# Present status of activities in Japan on hydrogen isotope retention studies for neutron-irradiated tungsten materials

Y. Hatano (U. Toyama), H. T. Lee, Y. Ueda (Osaka U.), Y. Oya (Shizuoka U.),
T. Toyama (Tohoku U.)

1. Japan-US PHENIX Project (2013–2018) [Japanese Universities, INL, ORNL]
2. Neutron-irradiation program in International Research Center for Nuclear Materials Science (IRCNMS) [U. Toyama, Tohoku U. and Kyoto U.]
3. Do transmutation elements affect on hydrogen isotope retention in W after irradiation? Yes, for Re. [U. Toyama, Kyoto U., Tohoku U., IPP Garching]
4. Summary

All specimens used in this study were prepared from sheets or rods of W and W alloys supplied by A. L. M. T. Co., Japan.

# 1. Japan-US PHENIX Project (2013–2018)

## PHENIX

PFC evaluation by tritium Plasma, HEat and Neutron Irradiation eXperiments

The goal of this project is to evaluate the feasibility of He gas-cooled divertor with tungsten material armor for DEMO reactors.

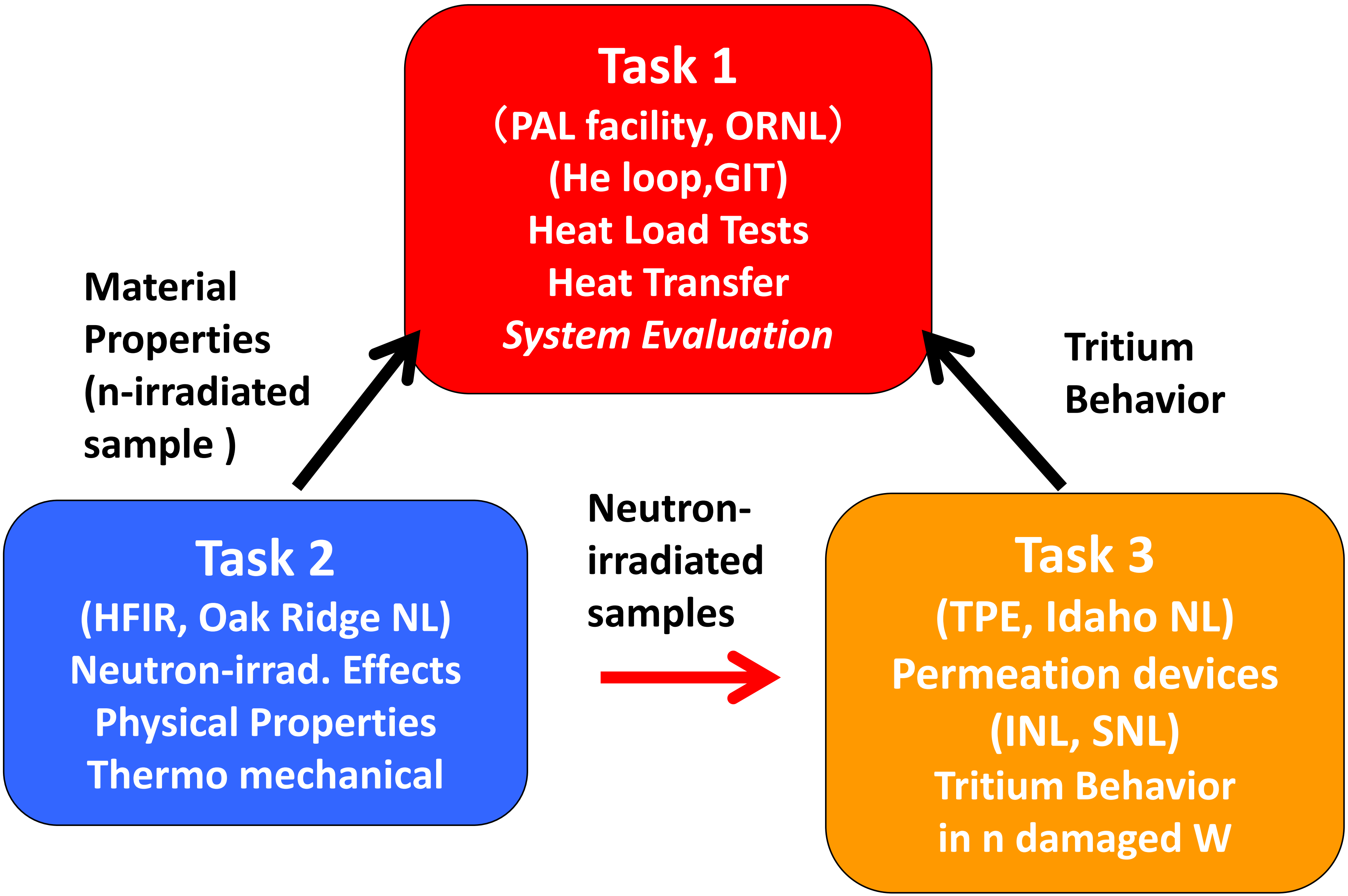
| | Japan | USA |
|---|---|---|
| Representative | Yoshio Ueda (Osaka U.) | Barry Sullivan (DOE)(interim) |
| Coordinator | Yuji Hatano (U. Toyama) | Barry Sullivan (DOE)(interim) |
| Task 1 | Takehiko Yokomine (Kyoto U.)<br>Yoshio Ueda (Osaka U.) | Adrian Sabau (ORNL)<br>Minami Yoda (GT) |
| Task 2 | Tatsuya Hinoki (Kyoto U.)<br>Akira Hasegawa (Tohoku U.) | Yutai Katoh (ORNL)<br>Lauren Garrison (ORNL) |
| Task 3 | Yasuhisa Oya (Shizuoka U.)<br>Yuji Hatano (U. Toyama) | Masashi Shimada (INL)<br>Dean Buchenauer (SNL) |

# Summary of the project

The goal of this project is to evaluate the feasibility of He gas-cooled divertor with tungsten material armor for DEMO reactors. Main research subjects are listed below;

1. Heat transfer mechanism and modeling in He-cooled systems, improvement of cooling efficiency and system design.
2. Response of tungsten layered materials and advanced tungsten materials to steady state and pulsed heat loads.
3. Thermo-mechanical properties measurement of tungsten basic materials, tungsten layered materials and advanced tungsten materials after neutron irradiation at elevated temperatures relevant to divertor conditions (500-1200 $^{o}$C).
4. Effects of high flux plasma exposure on tritium behavior in neutron-irradiated tungsten layered materials and advanced tungsten materials.
5. Evaluation of feasibility (under ~10 MW/m$^2$ heat load with irradiation of plasma and neutrons) and safety (tritium retention and permeation) of He-cooled PFCs and clarification of critical issues for DEMO divertor design.

# Structure of this Project



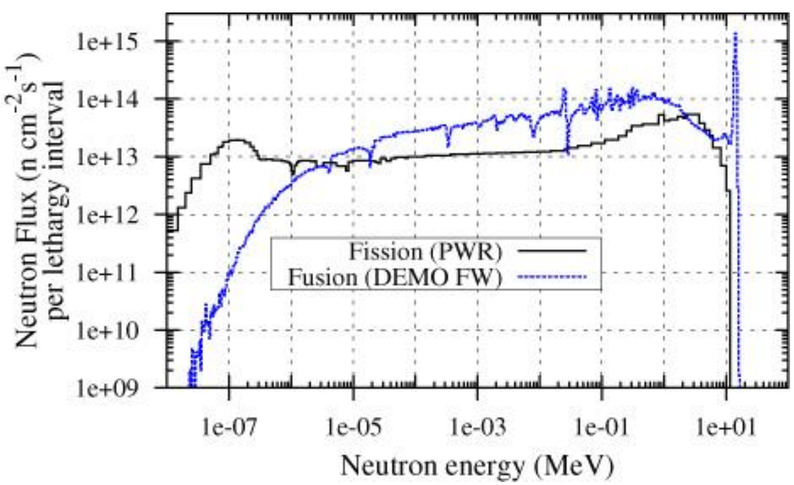
# Characteristics of PHENIX neutron irradiation in High Flux Isotope Reactor (HFIR), Oak Ridge National Laboratory

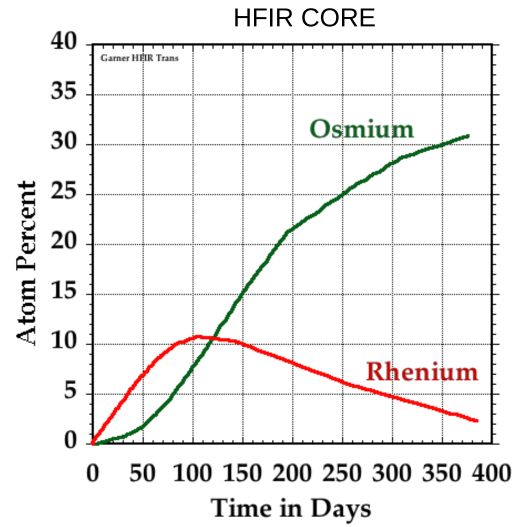
- ✓ Irradiation temperature: 500, 800 and 1200 ˚C.
- ✓ Irradiation dose: 1–1.5 dpa.
- ✓ Thermal neutron shielding (Gd)

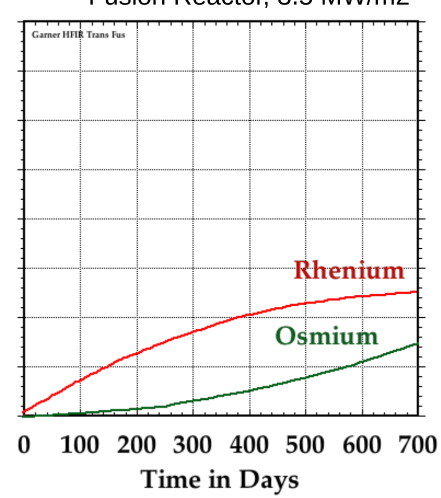
  Thermal neutron shielding is essential for fusion-relevant transmutation.
- ✓ Specimens for hydrogen isotope retention and permeation measurements

  $\phi$6 x 0.5 mm or $\phi$10 x 0.5 mm, W, W-Re, K-doped W-Re, UFG-W

*After L. Snead*



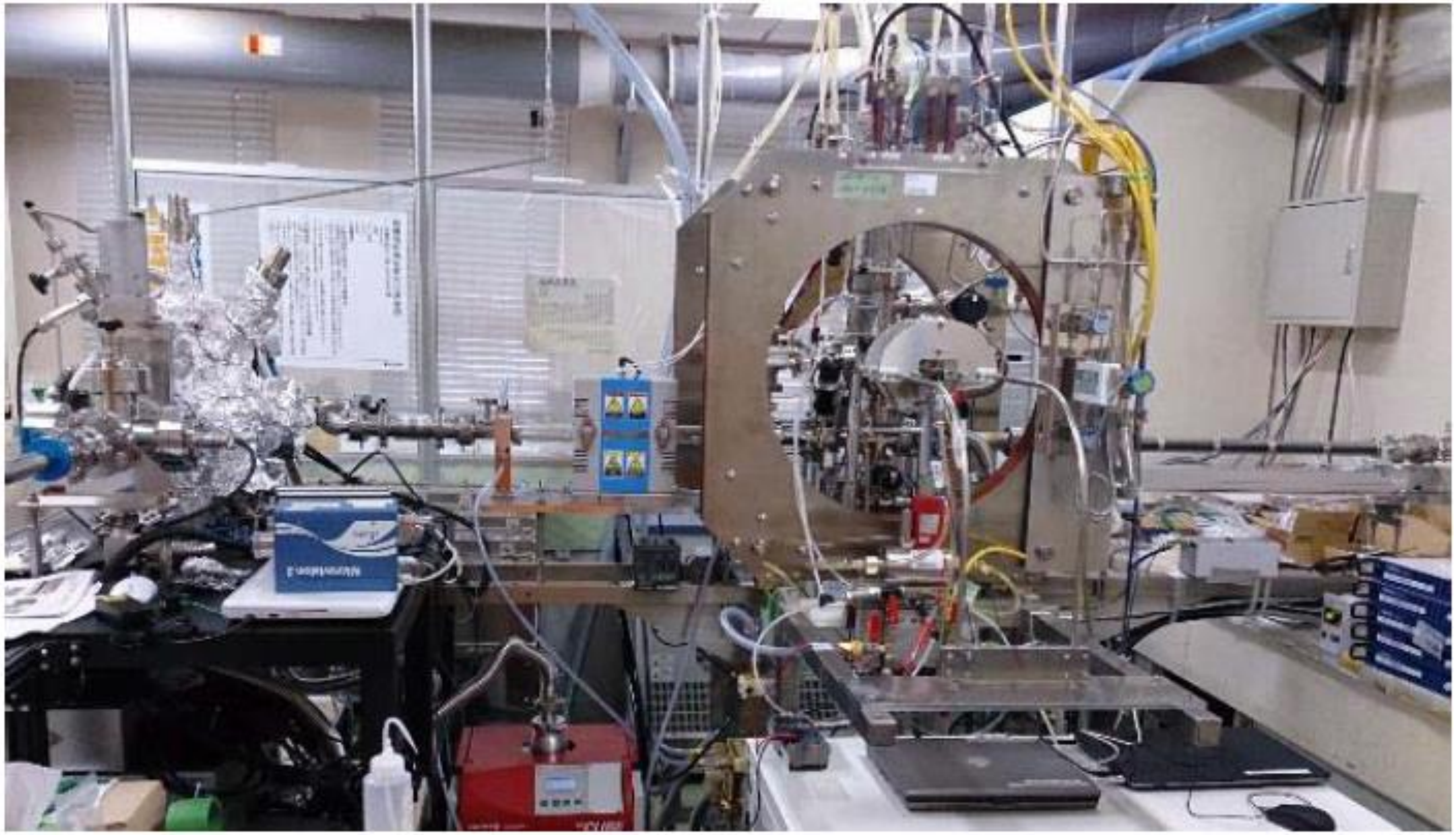




Greenwood/Garner JNM 212-215 (1994)

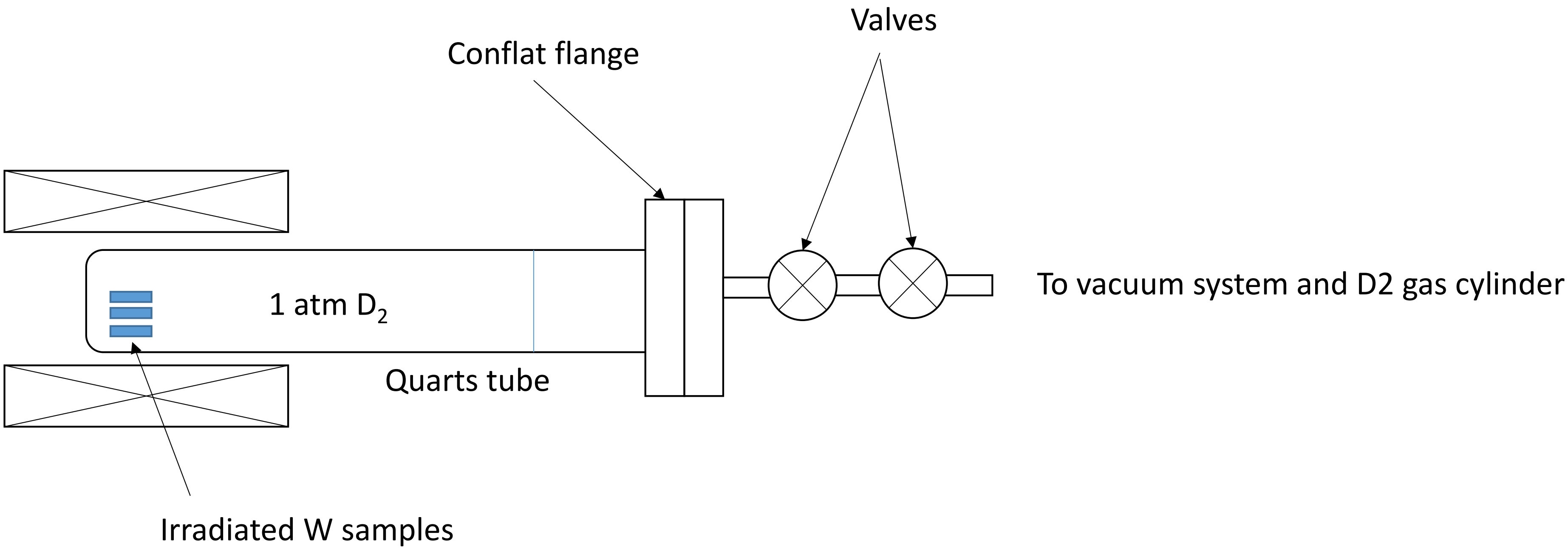
**Difference in neutron spectrum and transmutation between fusion and fission.**

# 2. Neutron-irradiation program in International Research Center for Nuclear Materials Science (IRCNMS)



The compact divertor plasma simulator (C-DPS) has been installed in the radiation-controlled area of IRCNMS.

- ✓ C-DPS Flux:$10^{24}$ D $m^{-2}s^{-1}$, Incident Energy: 20–140 eV, Sample temperature control feedback system Currently under test operation and real experiment will start soon.
- ✓ Long-term $D_2$ gas exposure experiments (several hundreds hours) in a sealed capsules have been performed to examine long-range diffusion of D and vacancy-type defects under the presence of D.



- ✓ Neutron irradiation in BR2 reactor has been completed and the specimens have been shipped to IRCNMS. (A part of materials were supplied by Prof. A. Hasegawa, Tohoku U.)

  Specimens: Disks ($\phi$6 × 0.5 mm)

  single crystal W, polycrystal W,

  W-Re alloy, K-doped W, K-doped W-Re

  Irradiation temperature: 290 ˚C

  Irradiation dose: 0.02–0.04 dpa

- ✓ Several TEM size W disks ($\phi$3 × 0.2 mm) irradiated at 300 ˚C to 0.3 dpa in HFIR in the previous Japan-US collaboration are also available.
- ✓ Positron annihilation spectroscopy (PAS, measurements of positron lifetime and coincidence Doppler-broadening (CDB) for defect characterization.
- ✓ Long-term $D_2$ gas exposure for HFIR specimens has already been started. Plasma exposure and gas exposure for BR2 specimens will follow soon.
- ✓ Tests for W specimens irradiated with high-energy electrons (8.5 MeV) have been performed to check experimental scenario, and some interesting results have been obtained as shown later.

# Experimental Scenario for n-irradiated specimens

PAS, TEM

Irradiation

PAS, TEM

D plasma exposure
Long-term $D_2$ gas exposure
Vacuum annealing to examine temperature effects

PAS, TEM

TDS

# Measurements for recrystallized W irradiated with 8.5 MeV electrons (surrogate irradiation)

PAS

Irradiation, 8.5 MeV e-beam, $10^{-3}$ dpa at ~100 C

PAS

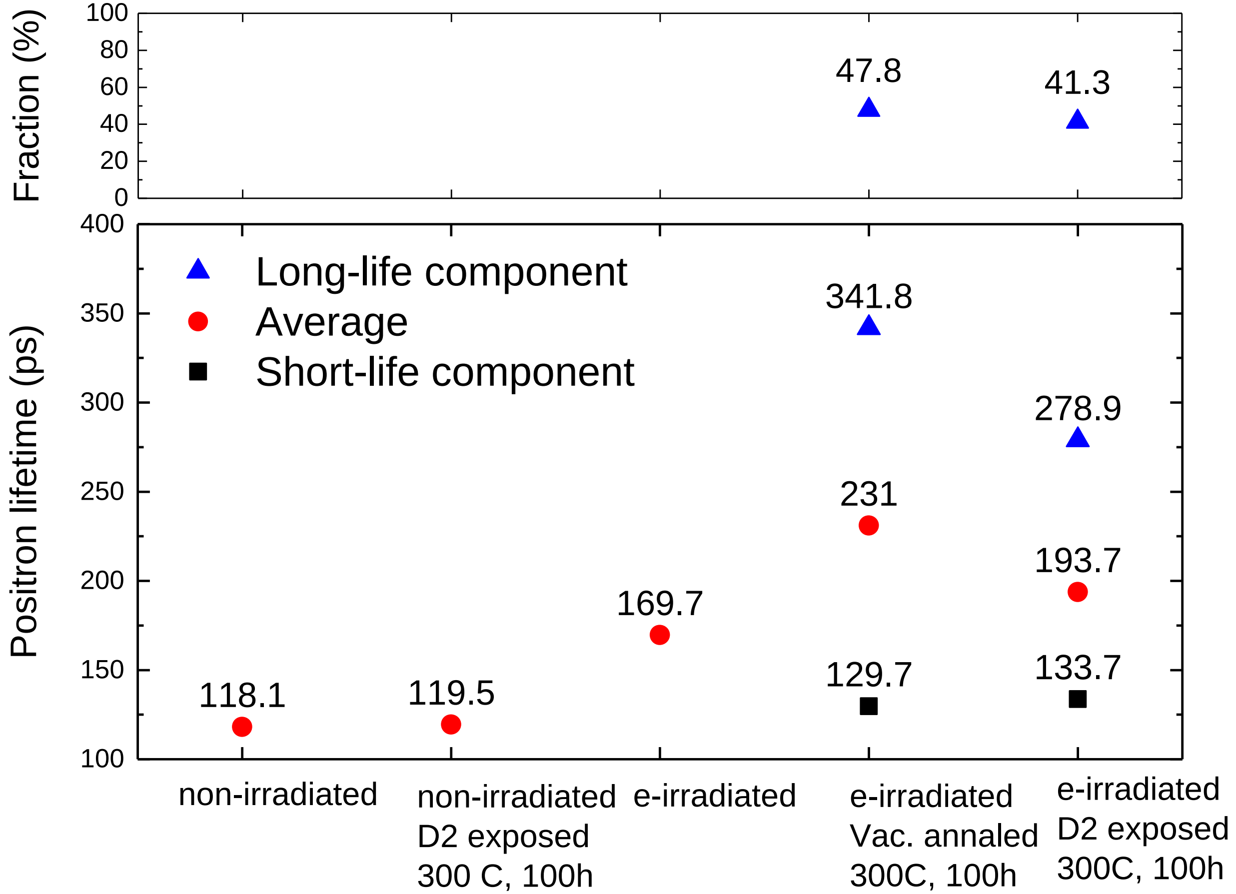
Long-term $D_2$ gas exposure (300 C, 100 h)
Vacuum annealing (300 C, 100 h)

PAS

TDS

**TDS spectra of non-irradiated and e-irradiated W after $D_2$ gas exposure at 0.1 MPa and 300 C for 100 h.**

Electron irradiation resulted in significant increase in D retention.




Clustering of vacancies was clearly observed at temperature as low as 300 C.

Coincidence Doppler broadening (CDB) is powerful method to detect H isotope in vacancy-type defect (shown later).

**Change in positron lifetime by e-irradiation, vacuum annealing and $D_2$ gas exposure.**

Electron irradiation resulted in formation of monvacancies (~170 ps). $D_2$ gas exposure as well as vacuum annealing at 300 C for 100 h led to clustering of vacancies.

Interestingly, lifetime of long-life component after $D_2$ gas exposure was shorter than that after vacuum annealing. This difference indicates the occupation of vacancy clusters by D (confirmed by CDB) and slower vacancy diffusion under the presence of D (needs to be confirmed by TEM). Similar measurements will be performed for n-irradiated specimens.

# Examples of positron lifetime in defects and defect-hydrogen clusters

| Type of defect | Positron lifetime (ps) |
|---|---|
| matrix | 108 |
| monovacancy | 258 |
| monovacancy + 1H | 162 |
| 5 vacancy cluster | 267 |
| 9 vacancy cluster | 330 |
| 6 vacancy cluster + 7H | 209 |

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

## 3. Do transmutation elements affect on hydrogen isotope retention in W after irradiation?

Re and Os are major transmutation elements in W after irradiation of fusion neutrons. To see the effects of Re, W-5%Re alloy was irradiated with 6.4 MeV Fe ions in DuET, Kyoto U. at different temperatures, and D retention was measured using nuclear reaction analysis (NRA) in IPP Garching. TDS and PAS were also performed.

Specimens: Sheets of W and W-5%Re

Irradiation: 6.4 MeV Fe ions to 0.5 dpa at the Bragg peak at 250, 500, 800 and 1000 C.
(DuET, Kyoto U.)

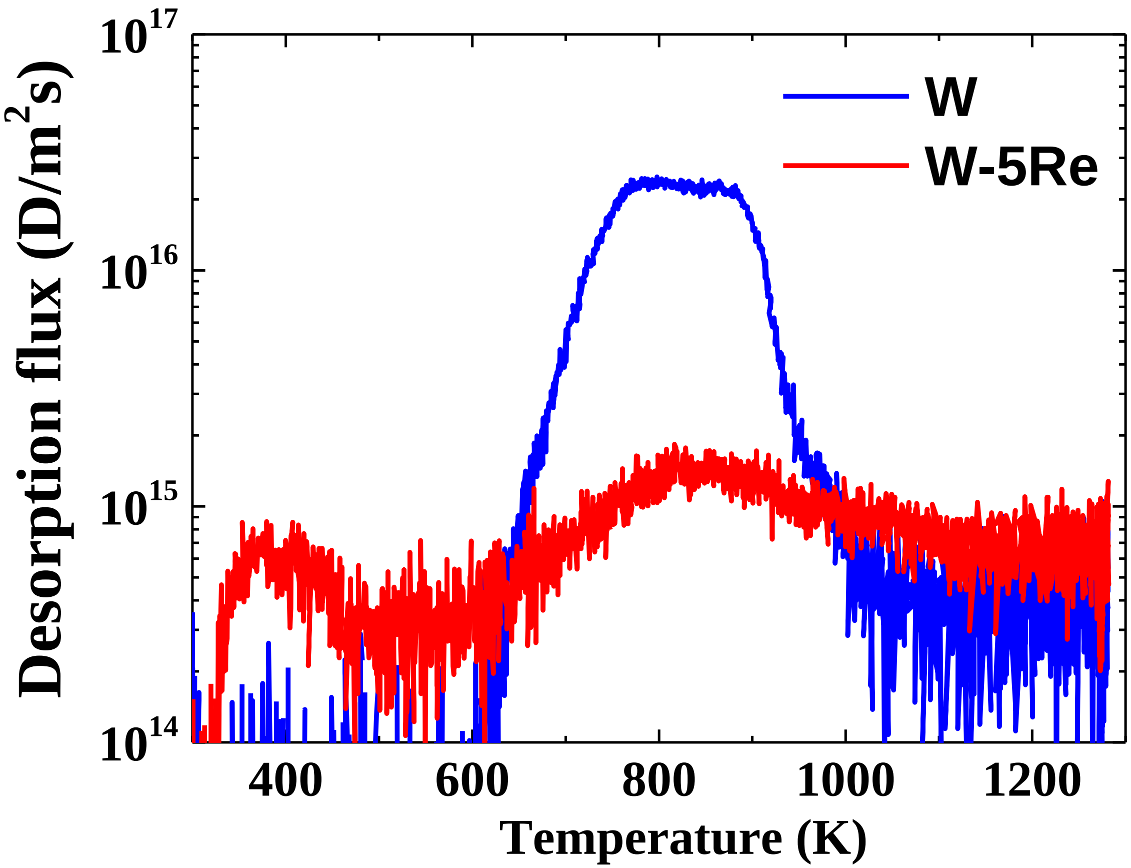
D loading: Exposure to $D_2$ gas at 0.1 MPa and 400 C for 10 h.

D detection: NRA (IPP Garching)

*Irradiation at different temperatures followed by D loading at the same condition clearly shows the correlation between trap density and irradiation temperature.*

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.




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

## Summary of positron lifetime measurements (average) for specimens irradiated at 1000 C

Positron lifetime in W significantly increased by 6.4 MeV Fe ion irradiation due to formation of vacancy-type defects.

In contrast, no significant increase in positron lifetime was observed for W-5%Re alloy. This observation suggests that the density of vacancy-type defect is significantly low in W-5%Re alloy.

The orders-of-magnitude lower D retention in W-5%Re alloy can be attributed to the low density of vacancy-type defects.

M. Fukuda et al. / Journal of Nuclear Materials 455 (2014) 460–463 463

**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 | $3.3 \times 10^{22}$ | 7.8 | 3.6 | $8.6 \times 10^{22}$ |
| | W–5%Re | – | – | 2.4 | $5.2 \times 10^{22}$ | 3.0 | 0.8 | $2.6 \times 10^{23}$ |
| | W–10%Re | – | – | – | – | 3.2 | 1.2 | $9.0 \times 10^{23}$ |
| 800 °C, 0.98 dpa | Pure W | 3.8 | $8.0 \times 10^{21}$ | – | – | 26.5 | 6.1 | $3.6 \times 10^{22}$ |
| | W–3%Re | ~1 | $<1.0 \times 10^{20}$ | – | – | 11.6 | 2.0 | $1.3 \times 10^{23}$ |
| | W–5%Re | ~1 | $<1.0 \times 10^{20}$ | – | – | 10.7 | 2.6 | $3.2 \times 10^{23}$ |
| | W–10%Re | ~1 | $<1.0 \times 10^{20}$ | – | – | 10.3 | 2.4 | $2.9 \times 10^{23}$ |
| | W–26%Re | – | – | – | – | 17.8 | 6.1 | $1.4 \times 10^{23}$ |

## 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.

# 4. Summary

- ✓ Construction of irradiation capsule for PHENIX project has been started. The characteristic points of this capsule is thermal neutron shielding by Gd layer for appropriate transmutation. Irradiation will be performed at 500, 800 and 1200 C to 1–1.5 dpa. PIE will starts in the summer of 2017.
- ✓ The new compact linear plasma device and long-term gas exposure system have been constructed in International Research Center for Nuclear Materials Science, Institute for Materials Research, Tohoku University. Lower-dose neutron irradiation (0.02–0.04 dpa) in BR2 reactor (and consequently low Re and Os) at 290 C has been completed. PIE will start in this autumn.
- ✓ Trapping of hydrogen isotopes in vacancy-type defects has been directly detected by positron annihilation spectroscopy (coincidence Doppler broadening).
- ✓ Re content and irradiation temperature affect very strong on hydrogen isotope retention and density of vacancy-type defects.
- ✓ High temperature irradiation with thermal neutron shielding is really necessary to simulate fusion environment in fission reactors. The irradiation in PHENIX project fulfills these requirements.