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Hydrogen Isotope Retention in Neutron-Irradiated Tungsten and Tungsten Alloys

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1. Japan-US TITAN Project (2007–2012) [Japanese Universities, INL, ORNL]
   First data on D retention in and release from n-irradiated W after exposure to high flux D plasma
2. Surrogate irradiation experiments to understand details of trapping mechanisms [U. Toyama, Kyoto U., IPP Garching]
   Trapping-detrapping equilibrium by D_2 gas exposure
   Isotope exchange
3. Japan-US PHENIX Project (2013–2018) [Japanese Universities, INL, ORNL]
4. Neutron-irradiation program in the International Research Center for Nuclear Materials Science (IRCNMS), Institute for Materials Research, Tohoku University
5. Tritium experiments in Hydrogen Isotope Research Center, U. Toyama
   Isotope effects in trapping by radiation-induced defects?
6. Summary

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1. TITAN Project (2007-2012)/ Overview

TITAN (Tritium, Irradiation and Thermofluid for America and Nippon)
Major objective of this project is to evaluate the consistency of the blankets with first wall and recovery systems with respect to tritium and heat control.

TITAN Structure (revised on Feb. 9, 2012)

Representatives Coordinators		JP : K. Okuno (Shizuoka U.) JP : T. Muroga (NIFS)		US : P. Pappano(USDOE) US : P. Pappano(USDOE)		
Task	Subtask	Facility	TC (JP)	STC/Deputy (JP)	TC (US)	STC/Deputy (US)
Task 1 Transport phenomena	1-1 Tritium and mass transfer in first wall	TPE PISCES	T. Terai (U.Tokyo)	Y. Ueda (Osaka U.)/ N. Ohno (Nagoya U.) K. Tokunaga (Kyushu U.)	P. Pappano (USDOE)	R. Doerner (UCSD)
	1-2 Tritium behavior in blanket systems	STAR		T. Terai (U. Tokyo)/ S. Fukada (Kyushu U.) S. Konishi (Kyoto U.)		B. Merrill (INL)
	1-3 Flow control and thermofluid modeling	MTOR		T. Kunugi (Kyoto U.)/ T. Yokomine (Kyoto U.)		S. Smolentsev (UCLA)/ K. Messadek (UCLA)
Task 2 Irradiation synergism	2-1 Irradiation-tritium synergism	HFIR STAR	A.Kimura (Kyoto U.)	Y. Hatano (Toyama U.)/ Y. Oya (Shizuoka U.)	R. Kurtz (PNNL)	Y. Katoh (ORNL)/ B. Merrill(INL)
	2-2 Joining and coating integrity	HFIR ORNL-HL (incl. T-test)		A. Kimura (Kyoto U.)/ N. Hashimoto (Hokkaido U.) T. Nagasaka (NIFS)		T. Yamamoto (UCSB)/ Y. Katoh (ORNL)
	2-3 Dynamic deformation			A.Hasegawa (Tohoku U.)/ T. Hinoki (kyoto U.)		Y. Katoh (ORNL)
Common Task System integration modeling	MFE/IFE system integration modeling		A.Sagara (NIFS)	A. Sagara (NIFS)/ H. Hashizume (Tohoku U) T. Norimatsu (Osaka U.)	R. Nygren (SNL)	R. Nygren (SNL)
Laboratory Liaisons	ORNL : INL : IMR-Oarai (Tohoku) :	Y. Katoh (ORNL) B. Merrill (INL) T. Shikama (Tohoku U.)				
IFE Liaisons			K. Tanaka (Osaka U.)	Kodama(Osaka U.) Yoneda(UTC)	M. Tillack (UCSD)	

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1. TITAN Project/ Experimental Procedures
(1) Neutron Irradiation and Post-Irradiation Examinations

(i) Irradiation in ORNL

W specimens

Specimen: Disks of pure W (99.99%)(A.L.M.T. Corp., Japan)
Stress-relieve treatment (900 °C, 1.5 h)
For retention measurements : ϕ6 mm , t = 0.2 mm

Grains are elongated in direction perpendicular to surfaces like ITER-Grade W.

n-irradiation: 0.025 and 0.3 dpa at 50 °C (coolant temp.) in the High Flux Isotope Reactor (HFIR) . No thermal neutron shield.

(ii) Deuterium retention measurements in INL

γ-ray spectroscopy to check activation of specimens.
Exposure to D plasma in Tritium Plasma Experiment (TPE)
Specimen temp. : 200 and 500 °C.
Energy : 100 eV
Flux : (5-7) × 10^{21} D $m^{-2}s^{-1}$
Fluence : (5-7) × 10^{25} D m^{-2} (10 ks)
D retention was measured by Nuclear Reaction Analysis (NRA) and Thermal Desorption Spectroscopy (TDS)

TPE in INL

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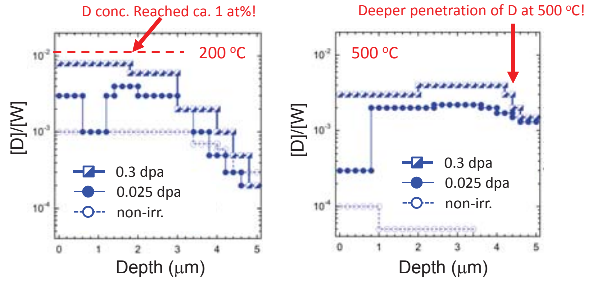
1. TITAN Project/ Results and Discussion (cont'd)
(i) D depth profiles by NRA

D conc. Reached ca. 1 at%!

Deeper penetration of D at 500 °C!



Depth profiles of D in n-irradiated W after exposure to D plasma at 200 and 500 °C. D profiles for 0.3 dpa specimens are preliminary; the specimens have been exposed to plasma only one time, and no guarantee on saturation of traps with D.

Significant increase in D concentration by n-irradiation!

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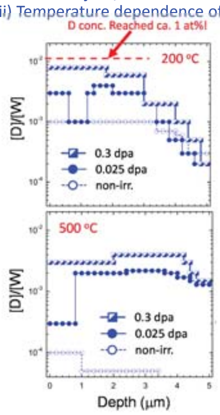
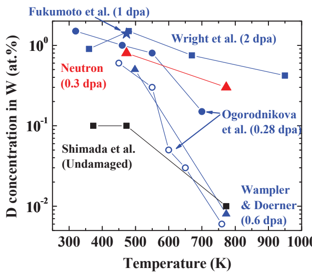
1. TITAN Project/ Results and Discussion (cont'd)
(ii) Temperature dependence of D retention in n-irradiated W



Depth profiles measured by NRA



Temp. dependence of D retention in irradiated W.

Relatively high retention even at 500 °C !

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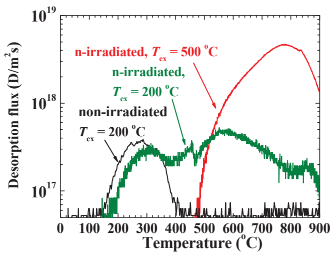
1. TITAN Project/ Results and Discussion (cont'd)
(iii) Thermal desorption spectroscopy (TDS)



TDS spectra of n-irr. (0.025 dpa) and non-irr. W. T_{ex} is temperature for D plasma exposure.

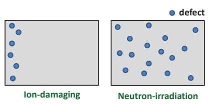
A large D retention at T_{ex} = 500 °C was ascribed to deep penetration of D into the bulk.

Desorption of D from n-irradiated W continued up to 900 °C!

Far larger retention in n-irradiated W than ion-damaged one.

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

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



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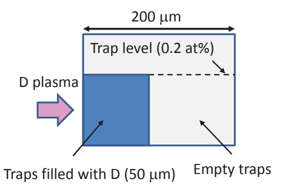
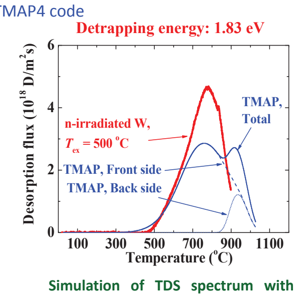
1. TITAN Project/ Results and Discussion (cont'd)
(iv) Simulation of TDS spectrum with TMAP4 code



Initial conditions for TMAP4 simulation corresponding to n-irradiated W exposed to D plasma at 500 °C.



Simulation of TDS spectrum with TMAP4 code (linear scale)

Position of main peak agreed by adjusting detrapping energy to ca. 1.8 eV.
The absence of high temperature shoulder in measured spectrum was due to annealing of defects.

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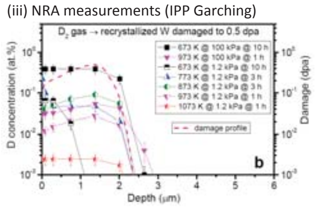
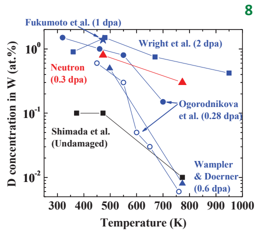
2. Surrogate irradiation experiments
(i) Trapping-detrapping equilibrium / Procedures

Recrystallized W (at 2070 K) and ITER-grade W

(i) Ion irradiation: 0.5 dpa at room temperature by 20 MeV W ions (IPP Garching)
(ii) Exposure to D_2 gas at 400–700 °C and 1.2 and 100 kPa for 1–10 h.
(iii) NRA measurements (IPP Garching)



Depth profiles of D measured by NRA in recrystallized W damaged with 20 MeV W ions and exposed to D_2 gas



$$\theta_t/(1-\theta_t) = \theta_L \exp(E_{bin}/kT),$$

θ_t : fraction of occupied traps
θ_L : fraction of occupied interstitial sites
E_{bin}: binding energy between D & traps

Under exposure to D_2 gas, θ_L is determined by Sieverts law!

$$\theta_L = k_0 \exp(-E_S/kT_{ex})P^{1/2}$$

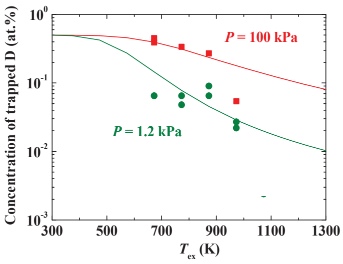
k_0: Solubility constant
E_S: heat of solution

2. Surrogate irradiation experiments

(ii) Trapping-detrapping equilibrium / Results

$\theta_t/(1-\theta_t) = \theta_L \exp(E_{bin}/kT)$,

$\theta_L = k_0 \exp(-E_S/kT_{ex})P^{1/2}$



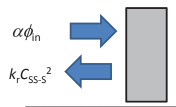
Dependence of concentration of trapped D on D_2 gas pressure and temp.

Concentration of trapped D was clearly dependent on solute D; trapping-detrpping equilibrium determines D retention at high temperatures !

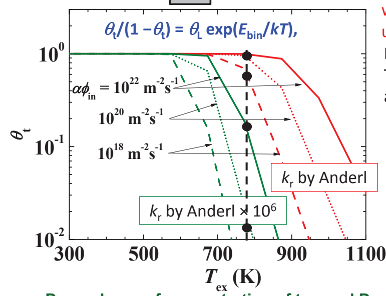
The observed correlation was reproduced by assuming E_{bin} = 1.4 eV.

Good agreement with the value evaluated for n-irradiated W.

Evaluation of D retention under plasma exposure



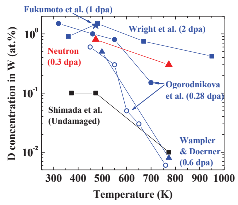
In steady state, $C_{SS\text{-}S} = \theta_L N_L = (\alpha\phi_{in}/k_r)^{1/2}$, where N_L is number of interstitial sites per unit volume of normal W lattice.



Dependence of concentration of trapped D on temp. and incident flux at E_{bin} = 1.4 eV.

The disagreement at high temp. was ascribed to difference in C_{SS} under different $\alpha\phi_{in}$ and k_r.

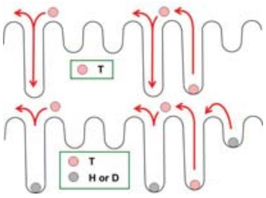
Important parameters for T retention are trap density, E_{bin} and solute T conc.



2. Surrogate irradiation experiments

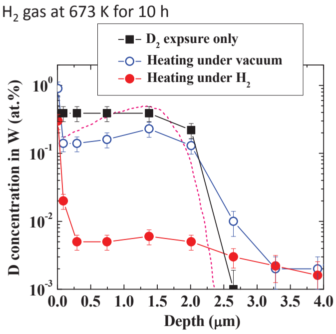
(iii) Isotope exchange/ Enhancement of D release under the presence of H

(i) Specimens irradiated with 20 MeV W ions were exposed to D_2 gas to saturate traps.
(ii) Heat treatments in a vacuum or H_2 gas at 673 K for 10 h
(iii) NRA analysis.



Schematics of diffusion of T in W without or with excess H or D

Strongly enhanced release of D with presence of excess H.



D profiles after loading and after heating under absence or presence of H

3. 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 <u>He gas-cooled divertor with tungsten material</u> armor for DEMO reactors.

	Japan	USA
Representative	Yoshio Ueda (Osaka U.)	Peter Pappano (DOE)
Coordinator	Yuji Hatano (U. Toyama)	Peter Pappano (DOE)
Task 1	Takehiko Yokomine (Kyoto U.) Yoshio Ueda (Osaka U.)	Richard Nygren (SNL)
Task 2	Tatsuya Hinoki (Kyoto U.) Akira Hasegawa (Tohoku U.)	Yutai Katoh(ORNL)
Task 3	Yasuhisa Oya (Shizuoka U.) Yuji Hatano (U. Toyama)	Brad Merrill (INL) Dean Buchenauer (SNL)

Summary of the project

The goal of this project is to evaluate the feasibility of <u>He gas-cooled divertor with tungsten material</u> 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 (500-1450 ºC).
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/m² 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

Task 1
(IR facility, ORNL)
Heat Load Tests
Heat Transfer
System Evaluation

Material Properties

Tritium Behavior

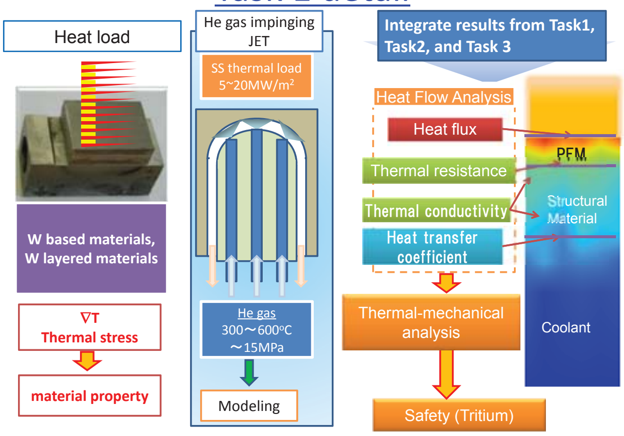
Task 2
(HFIR, Oak Ridge NL)
Neutron-irrad. Effects
Microstructure
Physical Properties

Neutron-irradiateds amples

Task 3
(TPE, Idaho NL)
Plasma-Surf. Interac.
Tritium Behavior

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

Task 1 detail



Task2 details

Material selection and modification
- W based material, advanced W material including alloy, surface treatment and composites: developed by US and JP
- Layered material: led by JP
- Information exchange and feedback for development

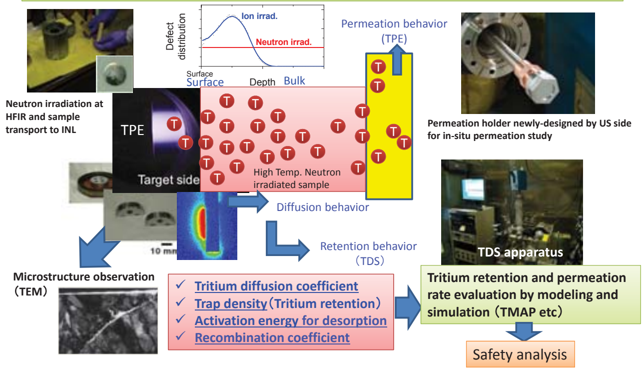
Post Irradiation Experiment

Neutron Irradiation
- Temperature: 500℃～1450℃, Fluence: ～3 dpa
- 3dpa irradiation: first half
- 1dpa irradiation: first half and second half
- Feedback from first half results to development
- High fluence（～15 dpa）: Utilizing ion irradiation

Post irradiation experiment
- LAMDA, IMET: thermal conductivity, toughness, strength, microstructure
- IR Facility: hear load
- Oarai, Japan: supporting

Task3 details

Neutron irradiation at higher temperature (500 – 1450 °C) is performed to simulate divertor conditions and the retention of hydrogen including tritium is also studied in high temperature region after neutron irradiation.



Tentative Neutron Irradiation Plan in PHENIX Project

Temperature range:
250–500 °C, 800–1000 °C, 1200 °C

Atmosphere:
He (all temperatures) and D_2 (1200 °C only)

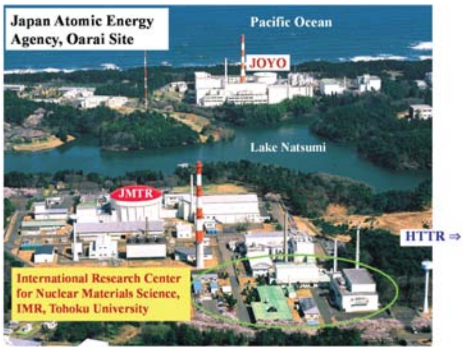
Thermal neutron shielding: Yes

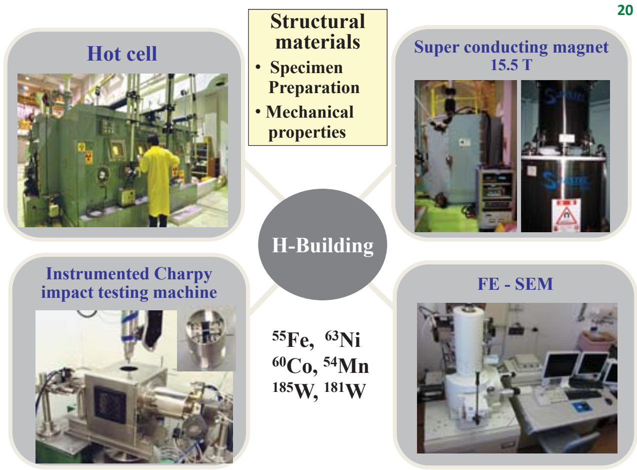
Dose:
~ 2 dpa and hopefully also ~ 0.2 dpa

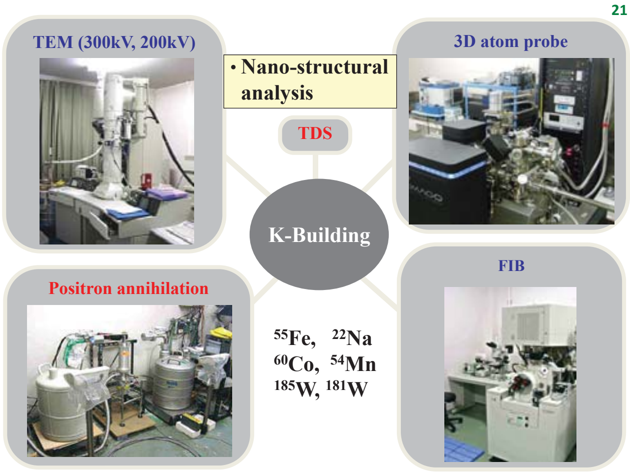
Materials:
W, W coatings on RAFM steels, TFGR W, W alloys and composites

PIE:
Microstructures, Mechanical properties (toughness & strength), Thermal conductivity, D/T retention at high temperatures, D/T permeation etc.

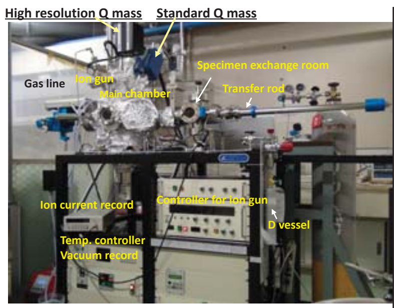
4. Neutron-irradiation program in the International Research Center for Nuclear Materials Science (IRCNMS), Institute for Materials Research, Tohoku University







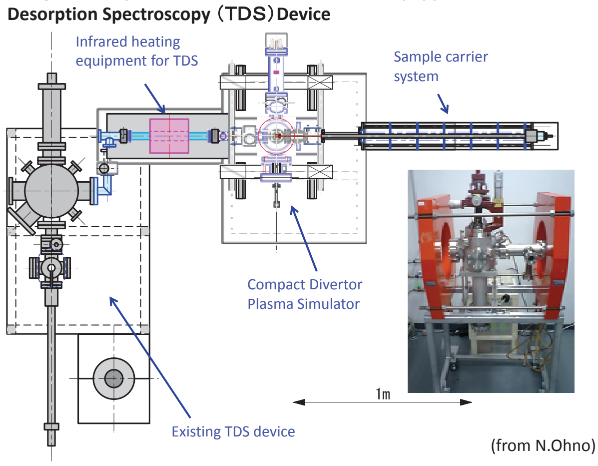
TDS installed in the radiation restricted area, Tohoku University



Ion gun
- D^{2+}, He^{2+}
- Beam dia.: ~ 3 mm
  No need of scan
  Energy: 0.3 ~ 3 keV
  Fluence: < 10^{23} D^{2+}
- Faraday Cup
  Specif. : 5Φ, 20Φ
- Amount of implanted ion
  = Fluence: current value at Faraday cup
- Current value
  Approximately constant through ion irradiation
- Irr. Temp.: RT~1000°C

TDS experiment : RT ~ 1000°C, 0.5~1°C/s
~ 3.0 x 10^{-7} Pa

Design of Compact Divertor Plasma Simulator Equipped with Thermal Desorption Spectroscopy (TDS) Device

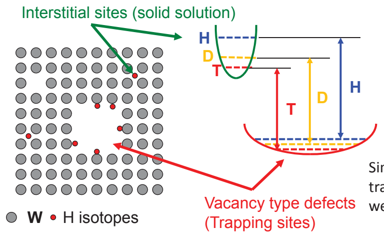


(from N.Ohno)

Research Plan in IRCNMS (Oarai Facility), IMR, Tohoku University

- ✓ The nearest neutron irradiation in Oarai Facility will be done in BR-2 reactor at SCK·CEN, Belgium. No thermal neutron shield.
- ✓ Hatano is going to order irradiation of W and W-5Re alloy at 290 °C to 1×10^{24} n m^{-2} (> 1 MeV).
- ✓ BR-2 reactor will be unavailable in 2015.
- ✓ A part of TITAN specimens are already in Oarai, and PHENIX specimens will be shipped also after neutron irradiation and appropriate cooling.
- ✓ Hatano is slowly constructing a gas loading device.
- ✓ The type of defects playing dominant roles in trapping may be identified with TEM observations and positron annihilation spectroscopy.
- ✓ Effects of transmutation elements could be understood by comparison of W and W-5Re alloy.

5. Tritium experiments in Hydrogen Isotope Research Center (HRC), U. Toyama



Vibrational states of H isotopes in interstitial sites and vacancy type defects

Simple model shows that the trapping in vacancy type is weaker for heavier H isotope.

Is it true?

Comparison of H, D and T is necessary.

Hydrogen Isotope Research Center (HRC), University of Toyama

HRC is one of the largest tritium research facilities in Japanese universities and licensed to handle 8 TBq (217 Ci) tritium per day and 555 TBq (15 kCi) per year.

The center was established at 1980, and this year is in celebration of 30 years anniversary. Safety equipment including ventilation, waste water processing and tritium monitoring systems were fully reconstructed 4 years ago.

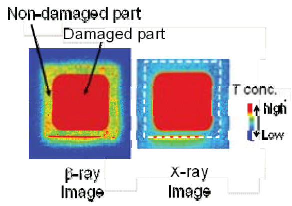
3 Full, 3 associate and 1 assistant professors,
1 Posdoc, 2 Technicians, Secretaries

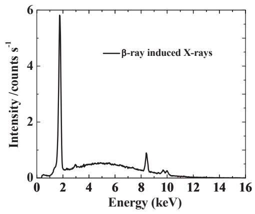
Uniqueness of Facility

(1) Handling of tritium in any chemical/physical form
(2) Various instruments for tritium measurements (gas, liquid and solid)
(3) Various tools for material characterization in tritium laboratory.

Research Plan in HRC, U. Toyama

- ✓ Defects are induced in W by heavy ion irradiation.
- ✓ The specimens were exposed to H_2, D_2 or T_2 (or high conc. T-D and T-H gas).
- ✓ TDS measurements.
- ✓ Isotope exchange for T.





Non-destructive measurements of T retention is possible using imaging plates and β-ray induced X-ray spectroscopy (BIXS) for ion-damaged tungsten.

6. Summary

- ✓ Results of TITAN project clearly showed the strong effects of neutron irradiation on hydrogen isotope retention in W.
- ✓ Hydrogen isotope retention at high temperature is determined by trap density and trapping-detrapping equilibrium.
- ✓ Isotope exchange appears to work for detritiation.
- ✓ Effects of high temperature neutron-irradiation will be examined in PHENIX project.
- ✓ Defect characterization will be performed in IRCNMS (Oarai Facility), IMR, Tohoku University.
- ✓ Isotope effects on trapping will be examined in HRC, U. Toyama.