Deuterium permeation in tungsten by mixed ion irradiation

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Outline

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  - General explanation of tritium behavior in wall materials
- Experimental details
  - Permeation apparatus
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- Pure D ion driven permeation
  - Microstructure (annealing temperature) dependence
- He/D mixed ion driven permeation
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- Discussion and Summary
Introduction

- T retention and permeation to coolant in blankets need to be studied for evaluation of T economy and safety.
- Tungsten is a leading candidate of armor materials of blankets.
- Tungsten has low diffusivity and solubility of T, and relatively high trapping energies of defects.
- Therefore, tungsten armor on reduced activation materials of blankets (Ferritic steel, V alloy) can be permeation reducer to coolant.
- However, still basic parameters to determine permeation such as effective diffusivity, recombination coefficient etc. are not well determined.
- In addition, effects of mixed ion irradiation (D+He,C) are not known well either.
Tritium behavior in a first wall

Intrinsic traps

T+(D+)

Reflection

Plasma

Coolant

T density
Tritium behavior in a first wall

$T^+$

Reflection

Plasma

Coolant

Intrinsic traps

Neutron induced traps

$T$ is trapped near surface damage sites (intrinsic and ion-induced)

T density
Tritium behavior in a first wall

- Near surface layer is saturated.
- Most of implanted T is desorbed from the surface.
- Small fraction of implanted T diffuses into the bulk.
- Bulk T is trapped at intrinsic or neutron induced traps.

T trapping at n-induced traps
Tritium behavior in a first wall

As temperature increased, trapped T decrease, but still **traps could affect effective diffusivity**
Necessary information for T behavior

- **Surface damage effects (How they affect T retention and release)**
  - Recoil damage effect
  - Effects of bubbles and blisters (due to oversaturation etc.)
  - Impurity mixing effects (He, wall impurities (C, Be etc.))

- **Effective diffusion mechanism (How they affect T transport)**
  - Trap sites effect
  - Microstructure effect (intergranular diffusion, grain boundary diffusion)

- **Neutron induced traps (How they affect T retention)**
  - Trapping energies
  - Production rate (as a function of dpa)
  - Saturation level with respect to dpa
Ion driven permeation model (Brice & Doyle)

Penetration
\( \phi_i (1 - R_e) \)

Recombination (recycling)
\( J_1 = 2k_1 C_1^2 \)

Recombination (Permeation)
\( J_2 = 2k_2 C_2^2 \)

Diffusion

Incident flux \( \phi_i \)

PLASMA

MATERIAL

IRRADIATION SURFACE

COOLANT

Permeation flux \( \phi_p \)

Solute hydrogen concentration

(a) Recombination limited condition :
\( \phi_p = \frac{D_2}{x_0 \sqrt{k_1}} \sqrt{\alpha \phi_i} \)

(b) Diffusion limited condition :
\( \phi_p = \frac{RD_2}{x_0 D_1} \alpha \phi_i \)


ISFNT10, Y. Ueda et al., September, 2011
Experimental setup
**Experimental device**

**HiFIT Device**

- Deflection coil
- Q-mass analyzer
- IR Heater (2kW)

**Permeation Device**

- Base pressure: \(~3 \times 10^{-7}\) Pa
- ECR plasma
- Magnetic coils
- Gas inlet
- Three spherical electrodes

**Specimen**

- Purity: 99.99%
- Annealed: 1573 K for 1h
- Thickness: 75 µm, 30 µm
- Roughness: \(R_a = 0.01\) µm
- Temperature: \(\leq 1050\) K

**Ion beam**

- Energy: 0.15 keV-3 keV
- Flux (D): \(~10^{20}\) m\(^{-2}\)s\(^{-1}\)
- Species: \(D_3^+, D_2^+, D^+\)
- Impurity (C): 0.1~0.2%

**Blanket first wall conditions**

- Tungsten
- Quartz Rod
- 8mm
- Thermo couple

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ISFNT10, Y. Ueda et al., September, 2011
Tungsten specimens

- **W1**: Sintered polycrystalline W → annealed at 1573 K.
  - Standard sample in our experiments (medium grains and low density defects)

- **W2**: Sintered polycrystalline W → annealed at 2273 K.
  - Large grains and almost no defects

- **W3**: Sintered polycrystalline W → annealed at 1203 K.
• Weak grain boundary dependence (factor of two)
• Peak in permeation flux observed $T \approx 800$ K

$E = 1$ keV

$\phi_i = 1.5 \times 10^{20}$ D m$^{-2}$ s$^{-1}$

normalized thickness: 25 $\mu$m

H. T. Lee et al., Phys. Scripta., accepted
Effective diffusivity values determined from lag time measurements.

There are some dependences on thickness and microstructure.

Effective diffusivities are close to Zakharov’s, but much less than Frauenfelder’s.

This could be the effect of some traps.

Need more investigation

He/D mixed ion driven permeation
He/D mixed ion driven permeation

- Addition of He (2%) greatly reduces permeation flux.
- Saturation time almost corresponds to He bubble formation time.

E = 1 keV

< 800 K more reduction (less than detection limit)

H. T. Lee et al., J. Nucl. Mater. (accepted)
Incident flux dependence

- $\phi_p \sim \phi_i$ (D only irradiation)
- $\phi_p \sim \phi_i^{1/2}$ (D/He irradiation)

  - $\phi_p$: Permeation flux
  - $\phi_i$: Incident flux

- Change of flux dependence suggests D release from the front surface could change from diffusion limited (D) to recombination limited (D/He).

- Two possibilities
  - Diffusion increased.
  - Recombination decreased.

E = 1 keV He:2%

Permeation flux $\phi_p$ vs. Incident flux $\phi_i$
Depth profile of He bubble layer

- He bubble layer was observed up to the depth of about 30 µm.
- Thickness of He bubble layer was larger than ion range (~10 µm).
- He bubbles are interconnected to form
He/D effects on retention and blistering

- Blistering disappeared (or reduced) for He/D irradiation.
- D retention greatly reduced for He/D irradiation.

PISCES results (573 K)


V. Alimov et al., 12th ITPA (SOL/DIV)
C/D mixed ion driven permeation
C/D mixed ion driven permeation

E = 1 keV

![Graph showing permeation flux vs irradiation time for D-only and D+C cases.]

- The steady state D permeation flux for simultaneous D+C case is **20 times larger** than that of D-only case.
- The lag time for simultaneous D+C case is **1.5 times larger** than that of D-only case.

Tw = 620 K
Temperature & concentration dependence

- D permeation greatly increased even with C (>0.9%) in ion beam.
- Strong temperature dependence.
- Surface elemental composition shows little dependence on temperature (C:1.4%). → only temperature affect change of parameters
- Maximum increment factor in the present exp. : ~250

Energy : 1 keV D, Ion flux ~10^{20} m^{-2}s^{-1}

Temperature dependence of permeation

Near surface atomic composition

H.Y. Peng, Phys. Scripta., accepted
How C/W mixed layer affect permeation

- Surface carbon concentration was determined by the balance between re-erosion and implantation of C ions.
- Either significant reduction in surface recombination (more than $10^{-4}$) or reduction in diffusion ($10^{-2}$) could cause significant increase in permeation.

**Diagram:**
- **Pure D irradiation**
- **C/D mixed irradiation**

**Equations:**
- $D_f \frac{\partial C}{\partial t}$
- $D_b \frac{\partial C}{\partial t}$
- $kr_1 C_1^2$
- $C_1$
- $C_2$
- $kr_1 C_2^2$
- $0$ to $d$ on the x-axis.

**Graph:**
- Comparison of permeation between irradiation side and permeation side.
Effect of C/W mixed layer on blistering

- For high carbon concentration in the beam (C:0.9%), blister appeared.
- C/W mixed layer effectively increases T diffusion into the bulk over certain fluence.
Mechanism for blistering

Implantation of H (a few nm ~ 20 nm)

Accumulation of H at grain boundaries

Cross section of blister (K-dope W)

Diffusion of D into the bulk is necessary for blistering.
Enhanced bulk diffusion by the C/W layer causes blistering.
An opposite effect was observed for He/D.
Planar DC magnetron plasma

- Energy: ~200 eV (D$_2^+$ mainly)
- Flux: $1 \times 10^{21} \text{ m}^{-2}\text{s}^{-1}$
- C plate on cathode surface to provide C into plasma

For D+C, D retention near surface (a) and bulk (b) increased at elevated temp.

D retention in W exposed to pure D plasma (□▽) and D+C plasma (▲■)

Comments on T behavior in blankets

- Surface phenomena greatly affect whole T behavior in blankets.

- Permeation barrier increases T concentration in materials
  - To reduce T permeation by putting permeation barrier on the permeation side, solute T density increase. Does it affect material performance?
  - $10^{-5}$ D/W for C/D irradiation (1.4%C, 800 K) (Assumption: Zakharov’s effective diffusivity)
  - Trapped T density (equilibrium with solute T) increases with the increase in solute T.

![Diagram](image)
Summary on D behavior for mixed irradiation

Mechanism of permeation reduction (He/D) and enhancement (C/D)

- **Addition of He**: Effective diffusion near surface area increased. Probably due to dense He bubble structure.
- **Addition of C**: Recombination or diffusion reduced: under investigation.

D: diffusion limited
R: recombination limited

~10² difference in permeation flux

~10³ difference in permeation flux
Concluding summary

- Mixed ion irradiation (He/D and C/D) into tungsten significantly changes permeation.
  - Addition of He: decrease \(10^{-1}\) at 800 K, and more at <800 K
    - Correlation with blister suppression
    - Correlation with decrease in retention
  - Addition of C: increase by two orders
    - Correlation with blister enhancement
    - Correlation with increase in retention
  - Strong temperature dependence
    - He or C effect significant at \(\sim 800\) K
      - close to blanket surface temperature (FS based)
  - Indicating importance to include material mixing effects in T modeling in wall materials

- Microstructure dependence on permeation
  - No significant effects of microstructure (W1, W2, W3) on steady-state permeation

Preferable for 1st wall