 0.17	0.3	500	1.1E+22	9.0E+21	5.0E+25	5.0E+25	1.0E+26



- To apply TMAP to TPE results, property data for tungsten diffusivity, solubility, and surface recombination are required
- Physical data of trap density, D^+ implantation depth, plasma surface flux intensity and target temperatures are also required
- Frauenfelder[1] measured diffusivity and solubility and Anderl's[2] determined surface recombination rates were adopted for this study
- An implantation depth of 3 nm estimated by Wampler[3] and TPE implantation flux (5×10^{21} D^+/m^2-s), reflection coefficient of 0.5, and flux and temperature histories specified were used
- NRA provide (near surface) trap concentration
- Only fitting parameter is detrapping energy

[1] R. Frauenfelder, Vac. Sci. Tech.,6 (1969) 338.
[2] R. A. Anderl, et al., Fusion Technol., 21 (1992) 745.
[3] W.R. Wampler, Nucl. Fusion, 49 (2009) 115023 (11pp).

Uncertainties in mass transport parameters in W

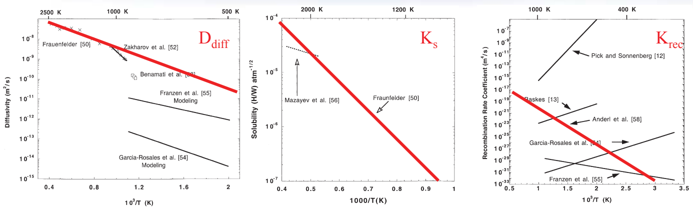


Fig. 6. Hydrogen diffusivity in tungsten.

Fig. 7. Hydrogen solubility in tungsten.

Fig. 9. Comparison of experimental and theoretical values for the hydrogen recombination rate coefficient for tungsten.

- Mass transport parameters of hydrogen in tungsten (red lines):
  - Causey's review paper recommends[1]:
    - Diffusivity (D_{diff}) and solubility (K_s) by Fraundelder[2]
    - Recombination coefficient (K_{rec}) by Anderl[3]
    - Dissociation coefficient by the assumption: $K_{dis} = K_{rec} \times (K_s)^2$
  - Recombination and dissociation coefficient have largest uncertainty and subject to the surface conditions

References:
[1] R.A. Causey, J. Nucl. Mater., 300 (2002) 91
[2] R. Frauenfelder, Vac. Sci. Tech.,6 (1969) 338.
[3] R. A. Anderl, et al., Fusion Technol., 21 (1992) 745.

Enhanced diffusion to simulate plasma implantation

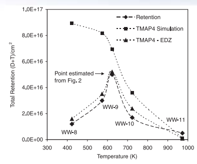
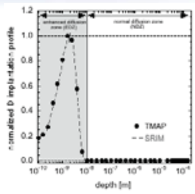


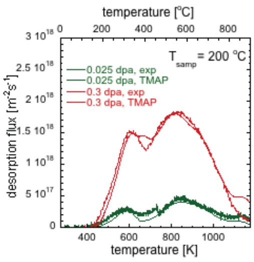
Fig. 1. Experimental retention results (◆), TMAP4 simulation using best-fit parameters (■), and TMAP4 retention results using C_s = 0 and enhanced diffusion in the implant zone (▲).



- TMAP does not treat plasma surface physics (e.g. erosion, sputtering and deposition).
- TMAP can simulate the ion implantation with ion implantation profile
- Venhaus and Causey[1,2] artificially increased hydrogen diffusivity in the Enhanced Diffusion Zone (EDZ) (<10 nm) during plasma implantation in order to reduce the mobile concentration and retention at low temperature in tungsten.
  - During plasma implantation:
    - Enhanced diffusivity (D_{diff}^{EDZ}) of (0.5-2.0)$\times10^{-9}$ m^2/s[1] matches the experiment well.
  - During thermal desorption:
    - Diffusivity (D_{diff}) by Fraundelder[4]

References:
[1] R.A. Causey, et. al. J. Nucl. Mater. 266 (1999) 467.
[2] T. Venhaus and R.A. Causey, J. Nucl. Mater. 290 (2001) 505.
[3] T. Venhaus and R.A. Causey, Fus. Technol. 39 (2001) 868.
[4] R. Frauenfelder, Vac. Sci. Tech.,6 (1969) 338.

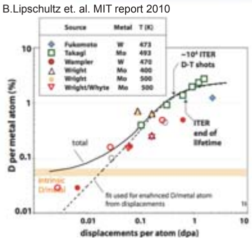
TMAP modeling in irradiated tungsten



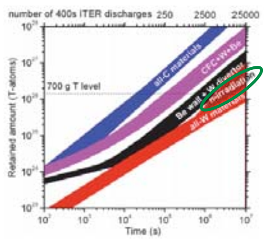
- TMAP fits the experimental data well with the following parameters for 200°C case:
  - 0.025 dpa, 200°C: E_{detrap}= 1.25 eV (5e-4 D/W) and 1.75 eV (1e-3 D/W)
  - 0.3 dpa, 200°C: E_{detrap}= 1.22/1.24 eV (2.3e-3 D/W) and 1.53/1.75 eV (2.5e-3 D/W)
- Having difficulty fitting experimental data from neutron-irradiated W at 500°C case
- Some physics needs to be understood:
  - Trap annealing with $E_{V,M}$ =1.7 eV to remove the very high temperature tail (> 800 °C)
  - Vacancy clustering, void formation, and its associated detrapping energies (E_{detrap})

Plasma/ion machine dependence at high temp. data

Reference: B.Lipschultz et. al. MIT report 2010

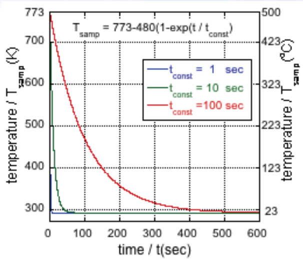


Reference: J. Roth et. al. PPCF 2008



- High energy ion beams have been used to simulate displacement damages by 14 MeV fusion neutron, and recent studies provided us three trends/assumptions:
  1. The trap concentration will most likely saturate at > 1 dpa
  2. Tritium will most likely stay with in a few micro meters from the surface
  3. Small tritium retention from damaged W at high (> 500C) temperature
- If the above three trends/assumptions are true (?), tritium retention in W will be small.

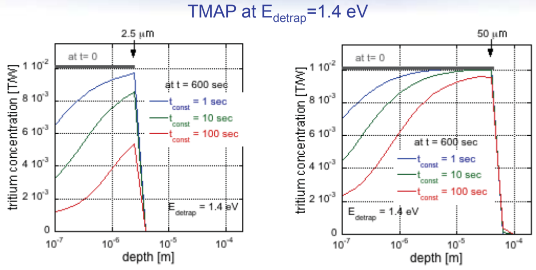
Effect of temperature profile during cooling down



- We need to know temperature profile not only during plasma/ion implantation but also during cooling down especially for high temperature case (> 500°C)
- There exists the machine dependence when comparing the D/T retention data
  - Low flux machine (< 10^{21} $m^{-2}s^{-1}$), t_{const} > 100 sec due to active heating
  - High flux machine (> 10^{22} $m^{-2}s^{-1}$), 10 < t_{const} < 100 sec due to active cooling
  - t_{const} depends on the location (divertor plate, strike point, dome) in ITER

Ion-damage W vs. neutron-irradiated W

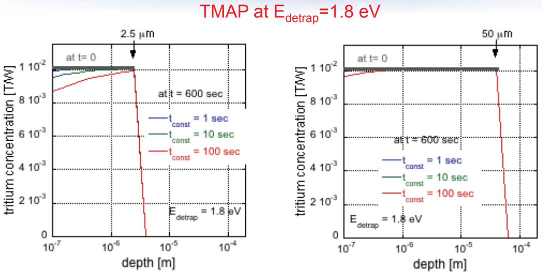
TMAP at E_{detrap}=1.4 eV



1. The trap concentration will most likely saturate at 1 at.% at > 1 dpa
2. T is trapped in ion-damaged range (2.5 μm) vs. neutron-irradiated (>50 μm)
3. T depth profile at high (> 500C) temperature:
   - For E_{detrap}=1.4 eV, Most of T can be desorbed during ramp-down if t_{const} >100 sec

Ion-damage W vs. neutron-irradiated W

TMAP at E_{detrap}=1.8 eV



1. The trap concentration will most likely saturate at 1 at.% at > 1 dpa
2. T is trapped in ion-damaged range (2.5 μm) vs. neutron-irradiated (>50 μm)
3. T depth profile at high (> 500C) temperature:
   - For E_{detrap}=1.4 eV, Most of T can be desorbed during ramp-down if t_{const} >100 sec
   - For E_{detrap}=1.8 eV, T is still trapped during ramp-down if t_{const} <100 sec

Key issues identified in TPE :

- <u>Key issues identified in TPE in the past 3 years:</u>

1. (0.1-1.5 at.% D/W) deuterium concentration even in low-moderate dose (0.025 - 0.3 dpa) HFIR neutron-irradiated tungsten
2. Deep (>> 10 μm) migration and trapping of deuterium and resulting high deuterium retention at high plasma exposure temperature (500 °C)
3. Raised the safety concern (e.g. possible tritium retention in bulk PFC) regarding the neutron-irradiated PFCs

<u>Challenges in DEMO envirionments</u>:

- W and W alloy, one of the promising candidate materials for first wall and divertor PFCs, are expected to receive dose up to >10 dpa at high temperature (> 500 °C)
- T will expected to migrate and find the trapping site in the bulk, increasing T retention

➔ We need to understand the physics of tritium behavior in irradiated W and find the mitigation/removal technique

Critical remaining issues:

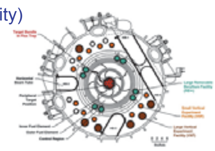
1. Radiation damage recovery temperature
   - Recovery temperature
   - Activation of defect migration
   - Defect characteristic etc.
2. Trapping mechanism(s) of tritium in radiation damage
   - Maximum trap concentration,
   - Trap concentration profile throughout material (on the order of mm),
   - De-trapping energy of tritium from trap site
   - Need to distinguish radiation damage from transmutation effects etc.
3. Diffusion mechanism(s) of tritium to <u>bulk (>10 μm) tungsten</u> under ITER/DEMO divertor relevant condition
   - High ion flux (>10^{22} $m^{-2}s^{-1}$)
   - Mixed (He, D, and T) plasma
   - High temperature (>500 C)
4. Mitigation of deep diffusion of tritium to <u>bulk (>10 μm) tungsten</u>
5. Removal of tritium from <u>bulk (>10 μm) tungsten</u>

<u>NOTE:</u>
<u>Neutron-irradiation is the best (and only) option to create high dose (> 1 dpa) radiation damage in bulk (>10 μm) W</u>

New challenges in PHENIX project (2013-2019):

Tritium behavior in realistic fusion nuclear environment

- Tritium behavior (retention and permeation):
  - Deuterium/tritium retention by high-flux TPE plasma
  - Gas-driven permeation in TGAP
  - Plasma-driven permeation in TPE
- Realistic plasma conditions in divertor:
  - High plasma exposure temperature (500-1000C)
  - Mixed (D/T/He) plasma
  - High flux (10^{22} – 10^{23} $m^{-2}s^{-1}$), high-fluence (10^{26} – 10^{27} m^{-2})
- Realistic fusion neutron irradiation
  - Larger irradiation port (removable beryllium facility)
  - HFIR irradiation with thermal neutron shielding
  - High irradiation temperature (500-1200C)
  - Deuterium gas environment



Summary:

- The FSP/INL possesses the unique capabilities (TPE, TMAP, NIMIIX, and TGAP) to study the PSI in irradiated tungsten, and will utilize them for this CRP (F43021)
- FSP/STAR continues to study the PSI in irradiated tungsten under the US-Japan PHENIX collaboration (April 2013-March 2019)
- Through this CRP, we hope to achieve the successful development of scientific understanding of tritium behavior in neutron-irradiated tungsten, which can be a major breakthrough in fusion technology and safety.
- This knowledge would minimize the uncertainty in tritium in-vessel inventory levels, and better understanding of tritium in-vessel inventories and ex-vessel releases will support licensing assessments.