



Tritium penetration by isotope exchange in tungsten

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Introduction

- Tritium (T) transport in tungsten (W) will be governed by its interaction with other isotopes due to the fact that tritium will always diffuse in the presence of deuterium (D) in a fusion device.
- Solute hydrogen isotopes have different diffusion coefficients as a consequence of their different masses. In its simplest non-quantum mechanical form, absolute rate theory yields:

$$\frac{D_\alpha}{D_\beta} = \left(\frac{M_\beta}{M_\alpha}\right)^{1/2}$$

- where α and β designate the two different isotopes and M_α and M_β are the isotopic masses.
- However in the temperature range of interest (300-1000 K), the effect of trapping must be considered and the evolution of the trapped tritium is of primary interest.

Purpose / Approach

The goal of our investigations is to understand how trapped tritium is transported and evolves in the presence of other isotopes.

- We utilize the far more sensitive radioactive tracer method using tritium and serial sectioning of the sample to obtain T depth profiles. The shape of the penetration profiles provide information of the effective diffusion rate.
- We perform complementary D-H isotope exchange experiments to determine the rate constants of D release by isotope exchanging with Hydrogen (H).

Summary/Conclusions

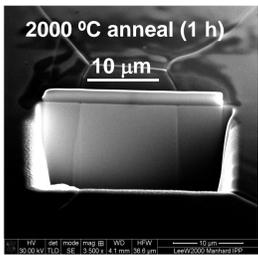
- The experimental results are summarized as follows:
 - T-penetration experiments:
 - The activation energy for diffusion ranges between 0.2-0.4 eV and is weakly dependent on the level of traps filled at $T < 450$ K.
 - D-H isotope exchange experiments:
 - Assuming the rate limiting step is diffusion/trapping/de-trapping process – effective diffusivity values are in the same order of magnitude with similar temperature dependence.
- These results suggest that a single characteristic rate constant can describe how trapped hydrogen isotopes evolve in the presence of other isotopes and that such a process is weakly temperature dependent.

Acknowledgements

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W samples

Recrystallized W sample



- ALMT, Japan (≥99.99%)
- Hot rolled to thickness
- 7 × 7 × 1 mm
- Surface polished to mirror like ($R_a \leq 10$ nm)
- Annealed at 2273 K in vacuum
- Results in parallel grain boundaries

1) Tritium penetration experiment

Performed at Hydrogen Isotope Research Center, Toyama U. Japan

Tritium loading by gas

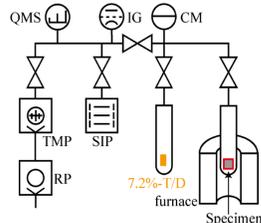
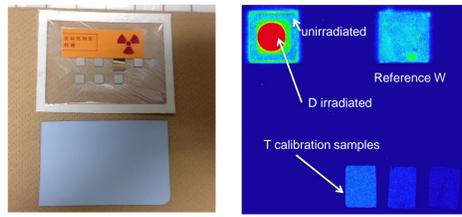


Image Plate Technique (residual T activity)



Sample sectioning by etching and mass loss

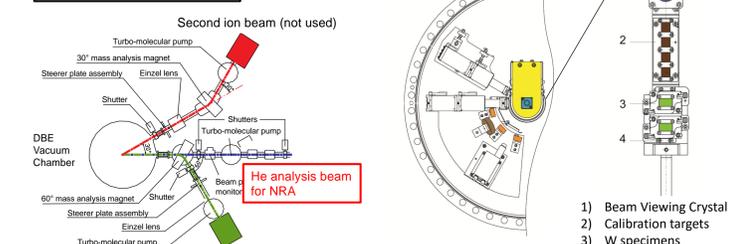
- H_2O_2 solution to dissolve W
- Caustic alkali (NaOH) with an oxidizing agent (H_2O_2) was used to etch samples
- Depth scale determined from mass loss
- $T_{etch} = 25$ °C

- D/T mixed gas exposure (7.2% T)
- Pressure = 1.2 kPa
- $T_{exp} = 200$ °C (3h), 300 °C (3h)
- Prior to D/T exposure samples are held at T_{exp} for 1 h in vacuum.

2) D-H isotope exchange experiment

Performed at Max Planck Institute, Garching, Germany

- D → W / NRA analysis
- H → W / NRA analysis
- Repeat step 2

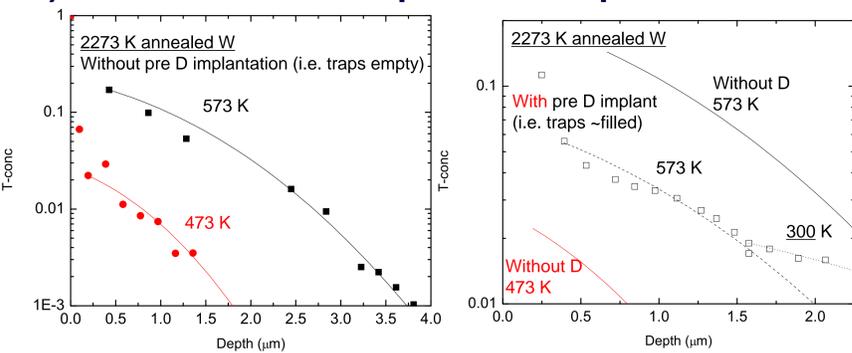


DBE device

- in-vacuo IBA (NRA/RBS etc)
- Beam mass analyzed (H_3^+ , D_3^+)
- $V_{acc} = 9$ keV
- Flux = $8 \times 10^{17} m^{-2}s^{-1}$
- Fluence = $\sim 1.5 \times 10^{22} m^{-2}$
- $T_{irr} = 150-450$ K

Results and Discussion

A) Normalized Tritium penetration profiles

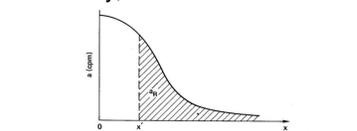


- Samples without and with D pre-implantation.
- Penetration profiles obtained following T-exposure at 473 /573 K.
- The samples were stored in Ar-atmosphere for a year to age at 300 K - then subsequent penetration profiles were further obtained.
- Data is fitted by two error functions - first part is diffusion during T loading at 473/573 K and the second part due to diffusion at 300 K.

The concentration profile of a tracer that is initially deposited as an infinitely thin planar source and subsequently allowed to diffuse from a free surface into the specimen is:

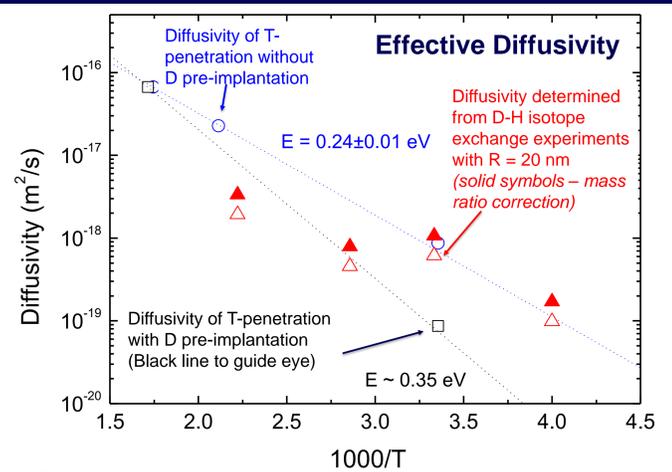
$$\frac{c(x, t)}{c_0} = \frac{1}{2\sqrt{\pi Dt}} \exp\left(-\frac{x^2}{4Dt}\right)$$

However since we are measuring the residual activity,



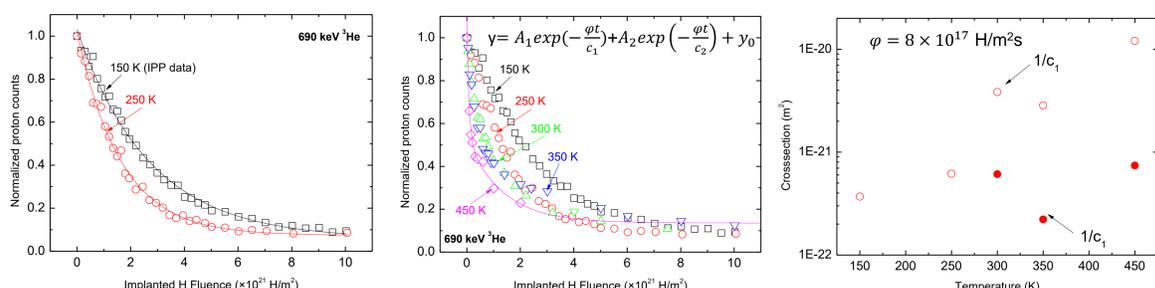
an error function solution can be applied to the data to obtain values of diffusivity, D:

$$\frac{a_R}{a_0} = \frac{1}{2} \operatorname{erfc}\left(\frac{x}{x\sqrt{Dt}}\right)$$



- T-penetration experiments:** values are order of magnitudes smaller than Frauenfelder or others – expected since we are measuring trapped not solute concentrations. The activation energy for diffusion ranges between 0.2-0.4 eV.
- D-H isotope exchange experiments:** Assuming the rate limiting step is diffusion/trapping/de-trapping process – effective diffusivity values are in the same order of magnitude with similar T dependence.

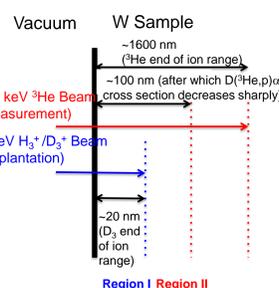
B) D-loss by H implantation: first-order process Fitted rate constants Ranges



- Experimental data indicates D-loss is a first-order process with exponential decay statistics.
- At $T > 250$ K the data is better fitted by a double exponential function, which may indicate two first-order processes.

- The rate constants can be determined by fitting using an exponential function.
- Two constants at $T > 250$ K
- Units of cross section in m^2 .

- The ion range for the 9 keV H and D beam is ~ 20 nm.
- The probed depth by NRA is ~ 100 nm.



- The probability D atom has not escaped in time t is given by:

$$p_{survival}(t) = \exp(-k_{total}t)$$

- Where k_{total} is the total escape rate which includes all pathways of escape.

- Taking the negative of time derivative of $p_{survival}(t)$ gives the probability distribution function $p(t)$ for the time of first escape:

$$p(t) = k_{total} \exp(-k_{total}t)$$

- The average time for escape τ is simply the first moment of $p(t)$:

$$\tau = \int_0^\infty t p(t) dt = 1/k_{total}$$

- Since the implanted H fluence is ϕt where ϕ is the H-flux in units of m^2s^{-1} , the relationship between the experimentally fitted decay constant c and τ is:

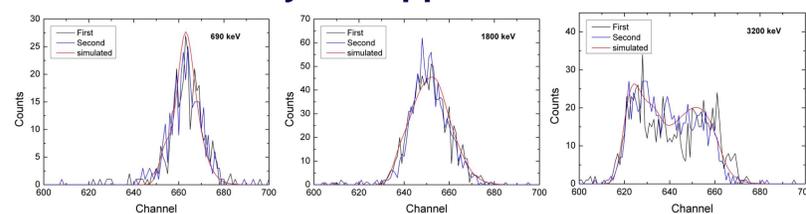
$$\tau = \frac{c}{\phi}$$

- Using the fundamental concepts of linking random walk to diffusivity, the most probable value to move a distance, R is given by:

$$R^2 = 2Dt$$

- Using the implanted ion range $R = 20$ nm, we can calculate the effective diffusivity values

Thermal stability of trapped D at 450 K



- Nuclear reaction analysis measurements with different energies was performed twice after D implantation (0.69, 1.8, 2.4, 3.2, 4.5 MeV).
- The measured D content does not change after energy scan (90 min)
- Confirms that D is stable at 450 K and NRA measurements are not perturbing the system.