Update on deuterium behaviors in neutron-irradiated tungsten

Y. Hatano, U. Toyama

on behalf of

Japan-US Collaboration TITAN Task2-1 Team,

Japan-US Collaboration PHENIX Task 3 Team,

IMR-Oarai Collaboration Team

Main contributions

Y. Oya (Shizuoka U.), T. Toyama (Tohoku U.), M. Yajima (NIFS), N. Ohno (Nagoya U.).,
T. Kuwabara (Aich Inst. Tech.), Y. Yamauchi, Y. Nobuta (Hokkaido U.)

M. Shimada, C. Taylor, P. Sharpe (INL), D. Buchenauer, R. Kolasinski, W. Wampler (SNL)

History

TITAN Project (2007-2012)

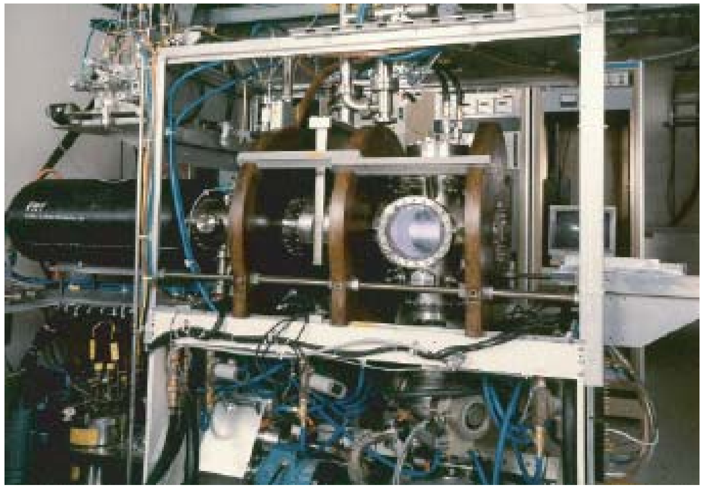
Deuterium retention in W irradiated with neutrons at coolant temperature of HFIR (~50 ^{O}C).

PHENIX Project (2013-2018)

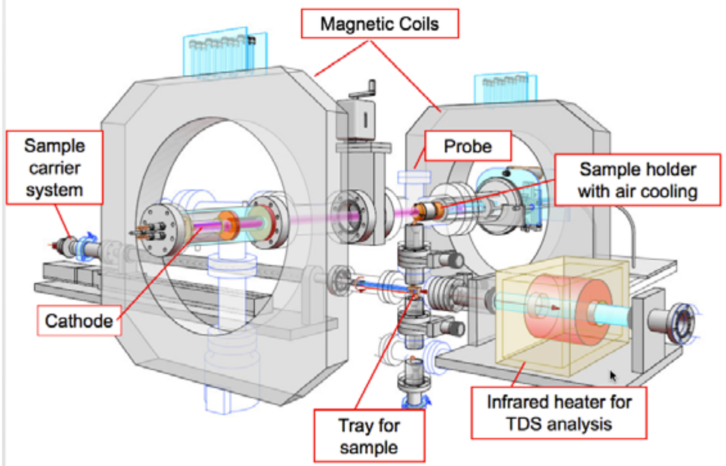
Deuterium retention in W irradiated with neutrons at elevated temps. (300–1100 ^{O}C).

IMR-Oarai (2015-)

Specimens irradiated in TITAN Project and Tohoku U.–SCK/CEN Collaboration (BR2, at 290 ^{o}C), state of the art positron annihilation, TEM and 3D Atom Probe analyses.

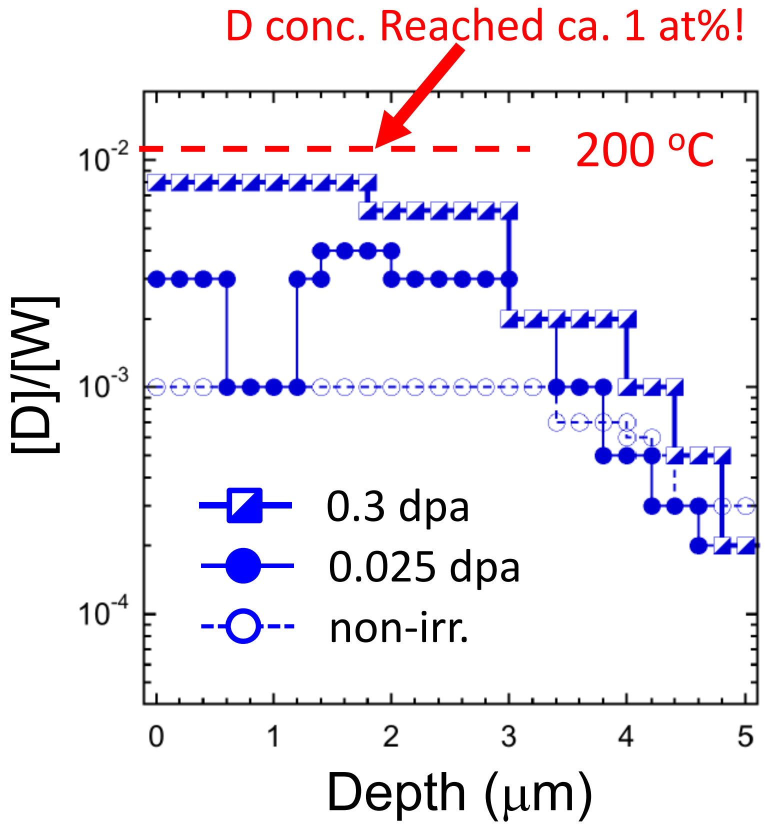


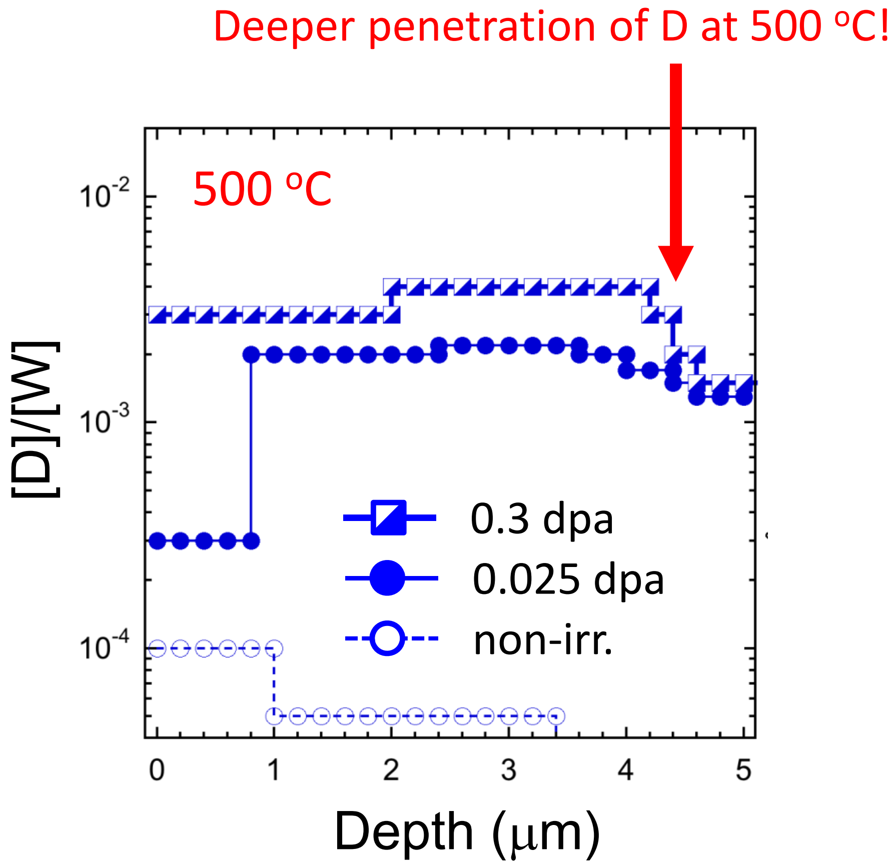
Tritium Plasma Experiment (TPE), Idaho National Laboratory (INL)



Compact Divertor Plasma Simulator (C-DPS), IMR-Oarai, Tohoku U.

Depth profiles of D in W after neutron irradiation in HFIR, ORNL at ~50 °C followed by exposure to high-flux D plasma at 200 or 500 °C.





Significant increase in D concentration by n-irradiation!

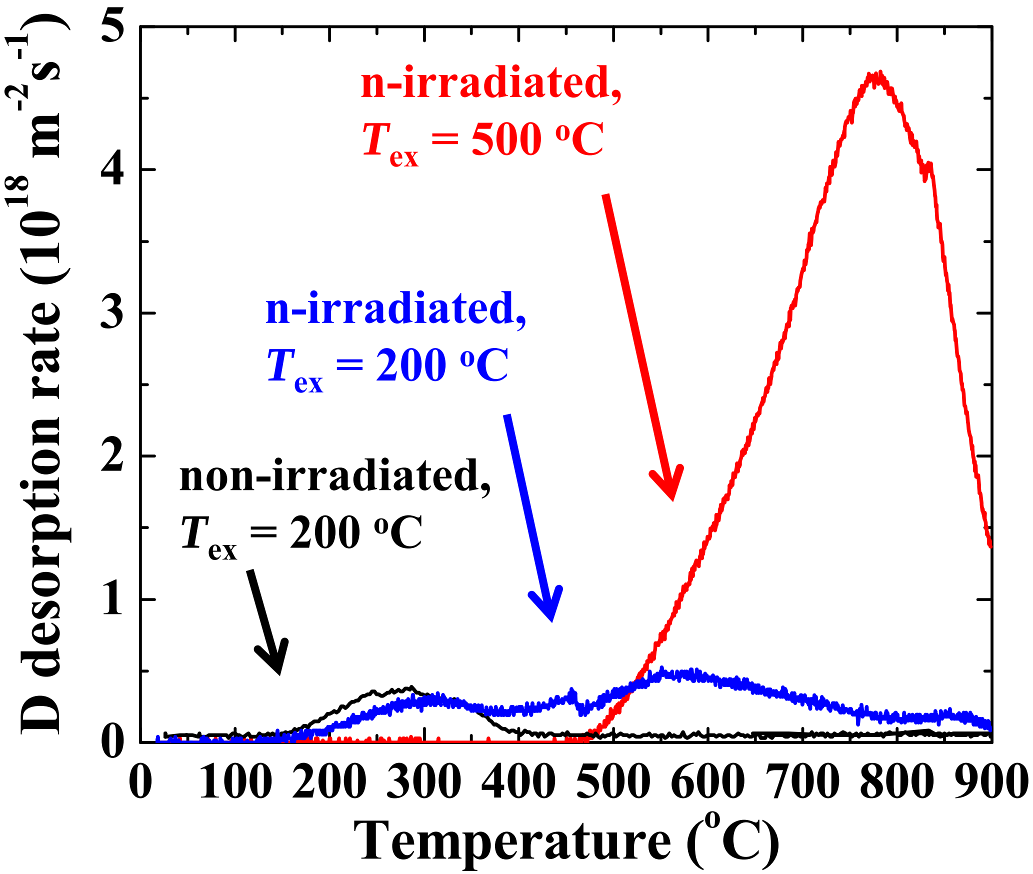
Exposure to D plasma in Tritium Plasma Experiment (TPE)

Sample temp. : 200 and 500 °C., D ion energy : 100 eV

Flux : (5-7) × 10^{21} D $m^{-2}s^{-1}$, Fluence : (5-7) × 10^{25} D m^{-2} (10 ks)

Y. Hatano et al., Nuclear Fusion, 53(2013)073006.

Thermal desorption spectra of D released from non-irradiated and n-irradiated W (0.025 dpa).



TDS spectra of n-irr. and non-irr. W at T_{ex} = 200 and 500 ^{o}C.

D retention at T_{ex} = 500 ^{o}C was 6.4×10^{21} D m^{-2}.

Because D concentration was 0.1–0.2 at.% (i.e., $6.3–12.6 \times 10^{25}$ D m^{-3}), penetration depth of D at T_{ex} = 500 ^{o}C was evaluated to be 50 – 100 μm.

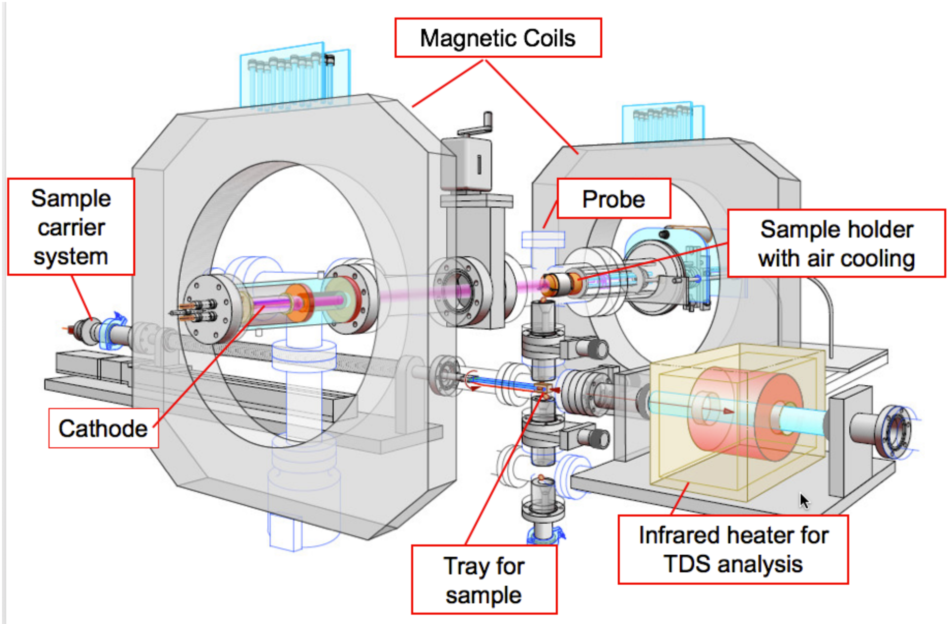
Y. Hatano et al.,
Journal of Nuclear Materials, 438(2013)S114-S119.
Nuclear Fusion, 53(2013)073006.

A large D retention at T_{ex} = 500 ^{o}C was ascribed to deep penetration of D into the bulk.

Contents

1. Long-range diffusion of D in n-irradiated W
2. Defects responsible for D trapping (positron annihilation)
3. Neutron-irradiation under hydrogen gas atmosphere : Capsule design and irradiation plan
4. Effects of Re (surrogate Fe ion irradiation)
5. Summary

1. Long-range diffusion of D in n-irr. W



Compact Divertor Plasma Simulator (C-DPS)

Specimen:
W disk (φ 6 × t 0.5 mm)
grains ⊥ surface
Belgium Reactor 2 (BR2), 290 °C, 0.06 dpa

Plasma exposure: C-DPS at 290 °C

Characteristics of C-DPS

- ✓ Good temperature control (±5 °C).
- ✓ Temperature rise before plasma ignition by electron bombardment.
- ✓ TDS without air exposure.
- ✓ TEM sample (ϕ 3 mm, t 0.1 mm) is acceptable.
- ◆ No tritium
- ◆ Max 500 °C at this moment

2. Defects responsible for D trapping (positron annihilation)

i. Neutron irradiation at 300 °C to 0.3 dpa (48 days in HFIR, ORNL, in TITAN Project) (ϕ3 × 0.2 mm W disk)

ii. Post-irradiation annealing at 300 °C in vacuum and 0.1 MPa D_2 gas for 100 and 400 h.

iii. Positron annihilation spectroscopy (PAS) and TDS.

T. Troev et al., Nuclear Instruments and Methods in Physics Research B 267 (2009) 535–541

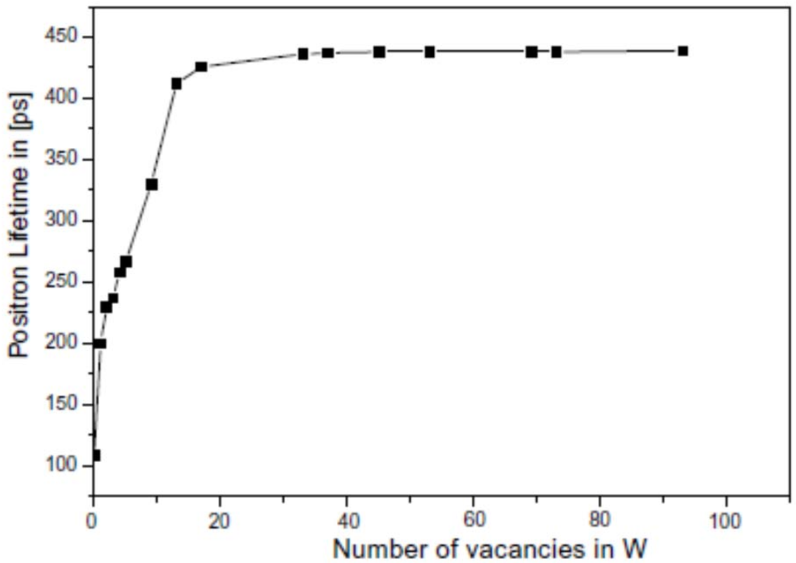
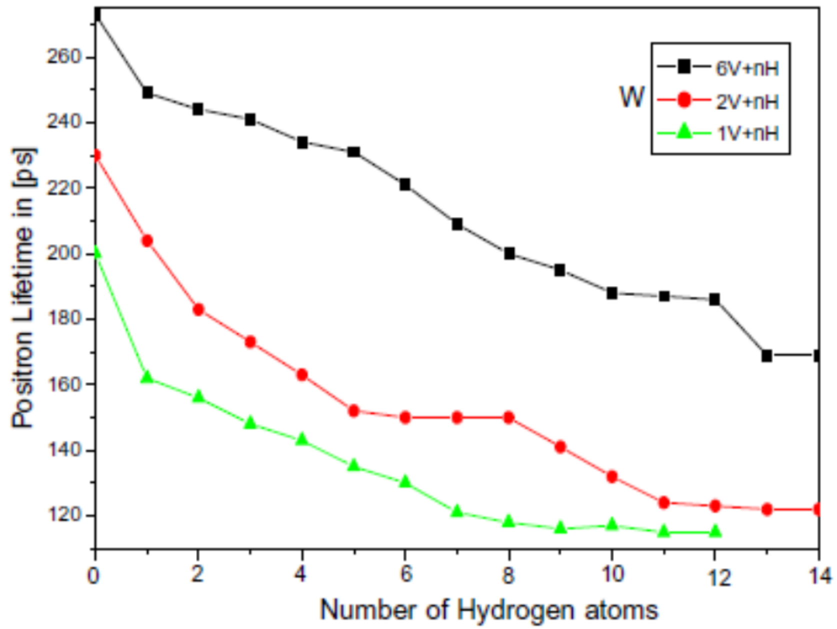


Fig. 3. Correlation between positron lifetime and the number of vacancies in a nano-void in tungsten.



3. Neutron-irradiation under hydrogen gas atmosphere: Capsule design and irradiation plan

(1) Motivation

In many experiments in this field, damages are created in hydrogen-free specimens, and then D is introduced by plasma- or gas-exposure. Only a limited number of studies examined defect migration under the presence of hydrogen.

In principle, vacancy-hydrogen clustering reduce mobility of vacancies.

Irradiation study under the presence of hydrogen is necessary!

In this study, metal hydride is used as a source of hydrogen gas.

Heating $MHx \rightarrow M + (x/2)H_2$
Cooling $M + (x/2)H_2 \rightarrow MHx$

H. Atsumi in Kindai University performed neutron irradiation of graphite with Mg_2NiH_4. (private comm.)

4. Effects of Re

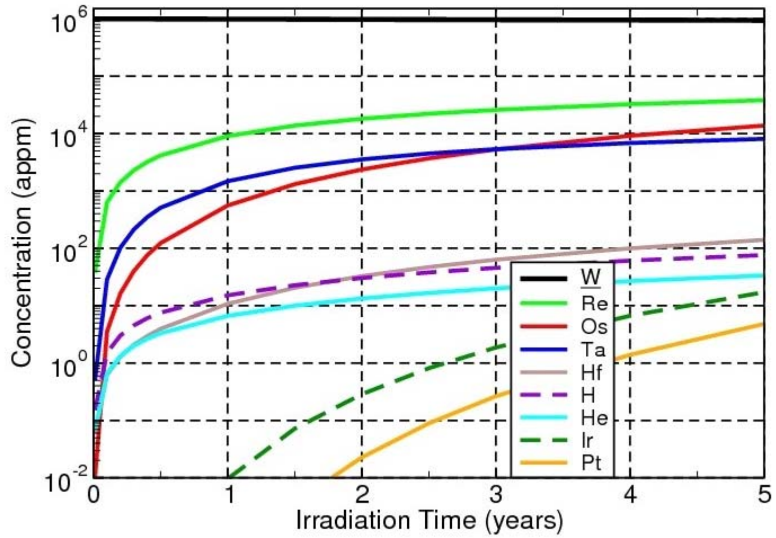
Due to transmutation, W automatically becomes W-xRe-yOs alloy under neutron irradiation. Hence, effects of Re have to be investigated.

Experimental Procedures

Specimens : Plates of pure W and W-5%Re alloy (10 × 10 × 0.5 mm)

- Irradiation with 6.4 MeV Fe ions to 0.5 dpa at 250–1000 °C.
- Exposure to D_2 gas at 0.1 MPa and 400 °C.
- NRA and TDS.
- Some specimens were irradiated at 250 °C and then annealed at 800 °C.

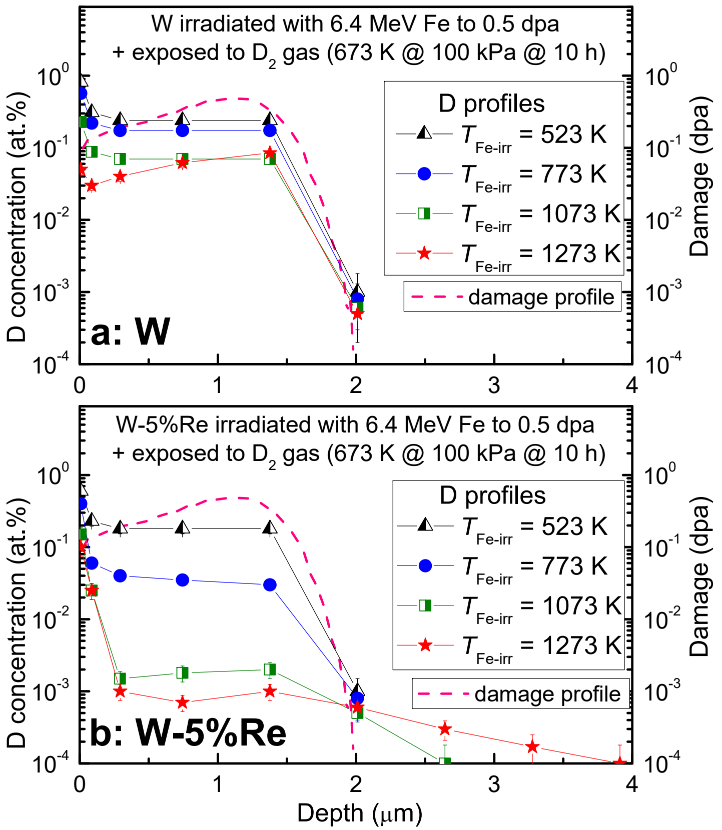
NRA was performed in IPP Garching, Germany by T. Schwarz-Selinger and M. Zibrov.



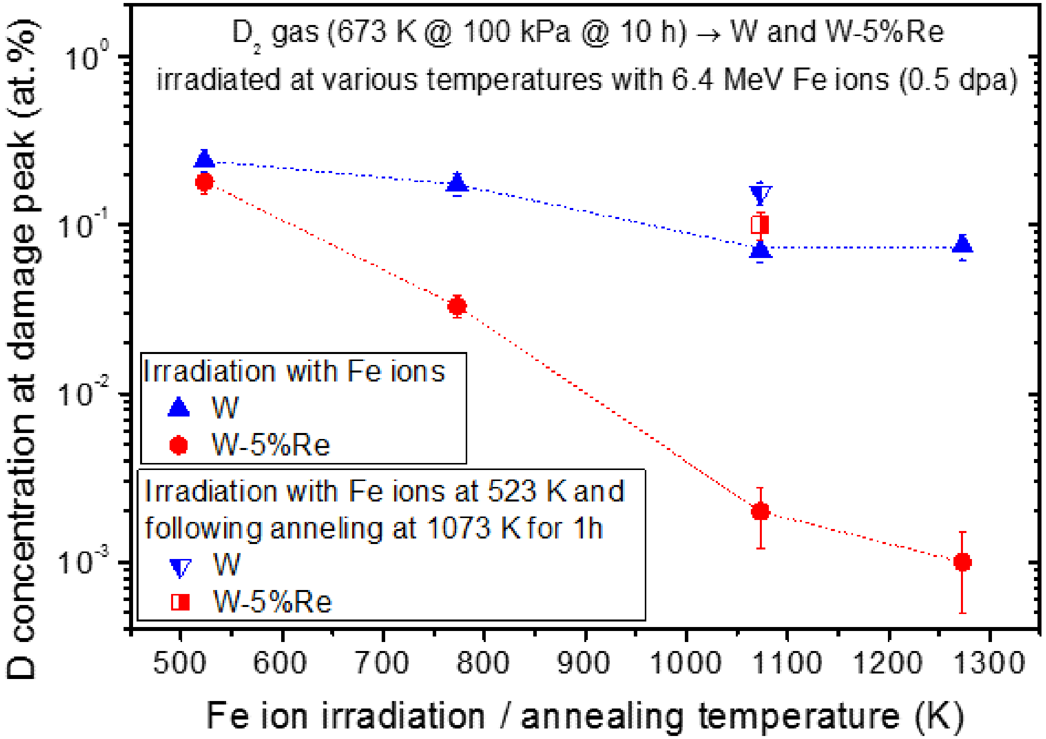
Transmutation in W under fusion neutron irradiation (M. R. Gilbert & J. -Ch. Sublet, Nucl. Fusion 51 (2011) 043005).

The presence of Re reduced trapped D by orders of magnitude after irradiation at high temperatures!

(Y. Hatano et al., Nuclear Materials and Energy 9 (2016) 93-97)



Depth profiles of D in W and W-5%Re irradiated with 6.4 MeV Fe ions to 0.5 dpa at the Bragg peak at different temperatures.

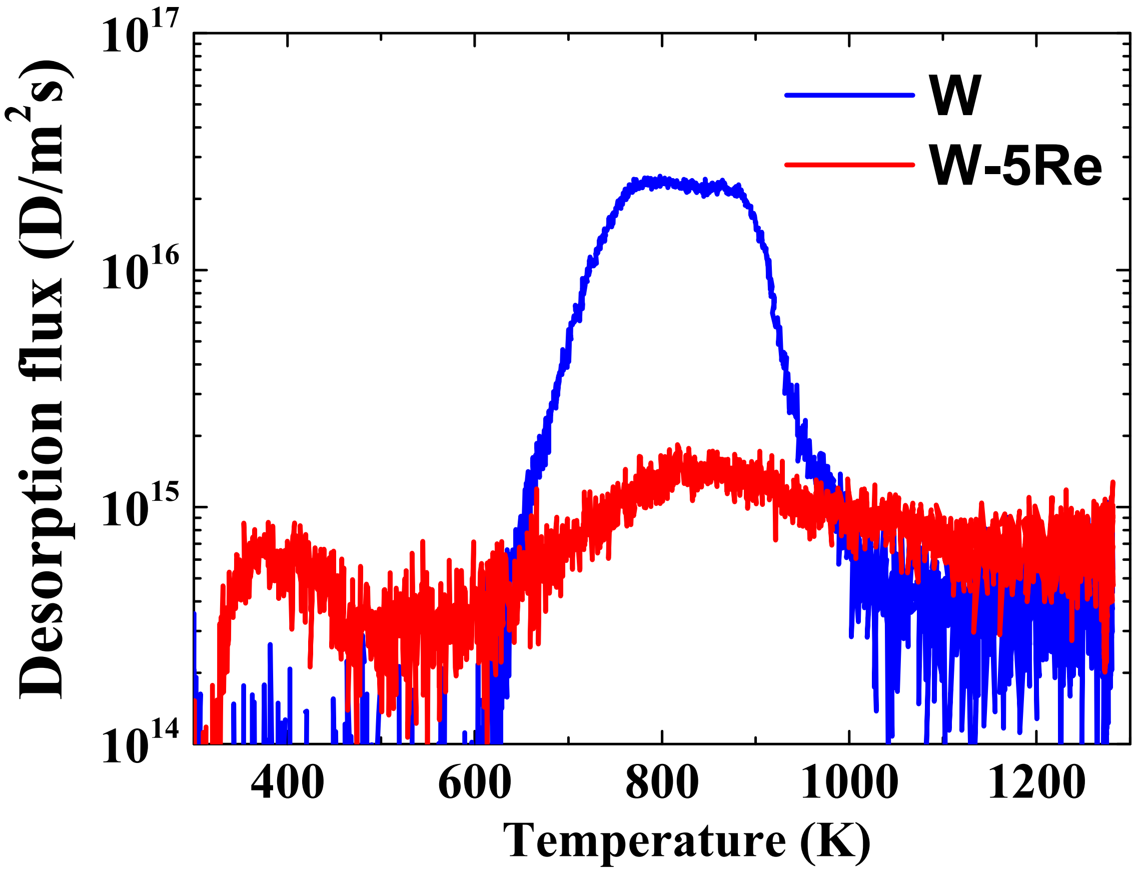


Correlation between irradiation temperature and concentration of D trapped in radiation-induced defect at the Bragg peak. Half-filled points showed data after irradiation at 523 K and post-irradiation annealing at 1073 K.

Reduced amount of trapped D can be explained by either of (1) reduced density of traps, and (2) reduced binding energy between D and traps.

Thermal desorption spectra giving the desorption peaks at almost the same temperature showed that the binding energy was comparable between W and W-5%Re alloy. In other words, reduced D retention by Re was due to reduced trap density.

(Y. Hatano et al., Nuclear Materials and Energy 9 (2016) 93-97)



Thermal desorption spectra of D from W and W-5Re alloy irradiated at 800 C.

Table 1 Results of positron lifetime measurements for W and W-5%Re samples before and after the irradiation with 6.4 MeV Fe ions at 1273 K to 0.5 dpa.
(Y. Hatano et al., Nuclear Materials and Energy 9 (2016) 93-97)

Sample	W (non-irradiated)	W (irradiated)	W-5%Re (non-irradiated)	W-5%Re (irradiated)
Positron lifetime (ps)	133.9	124.7 467.4	137.7	138.9

✓ Orders-of-magnitude reduction in D retention due to Re-assisted vacancy-interstitial atom recombination.
T. Suzudo et al., Modelling Simul. Mater. Sci. Eng. 22 (2014) 075006
X. -S. Kong et al., Acta Mater., 66(2014)172

Table 3
Summary of the microstructural observation in the matrix of pure W and W-Re alloys after the neutron irradiation.

Irradiation condition	Material	Void		Dislocation loop		Precipitate		
		Size (nm)	Number density (m^{-3})	Size (nm)	Number density (m^{-3})	Size		Number density (m^{-3})
						Major axis (nm)	Minor axis (nm)	
500 °C, 0.90 dpa	Pure W	–	–	2.9	3.3×10^{22}	7.8	3.6	8.6×10^{22}
	W–5%Re	–	–	2.4	5.2×10^{22}	3.0	0.8	2.6×10^{23}
	W–10%Re	–	–	–	–	3.2	1.2	9.0×10^{23}
800 °C, 0.98 dpa	Pure W	3.8	8.0×10^{21}	–	–	26.5	6.1	3.6×10^{22}
	W–3%Re	~1	$<1.0 \times 10^{20}$	–	–	11.6	2.0	1.3×10^{23}
	W–5%Re	~1	$<1.0 \times 10^{20}$	–	–	10.7	2.6	3.2×10^{23}
	W–10%Re	~1	$<1.0 \times 10^{20}$	–	–	10.3	2.4	2.9×10^{23}
	W–26%Re	–	–	–	–	17.8	6.1	1.4×10^{23}

Microstructure after HFIR irradiation reported by Fukuda et al., Tohoku U.

Fukuda et al. observed significant reduction in void density and formation of precipitates by Re addition after neutron-irradiation in HFIR at 800 C.

The present observation is consistent with their observations.

5. Summary

- ✓ Neutron irradiation induces V and V clusters in various sizes, and those act as strong trap sites with detrapping energy of 1.8–2 eV.
- ✓ Penetration depth of D was proportional to square root of plasma exposure time.
- ✓ Re significantly reduces trap concentration at temperatures ≥ 500 ^{o}C. Post-irradiation annealing did not work. Dynamic process under irradiation plays key-roles.
- ✓ High temperature neutron irradiation (800 and 1100 ^{o}C) of W-Re alloy in HFIR was completed in PHENIX Project. Specimens are waiting PIE.
- ✓ Neutron irradiation under H_2 gas atmosphere (400 ^{o}C, 0.1 dpa) is in progress under the framework of IMR-Oarai–ORNL collaboration, to understand effects of hydrogen on microstructure development under irradiation.