Hydrogen isotopes permeation through the tungsten-coated F82H first wall

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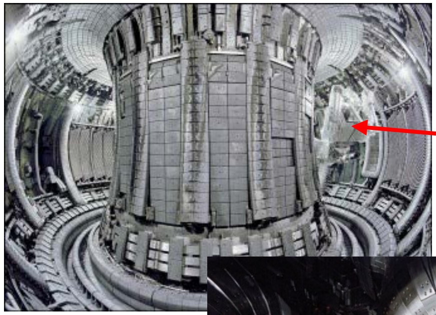
4. Summary

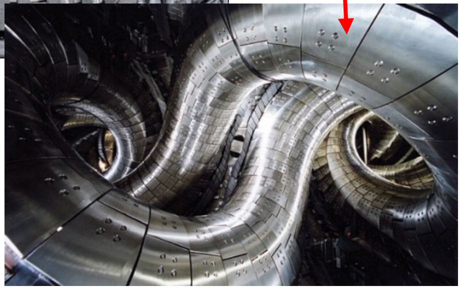
1. Motivation

The "First Wall" of a fusion power reactor

- **Definition of the first wall:**
  - ✓ **All the fusion experimental devices:** ***a vacuum chamber wall.***
  - ✓ **Fusion reactors:** ***the plasma-facing surface of breeding blanket units.***



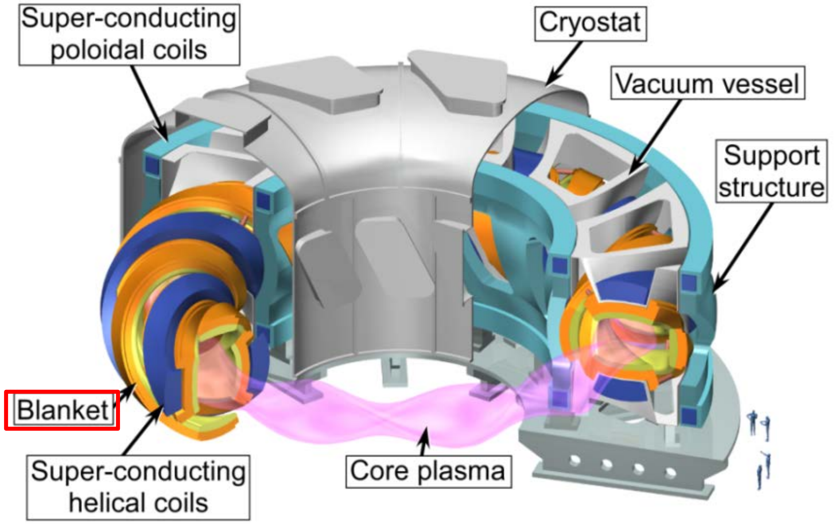
JET



First wall

LHD

The first walls of existing fusion devices.



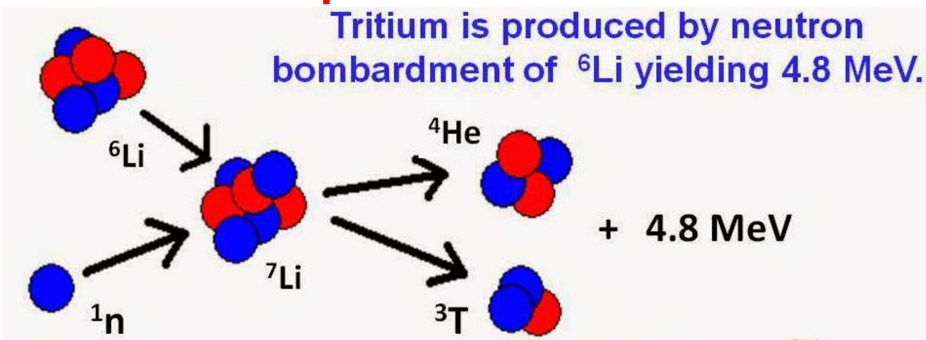
FFHR

(Force-Free Helical Reactor)

A. Sagara et al., research report, NIFS-MEMO-64 (2013).

Breeding blanket structure

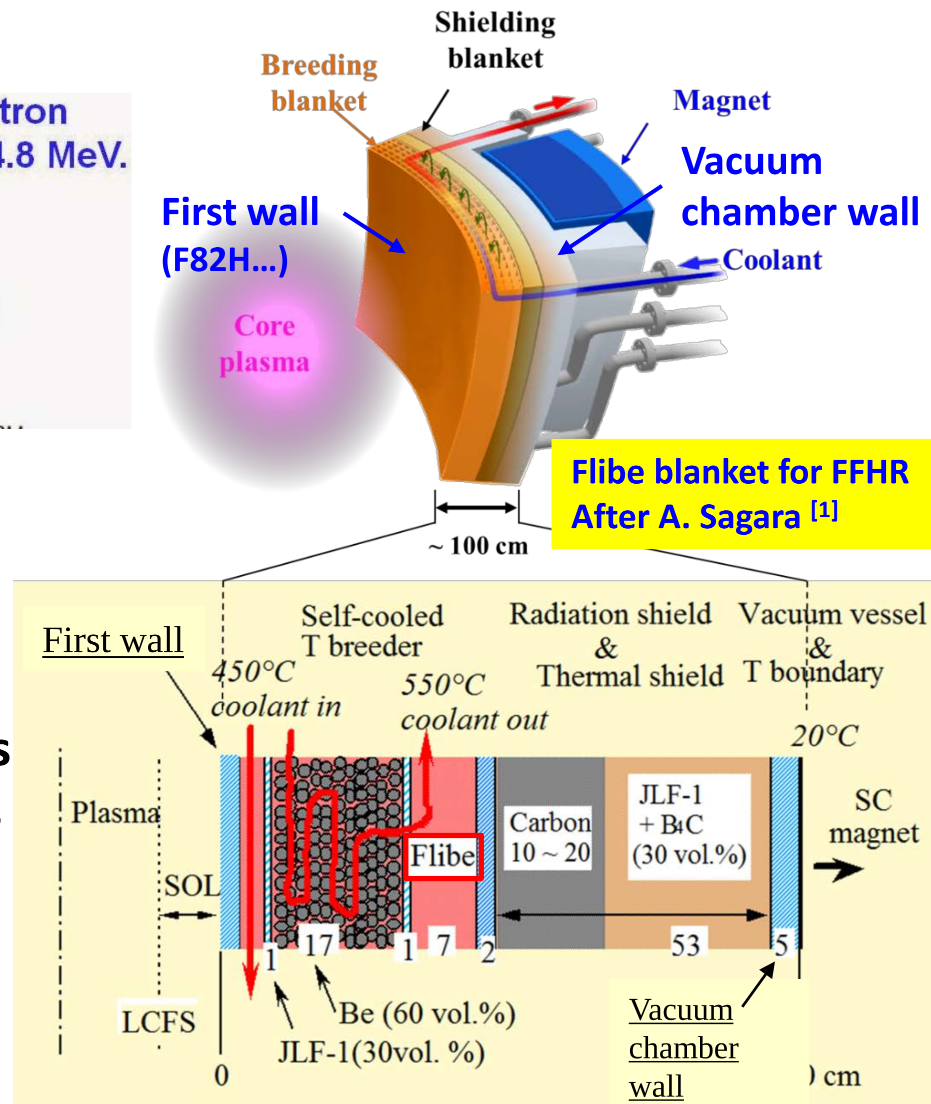
- **Blanket concepts include:**



- **Characteristics of the first wall of a fusion power reactor are:**

- **A large surface area: ~1000 m^2,**
- **Made of reduced activation materials (e.g. RAFS (F82H…), V-alloy, and SiC$_f$/SiC),**
- **Operated at high temperatures**

  (e.g. ~500 ℃ for ferritic steel alloys),
- **Thin wall design to reduce thermo-mechanical stress (~5 mm for RAFS).**



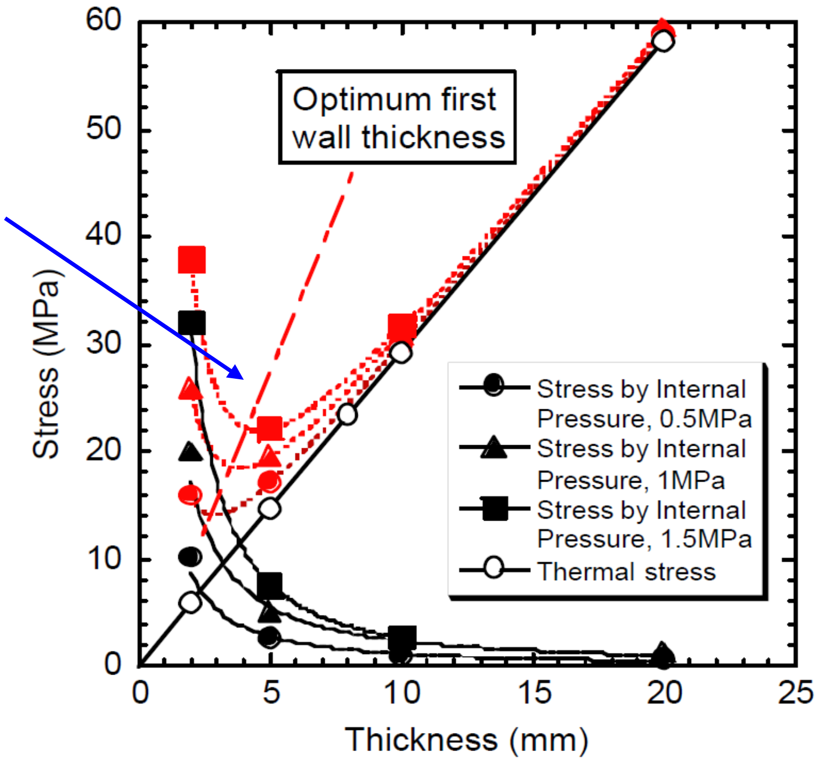
Flibe blanket for FFHR After A. Sagara [1]

[1] A. Sagara et al., Fusion Technol. 39 (2001) 753–757.

Optimum thickness of the first wall

- **Optimum first wall thickness: 5 mm or even less, although these concepts employ various first wall materials.**

Thickness optimization [2]



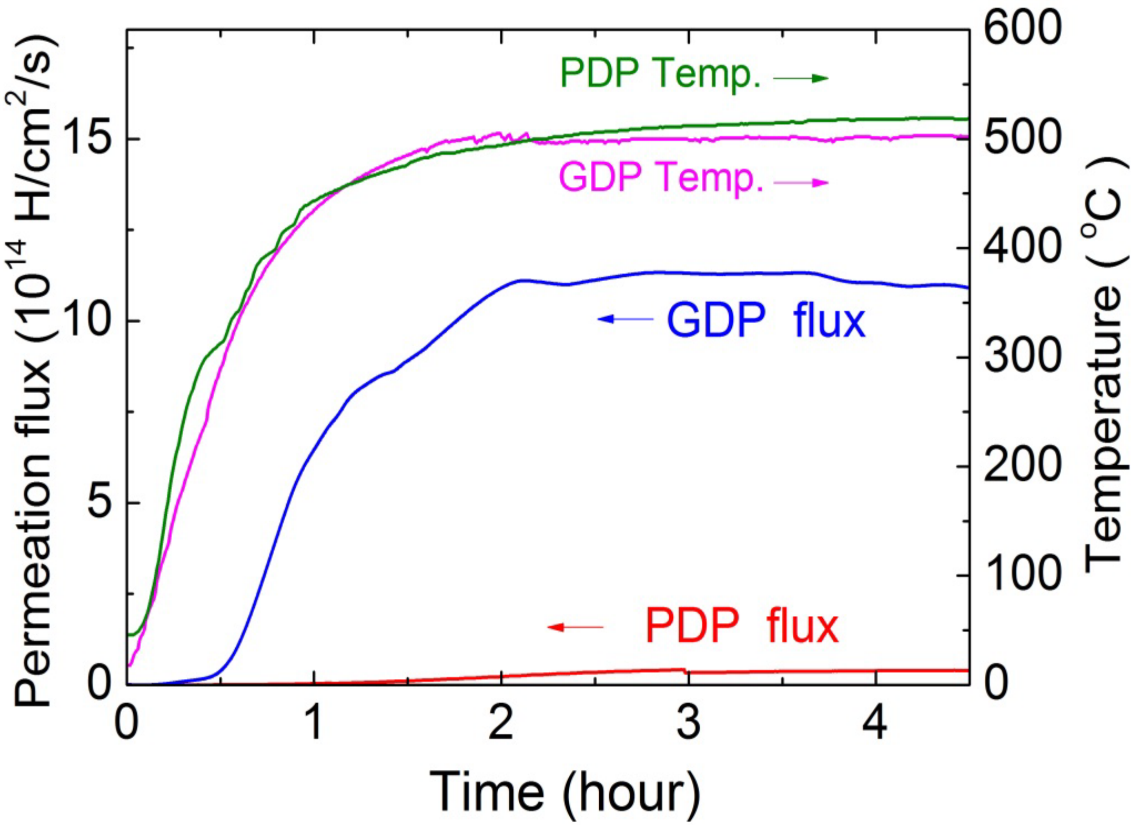
[1] Y. Ueda et al., J. Nucl. Mater. 313-316 (2003) 32-41.
[2] A. Sagara et al., Fusion Technol. 39 (2001) 753-757.

T_2 pressure in breeder

- **For blanket employing FLiBe, the tritium thermodynamic equilibrium pressure is ~10^4 Pa at 800 K at a tritium concentration (at%) of ~0.1 ppm (~12 g tritium for FFHR).**

~1 torr liter/s/m²

~1000 torr liter/s

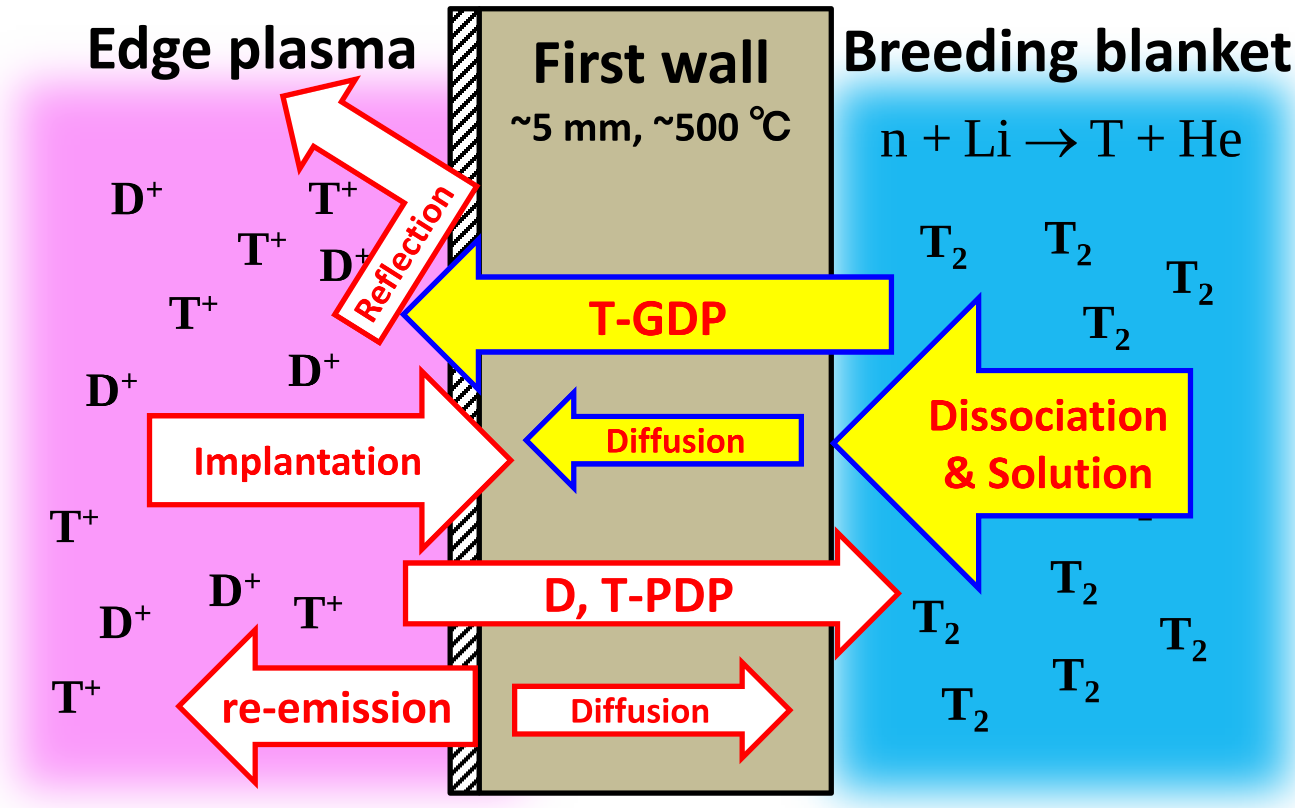


S. Fukada, et al., Fusion Eng. Des. 83, 747 (2008).

Bi-directional permeation

Do we want bi-directional permeation?

No!



1. W as a plasma-facing material [1,2]:
   - High melting point (3410℃)
   - High thermal conductivity
   - Low sputtering yield
   - Low activation
2. Applications:
   - Divertor: JET, EAST, ITER…
   - First wall: ASDEX-U…
3. W coatings methods:
   - Vacuum Plasma-Sprayed W (VPS-W)
   - SPutter-deposited W (SP-W)
   - Chemical Vapor Deposition W (CVD-W)…

PDP ⟷ GDP: Bi-directional permeation

[1] A. Sagara et al., Fusion Technol. 39 (2001) 753-757.
[2] R. A. Pitts et al., J. Nucl. Mater. 438 (2013) S48–S56.

2. Experimental facility and setup

Experimental facility

Plasma parameters:

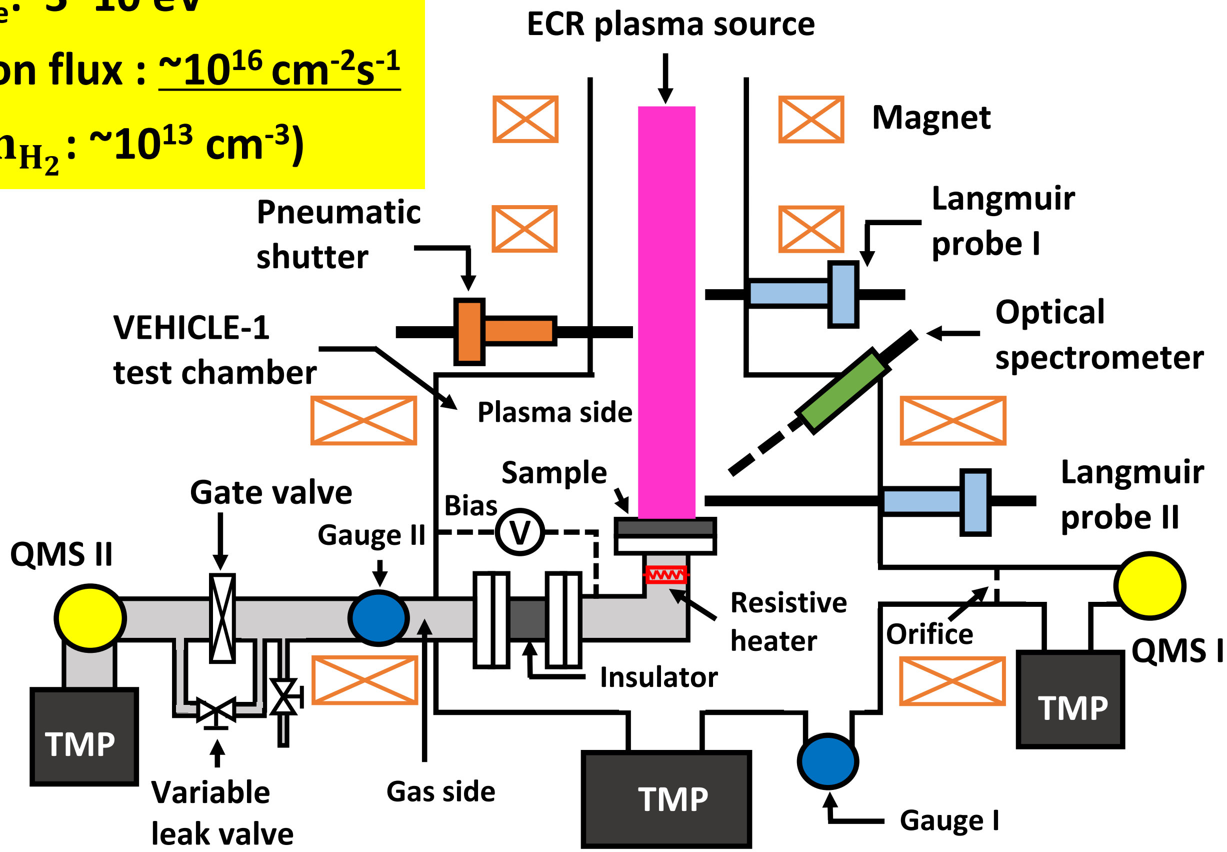
n_e: ~10^{10} cm^{-3}

T_e: 3~10 eV

Ion flux : ~10^{16} $cm^{-2}s^{-1}$

(n_{H_2}: ~10^{13} cm^{-3})

VEHICLE-1

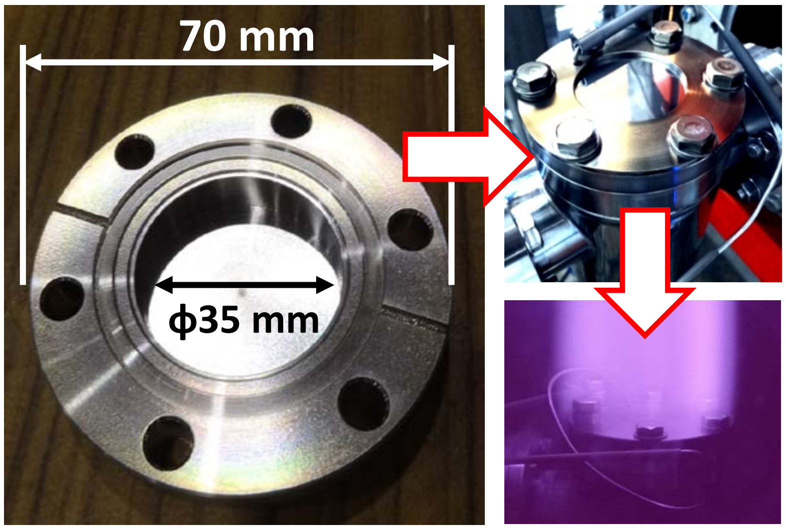


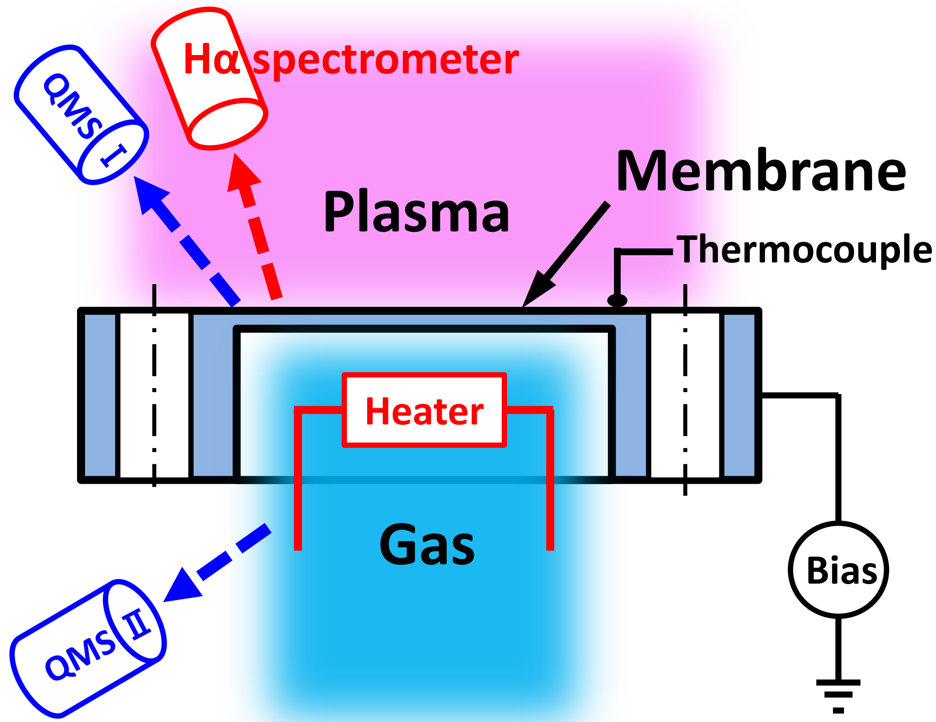
Y. Hirooka et al., J. Nucl. Mater. 337-339(2005)585-589.

QMS: Quadrupole Mass Spectrometer
TMP: Turbo Molecular Pump

Experimental setup

Permeation membrane sample





Samples:

- F82H (Fe-8Cr-2W)
- VPS-W coated F82H
- SP-W coated F82H

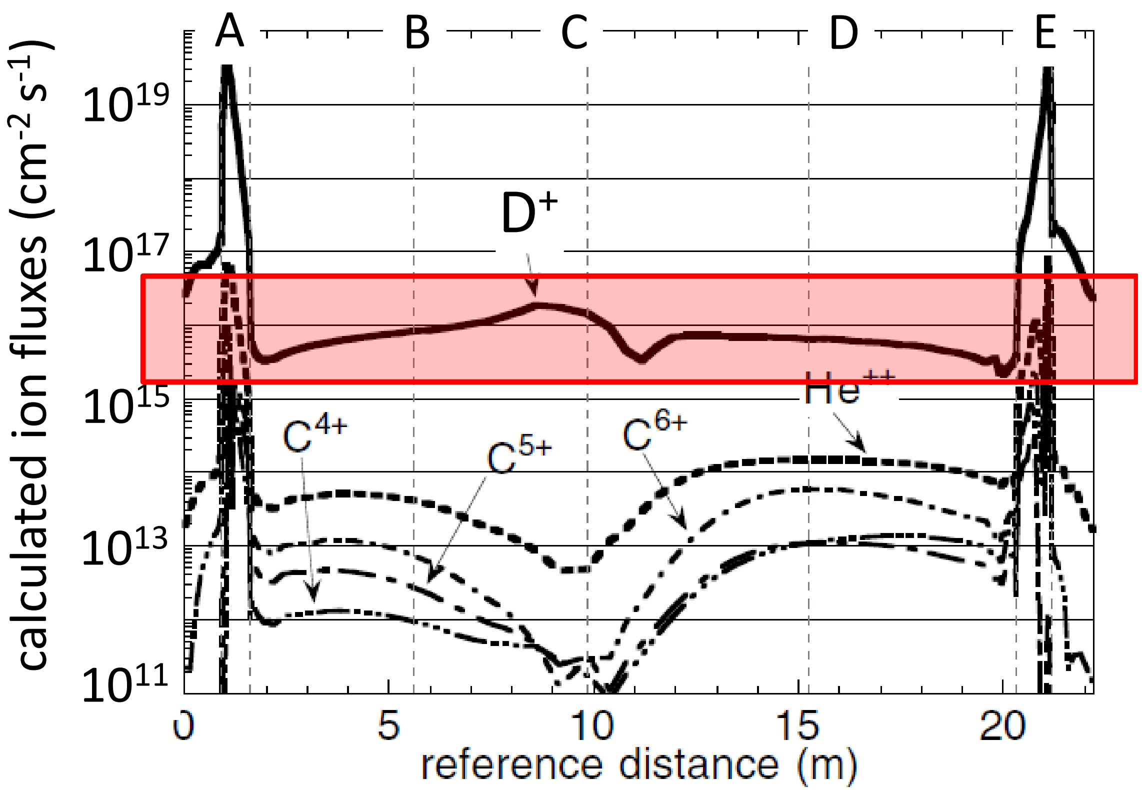
> F82H thickness: 0.5 - 2 mm
> VPS-W thickness: 50 - 200 μm
> SP-W coatings thickness: 0.5 - 4 μm

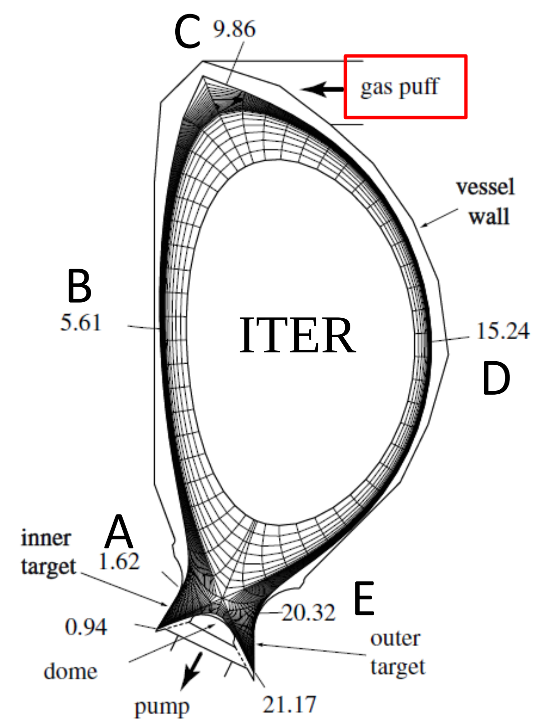
VPS-W: Vacuum Plasma-Sprayed W
SP-W: SPutter-deposited W

- PDP and GDP fluxes are measured by two QMS (Quadrupole Mass Spectrometer), respectively.
- Ion bombardment energy is provided by a DC bias (-100 V).
- Temperature: ~150 - 550 ℃.
- Argon plasma bombardment pre-treatment to reduce surface contamination.

Ion flux to the first wall

- D^+ ion flux to the first wall has been estimated to be of the order of 10^{16} D cm^{-2} s^{-1}.





R. Behrisch, et al., J. Nucl. Mater. 313–316 (2003) 388–392.

Gas-driven permeation (GDP) model (single layer)

Solubility of gases in metals (Sieverts' law):

$$C = S(T) \cdot \sqrt{p_{H_2}}$$

- $S(T)$: Sieverts' constant as a function of temperature;
- p_{H_2}: hydrogen pressure.

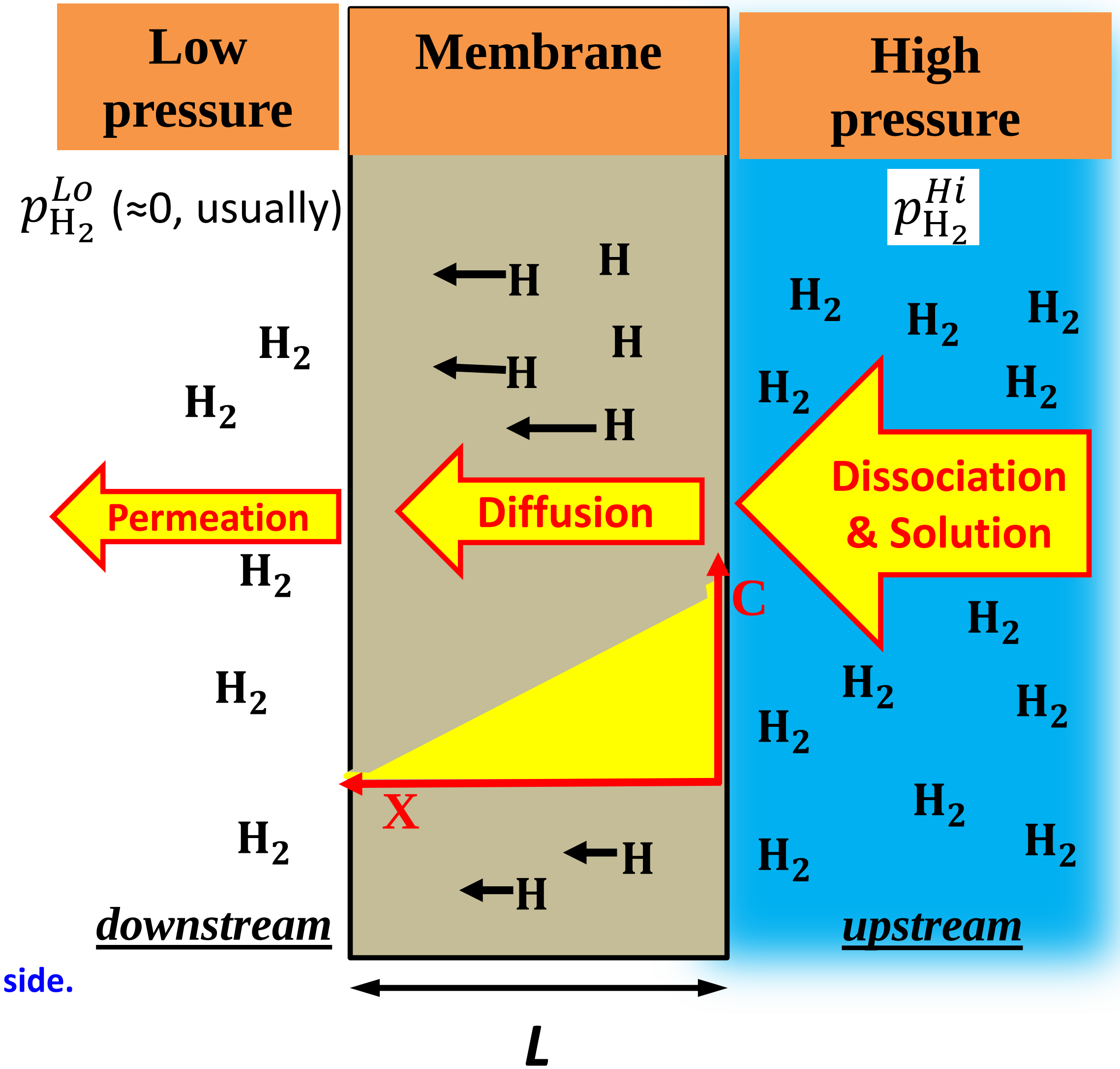
Diffusion limited gas-driven permeation flux:

$$J = -D(T)\frac{dC}{dx}$$

$$= \frac{D(T)S(T)}{L}\sqrt{p_{H_2}^{Hi}}$$

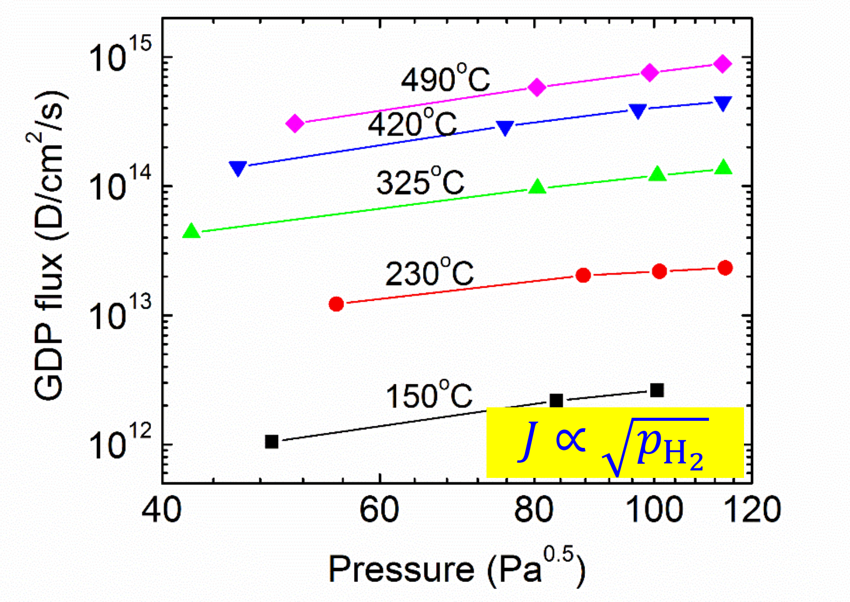
- L: membrane thickness;
- $D(T)$: diffusion coefficient;
- $p_{H_2}^{Hi}$: H_2 pressure at the high pressure side.

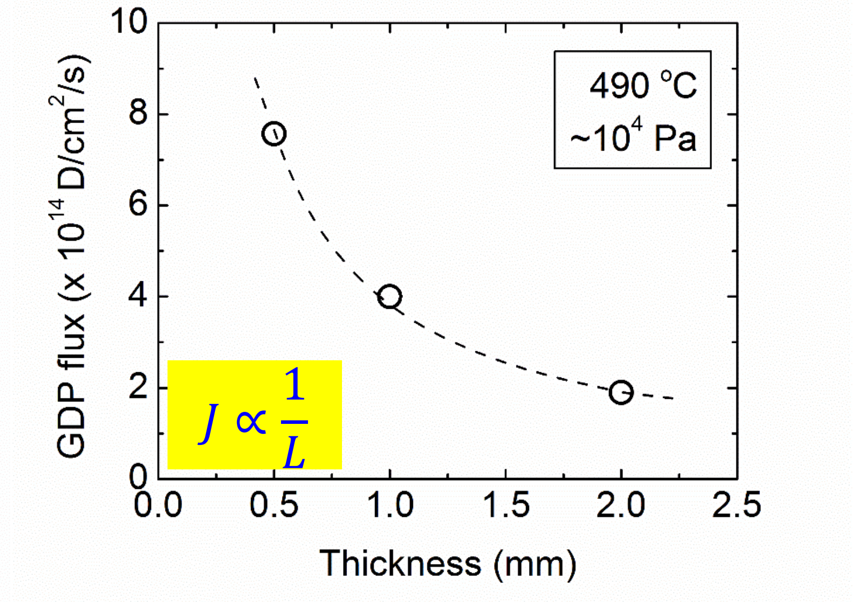
Permeability: $\Phi(T) \equiv D(T)S(T)$



F. Waelbroeck et al., J. Nucl. Mater. 85-86 (1979) 345-349.

F82H: pressure and thickness effects on GDP

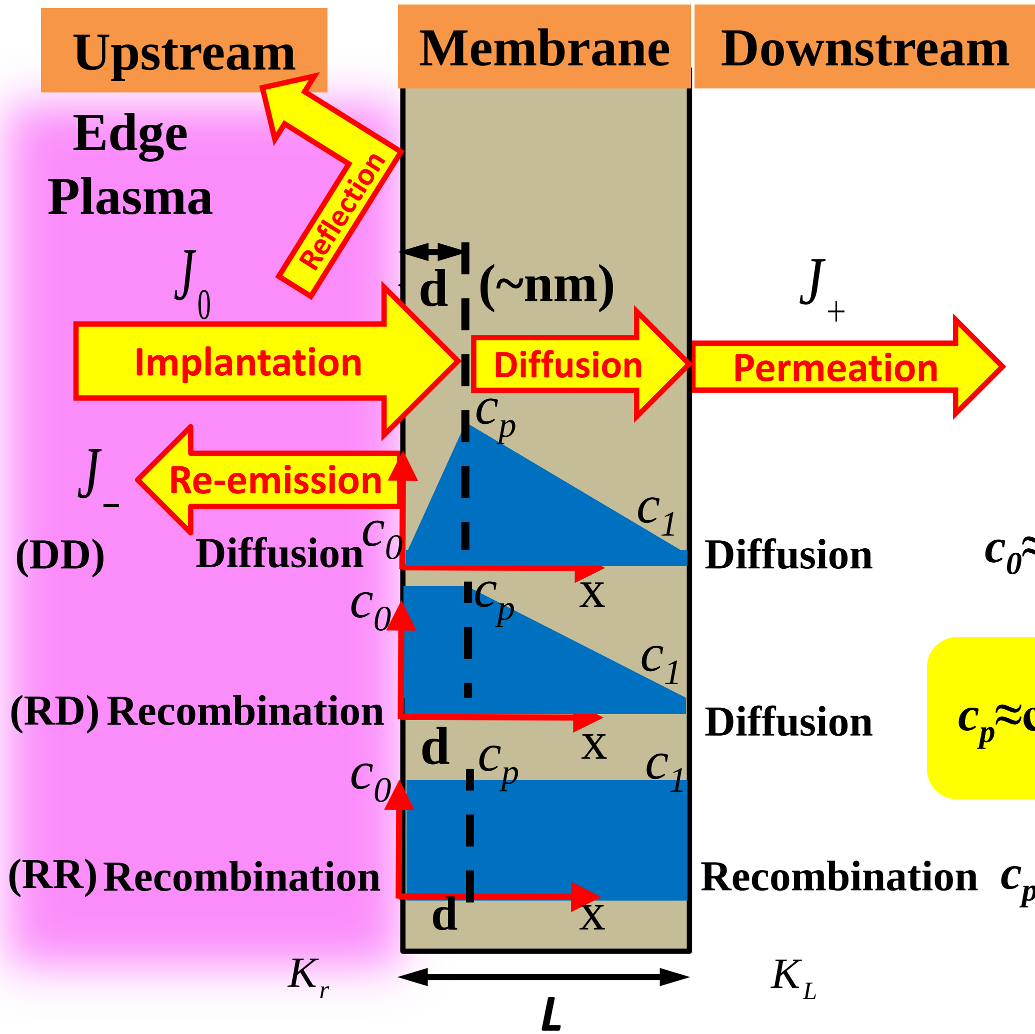




- **The GDP flux is proportional to the <u>square-root of upstream pressure</u>, and <u>inversely proportional to the membrane thickness</u>.**

GDP is diffusion-limited: $$J_{GDP} = -D(T)\frac{dC}{dx} = \frac{D(T)S(T)}{L}\sqrt{p_{H_2}}$$

Plasma-driven permeation (PDP) model



Particle balance:

$$J_0 = J_- + J_+$$

implantation re-emission permeation

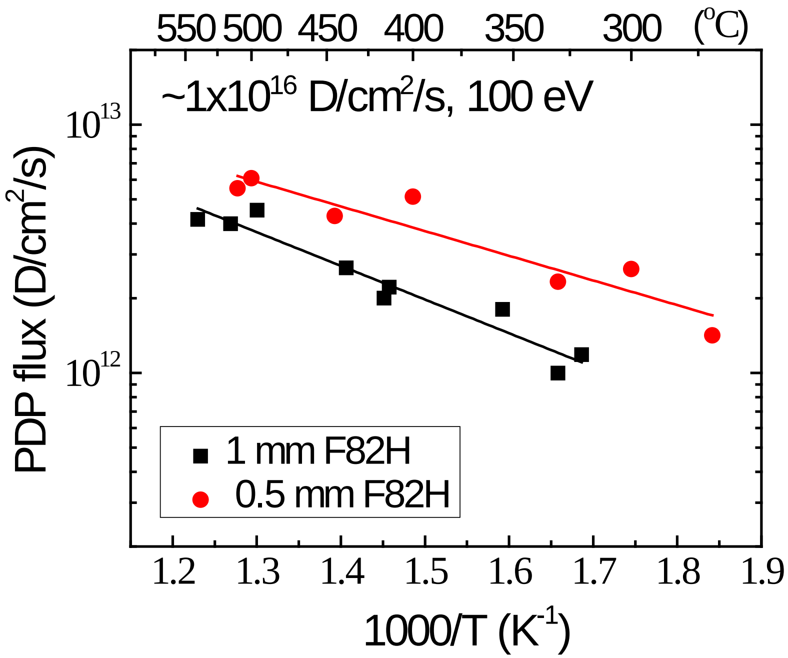
$c_0 \approx 0,\ c_1 \approx 0$ ⇨	$J_+ = \frac{d}{L} J_0$	$J_- \approx J_0$
$c_p \approx c_0,\ c_1 \approx 0$ ⇨	$J_+ = \frac{D}{L}\sqrt{\frac{J_0}{K_r}}$	$J_- = K_r C_0^2$
$c_p \approx c_0 \approx c_1$ ⇨	$J_+ = \frac{K_L}{K_r + K_L} J_0$	$J_- = \frac{K_r}{K_r + K_L} J_0$

- c_0 and c_1: H concentrations of the plasma side and back side;
- d: implantation range;
- c_p: H concentrations at x=d;
- K_r and K_L: H recombination coefficients.

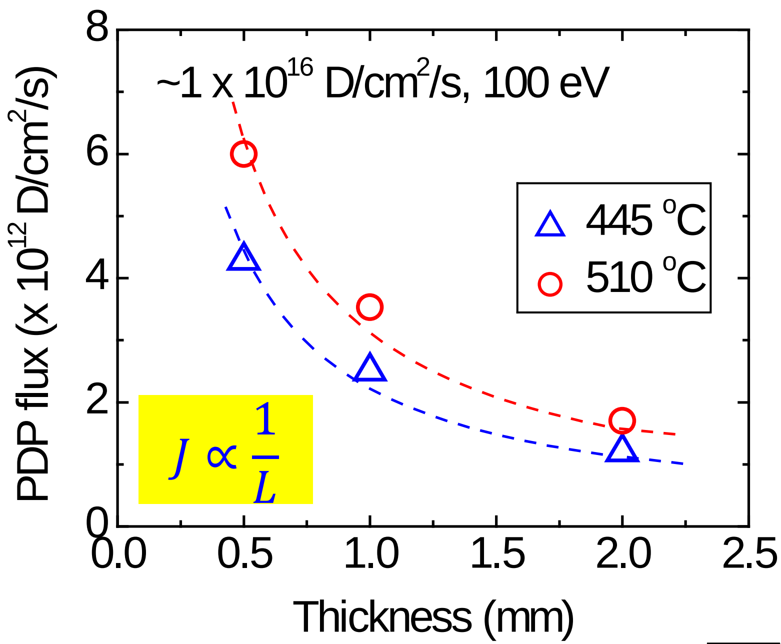
B. L. Doyle, J. Nucl. Mater. 111 & 112 (1982) 628-635.

F82H: temperature and thickness effects on PDP

PDP flux increases as temp. increases



PDP at 445 °C and 500 °C



PDP in RD-regime [1]: $J_+ = \frac{D}{L}\sqrt{\frac{J_0}{K_r}}$

- **The steady-state PDP flux is <u>inversely proportional to the membrane thickness</u>, meaning that hydrogen PDP is in the RD-regime.**

[1] B. L. Doyle, J. Nucl. Mater. 111 & 112 (1982) 628-635.

Permeation studies by modelling: DIFFUSE code

Hydrogen transport in solids can be described as:

Mobile hydrogen:

$$\frac{\partial C(x,t)}{\partial t} = D(T)\frac{\partial^2 C(x,t)}{\partial x^2} - \frac{\partial C_t(x,t)}{\partial t} + G(x,t)$$

Mobile — Trapped — Implantation profile

Trapped hydrogen:

$$\frac{\partial C_t(x,t)}{\partial t} = D(T)\frac{C(x,t)C_t^e(x,t)}{\lambda^2} - C_t(x,t)\nu_0 \exp(-U_t / kT)$$

$$C_t^e(x,t) = C_t^0(x) - C_t(x,t)$$

***C*(*x,t*) and *C*$_t$(*x,t*): concentrations of mobile and trapped; *D*: diffusion coefficient; *G*(*x,t*): hydrogen implantation profile; $C_t^0(x)$ and $C_t^e(x)$: concentrations of intrinsic and empty trapping sites; *λ*: mean distance between trapping sites; v_0: jumping frequency; *k*: Boltzman's constant; U_t: the de-trapping energy.**

Isotopic effects:

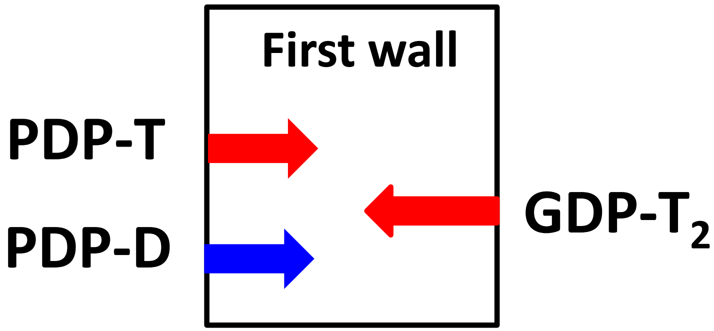
I. Isotope dependence of diffusivity, trapping energy, etc.

II. Coupling of isotopes through the process of surface recombination.

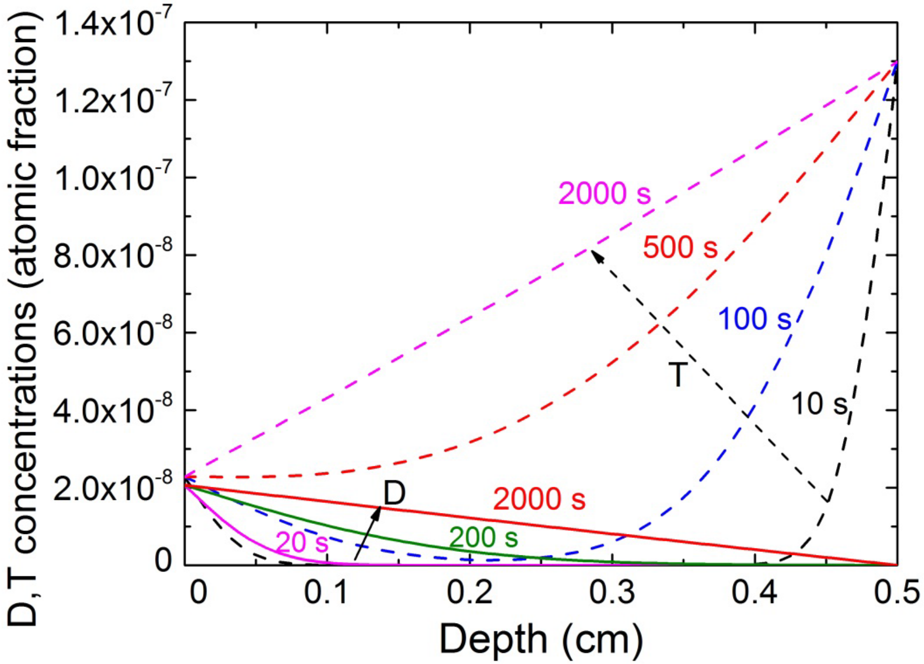
III. Competition of the various isotopes for traps.

One-dimensional modelling by DIFFUSE (2)

Bi-directional PDP-D/T and GDP-T_2 flows.

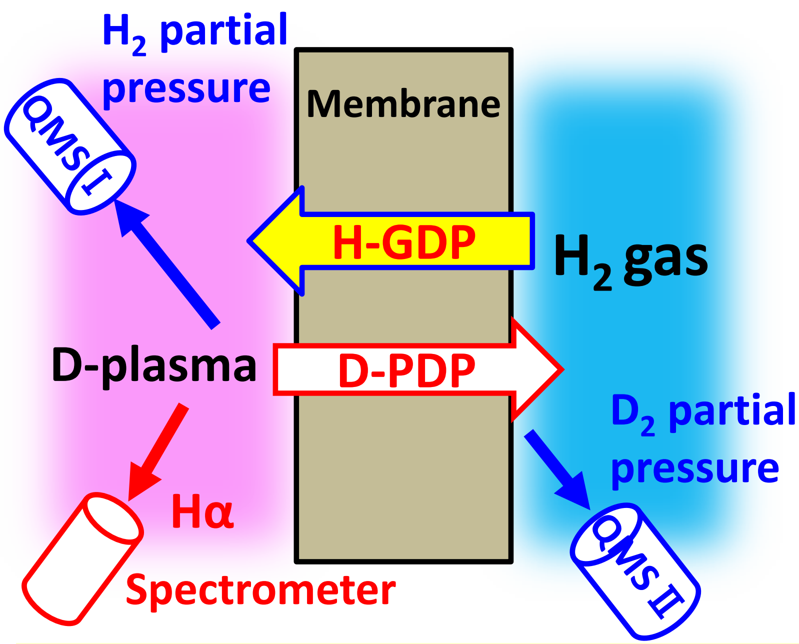


- GDP-T_2 pressure: ~1 Pa
- Plasma:
  5×10^{15} $D\cdot cm^{-2}\cdot s^{-1}$ 5×10^{15} $T\cdot cm^{-2}\cdot s^{-1}$
- Membrane: 5 mm thick α-Fe
- Temperature: 527°C
- Boundary conditions:
  Gas side: Sieverts' law
  Plasma side: recombination
- Intrinsic trap density: 1%
- Trapping energy: 0.62 eV



- The tritium concentration profiles interact with each other in the two counter flows.
- Deuterium flow appears to be independent of these tritium flows, driven by its own concentration gradient, i.e. "random walk" diffusion.

Demonstration of simultaneous bi-directional H/D permeation for the first time



D – PDP: T_e: ~10 eV

n_e: ~10^{10} cm^{-3}

Flux: ~10^{16} D cm^{-2} s^{-1}

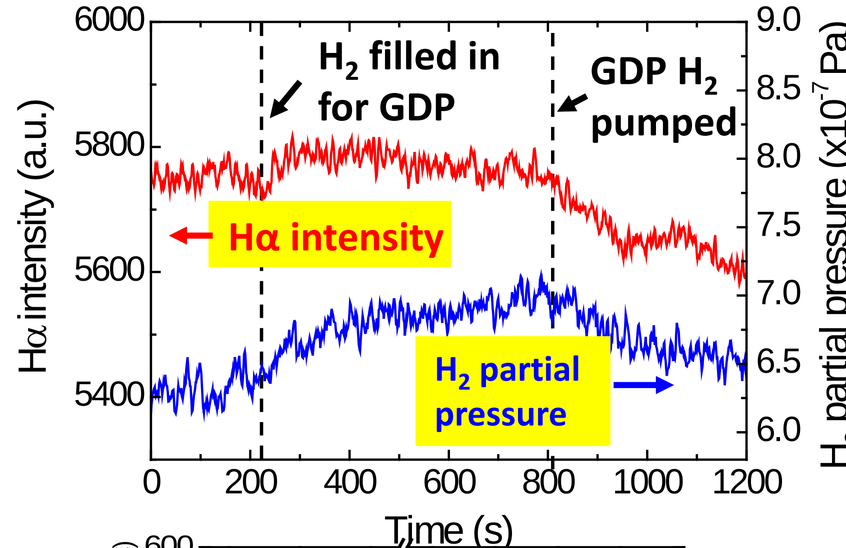
H - GDP: ~10^4 Pa (Fukada [1])

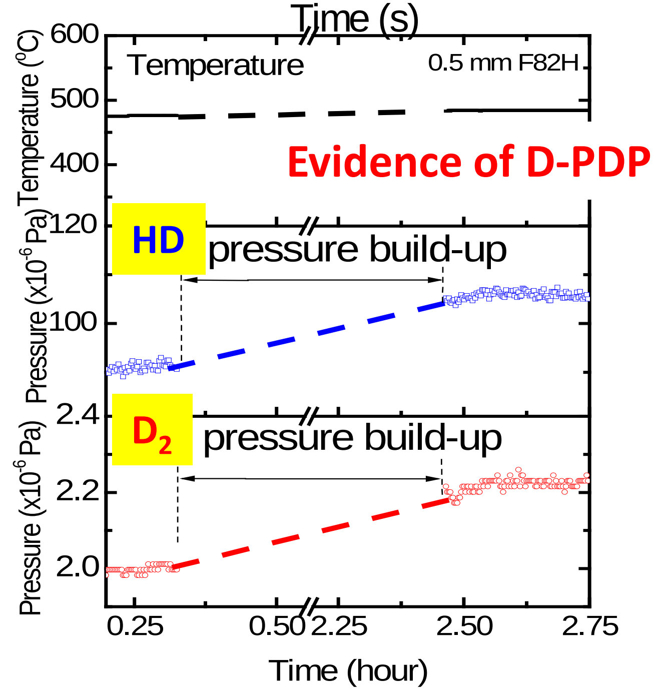
Membrane: 0.5 mm F82H

Temperature: ~470℃

[1] S. Fukada, et al., Fusion Eng. Des. 83, 747 (2008).

Evidence of H-GDP





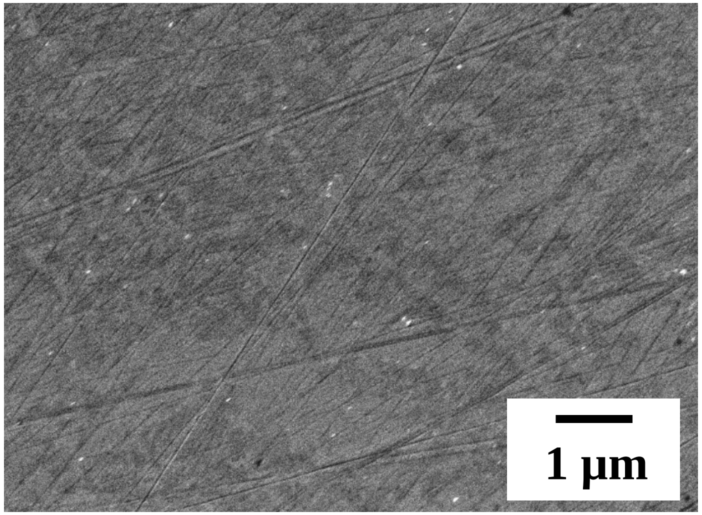
Y. Xu et al., Fusion Eng. Des. DOI: 10.1016/j.fusengdes.2017.04.022 (2017).

3. Tungsten coatings effects on hydrogen permeation

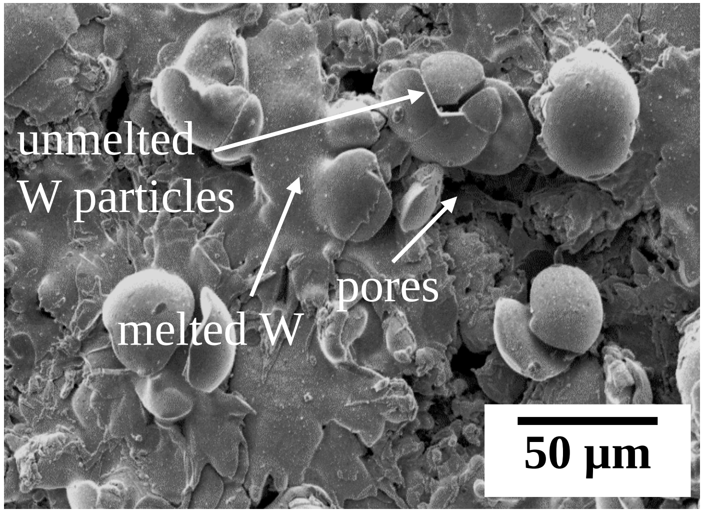
3.1. VPS-tungsten coated F82H

Microstructure of VPS-W coatings

(a) F82H surface, polished

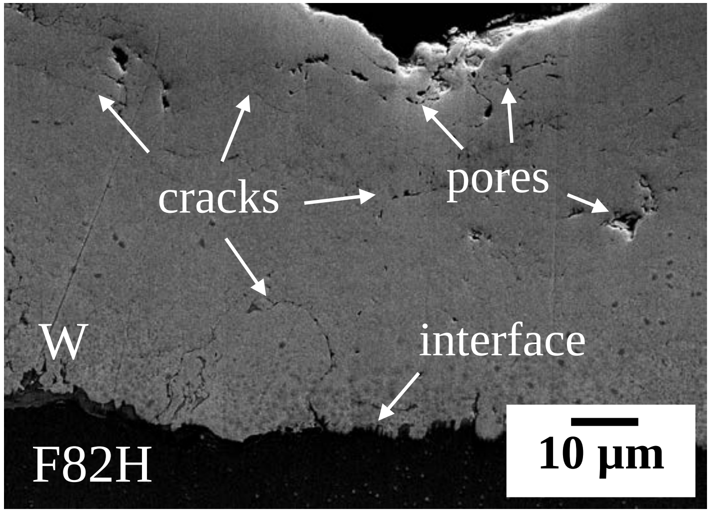


(b) VPS-W surface, as-received



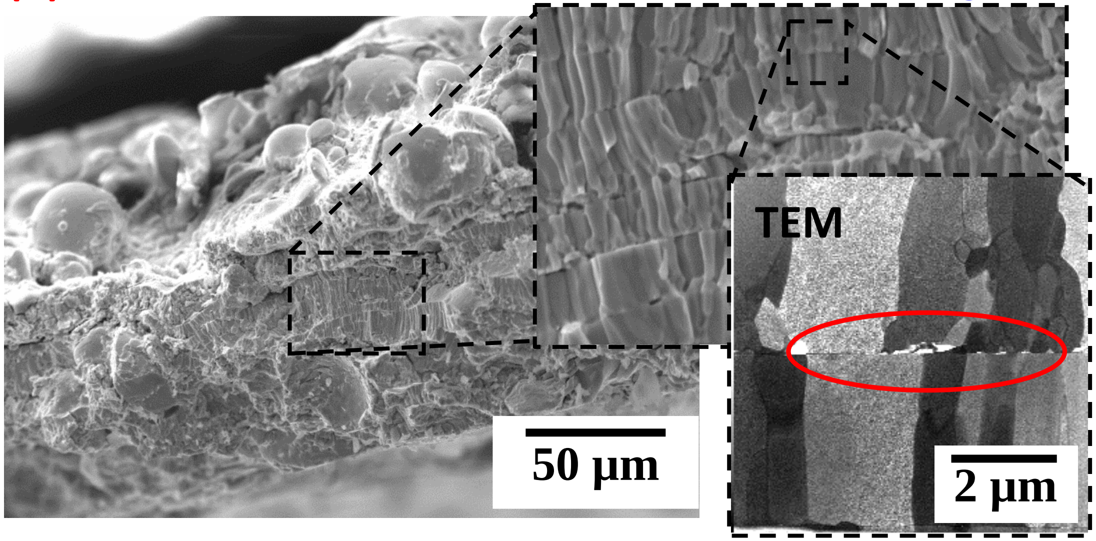
- Deposition temperature: ~600 ℃
- W powder particles: ~25 µm
- Density: ~90% of bulk W

(c) W/F82H cross-section, polished



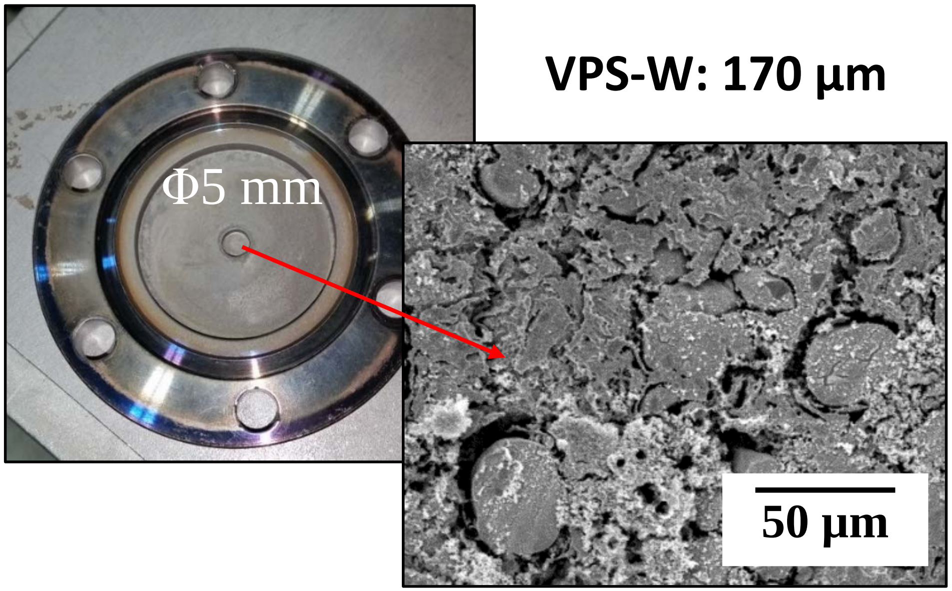
(d) VPS-W cross-section, fractured

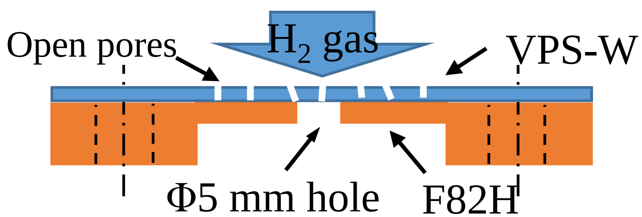
lamellar structure
columnar grains



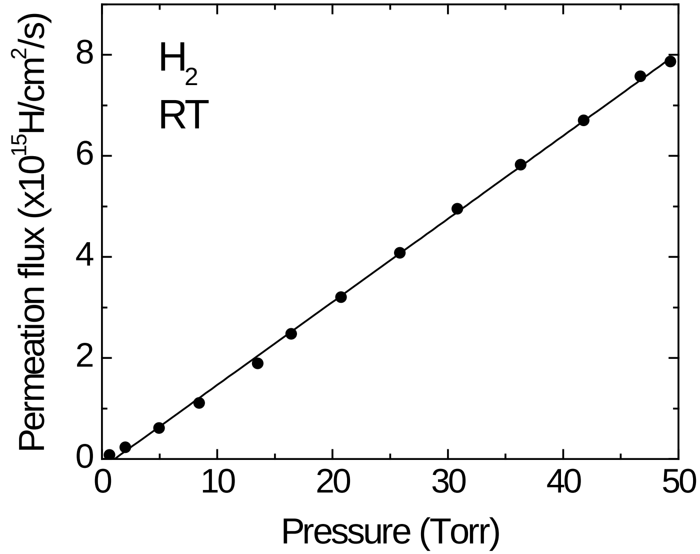
Experimental proof of connected pores of VPS-W

VPS-W: 170 μm





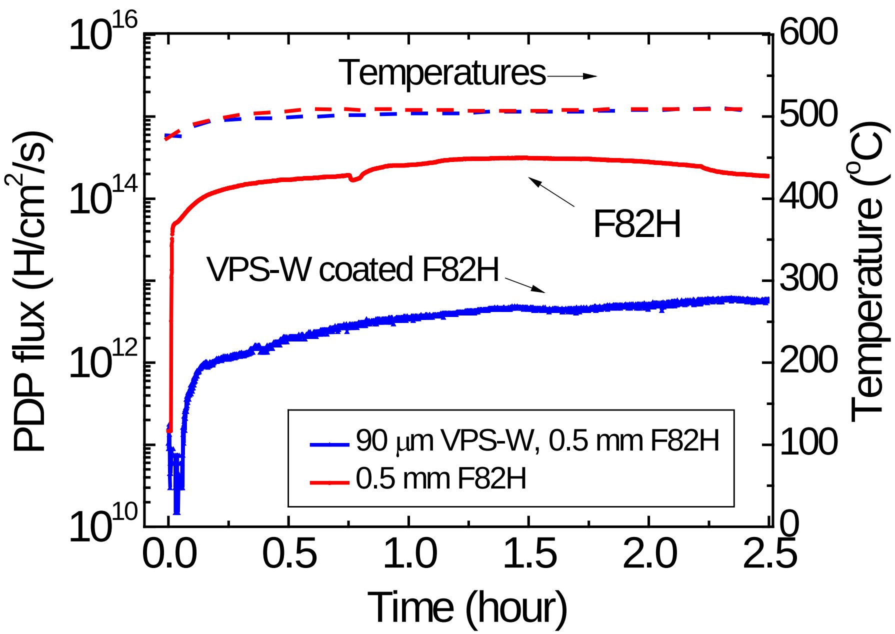
H_2 flows through VPS-W coatings at room temperature



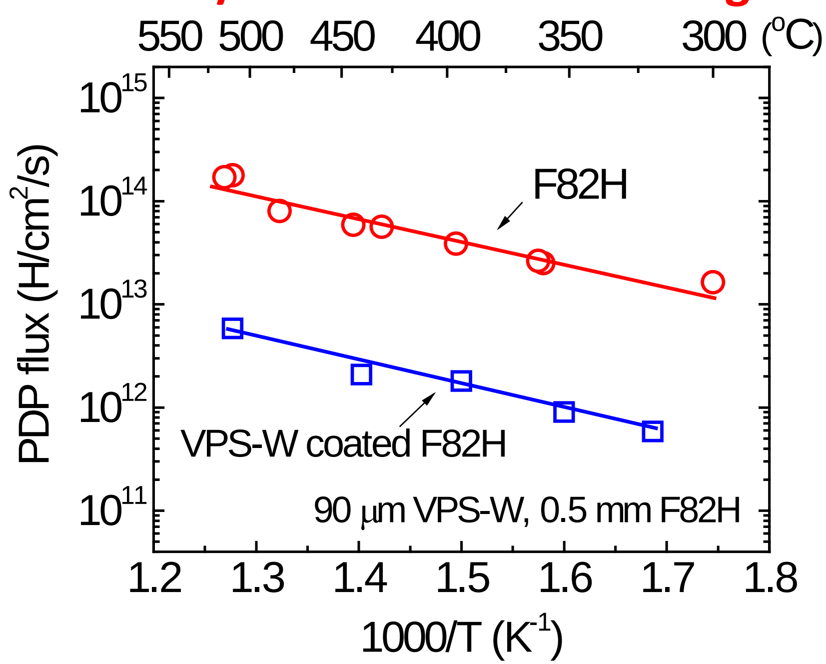
- ✓ **Machining down to the coating surface by removing F82H substrate.**
- ✓ **The permeation flux was detected immediately after gas loading.**
- ✓ **The permeation fluxes increase <u>proportional to the pressure difference</u>, indicating that gas molecules penetrate through the connected pores in the coating but not through the W bulk.**

VPS-W coatings suppress PDP

Hydrogen PDP: at ~500℃



Hydrogen PDP through F82H with/without VPS-W coatings

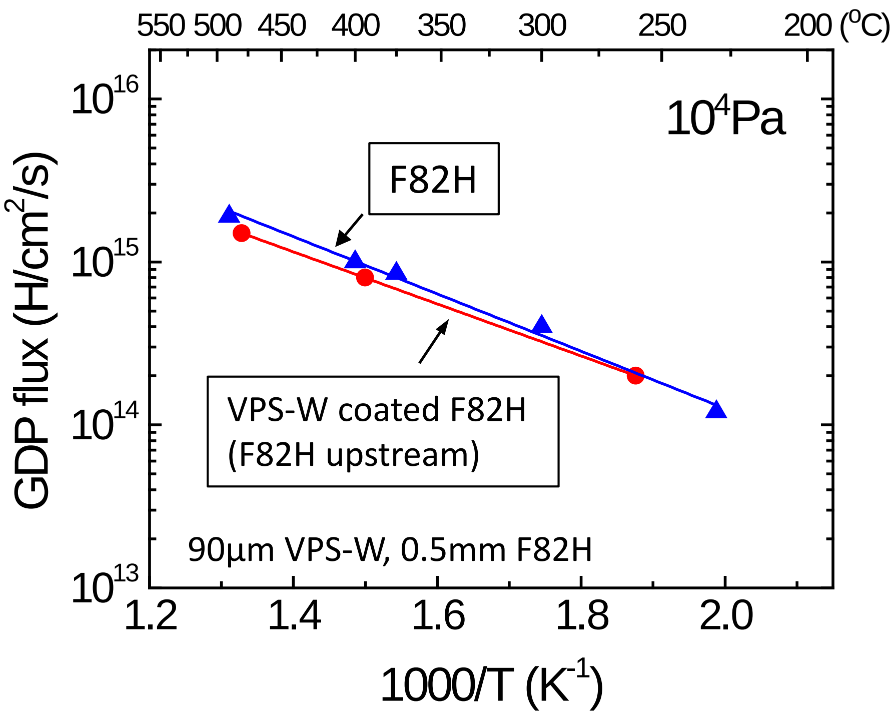


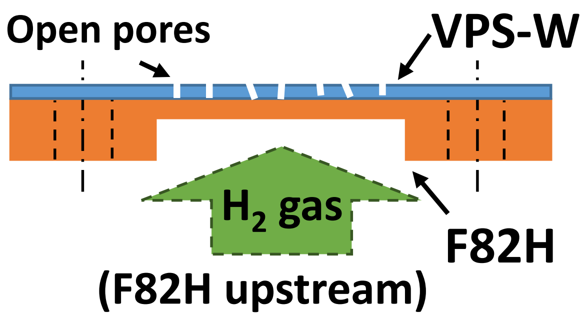
VPS-W coatings effects on PDP:

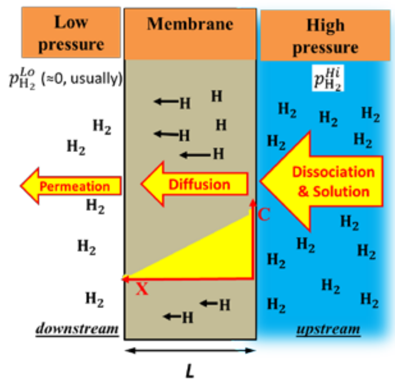
- **VPS-W suppresses DT-PDP from the plasma side.**

VPS-W coatings do not suppress GDP

Hydrogen GDP through F82H with/without VPS-W coatings







VPS-W coatings effects on GDP:

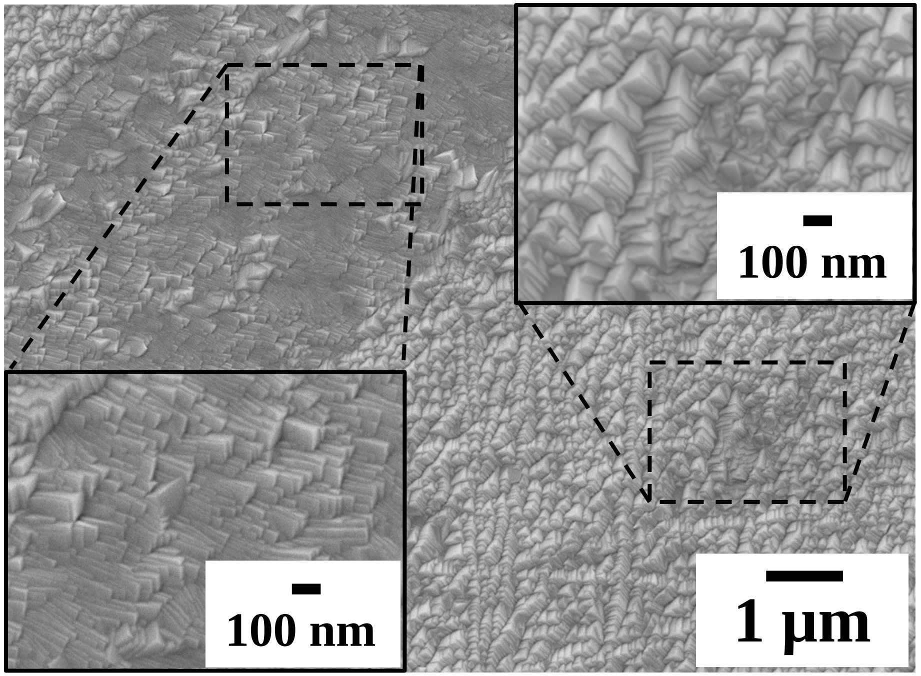
- **VPS-W does not suppress T-GDP from the blanket side.**

Y. Xu, et al., Plasma Fusion Res. 11 (2016) 2405064.

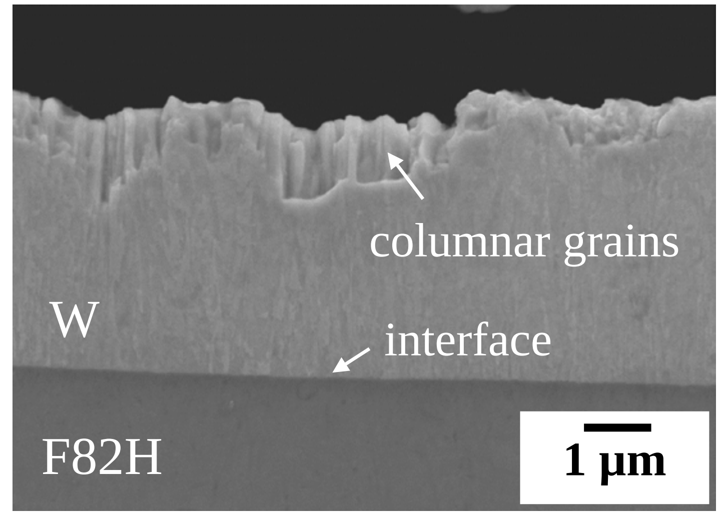
3.2. SP-tungsten coated F82H

Microstructure of SP-W coatings

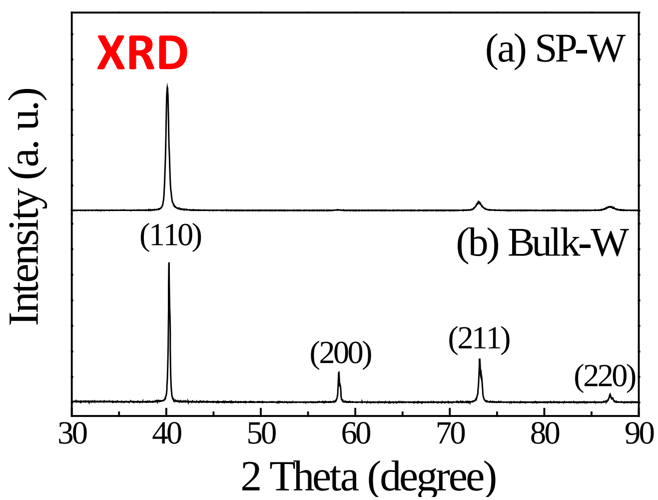
SP-W surface, as-deposited (300 ℃, 0.19 Pa)



SP-W/F82H cross-section, polished



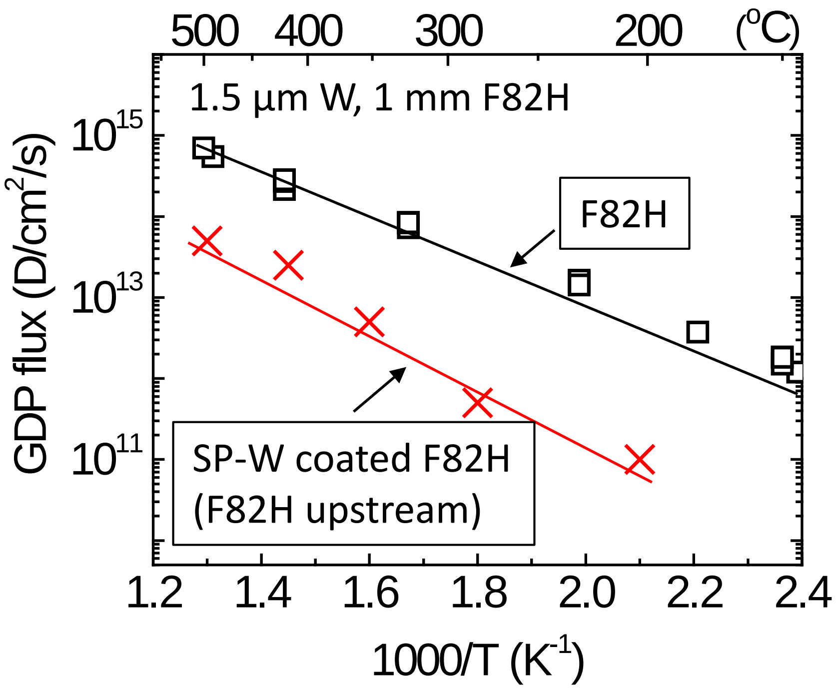
Density: 19.2 g/cm^3, ~99.5% of bulk W

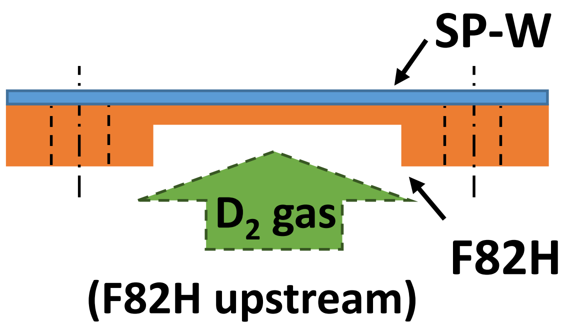


- **Fine grain.**
- **Dense structure.**
- **Preferred orientation.**

SP-W coatings suppress GDP

Deuterium GDP through F82H with/without SP-W coatings





- **Effective permeability Φ_{eff} [1]:**

$$\frac{L}{\Phi_{\mathrm{eff}}} = \frac{L_1}{\Phi_1} + \frac{L_2}{\Phi_2}$$

SP-W coatings effects on GDP:

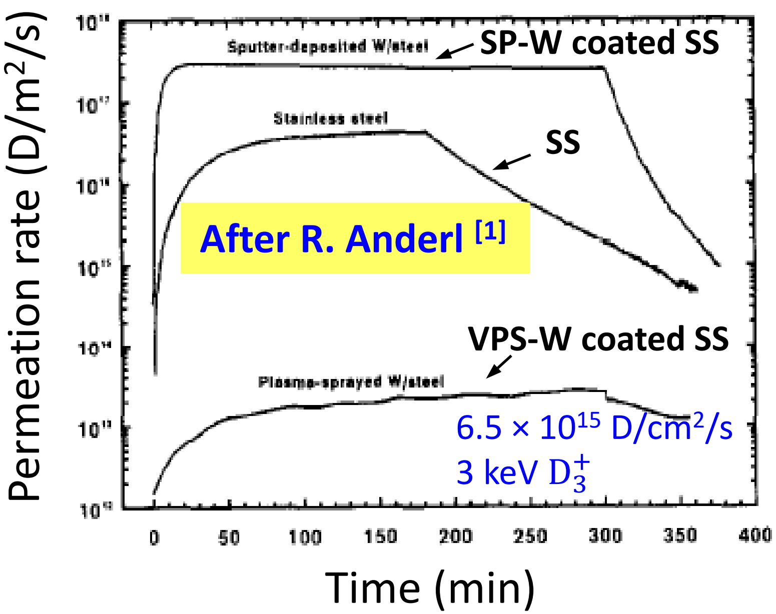
- **SP-W suppresses T-GDP from the blanket side.**

[1] J. Crank, The Mathematics of Diffusion, 2nd Edition, 1975.

SP-W coatings enhance PDP in VEHICLE-1

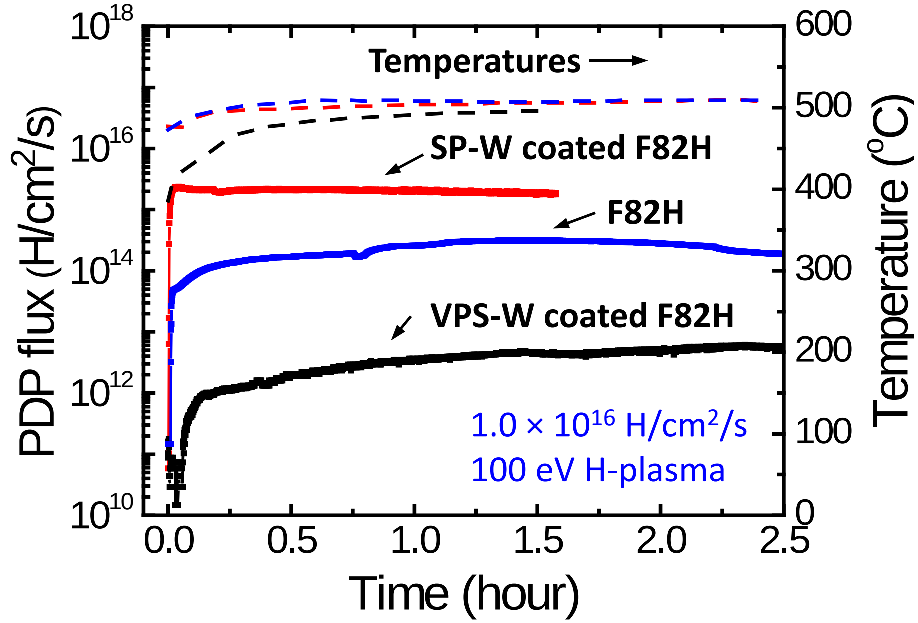
Literature data [1]:
Deuterium IDP at ~ 500 ℃

SS: 0.5 mm, SP-W: 1 μm, VPS-W: 21 μm



Present data [2]:
Hydrogen PDP: at ~500 ℃

F82H: 0.5 mm, SP-W: 0.5 μm, VPS-W: 90 μm



IDP: Ion-Driven Permeation
SS: Stainless Steel
SP-W: SPutter-deposited W
VPS-W: Vacuum Plasma-Sprayed W

SP-W coatings effects on PDP:

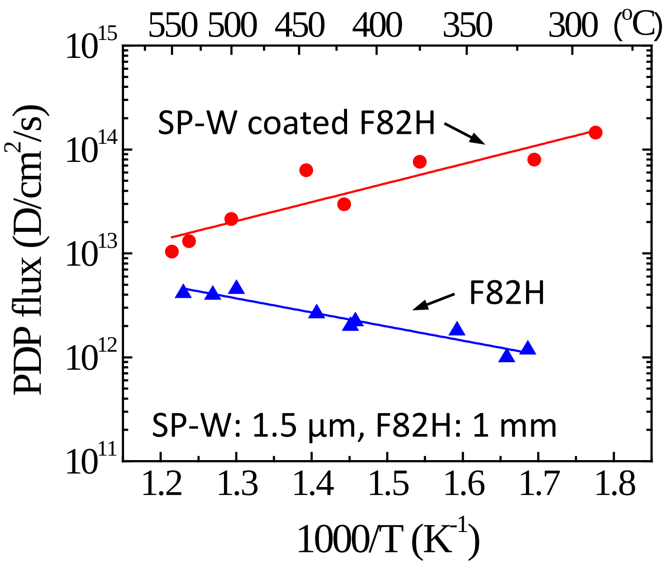
- **SP-W enhances DT-PDP from the plasma side.**

[1] R. Anderl et al., J. Nucl. Mater 212-215 (1994) 1416-1420.

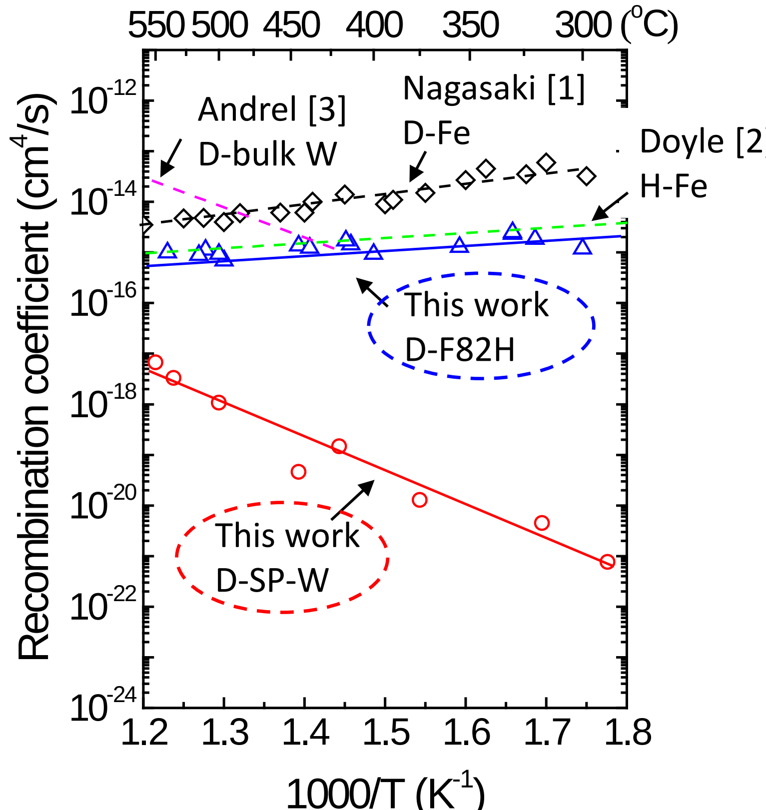
[2] Y. Xu et al., Plasma Fusion Res. 12 (2017) 1205009.

Surface recombination coefficient K_r is a key parameter relating to PDP flux

Steady-sate D-PDP fluxes



Recombination coefficient



PDP in RD-regime [2]: $J_+ = \frac{D}{L}\sqrt{\frac{J_0}{K_r}}$

Materials with small K_r tend to suppress re-emission, increasing the hydrogen concentration. As a result permeation flux increases.

Particle balance:

$$J_0 = J_- + J_+$$

implantation re-emission permeation

$$K_r(\mathrm{D-W}) = 1.2\times10^{-10}\exp\left(\frac{-1.21\ eV}{kT}\right)\ \mathrm{cm^4\,s^{-1}}$$

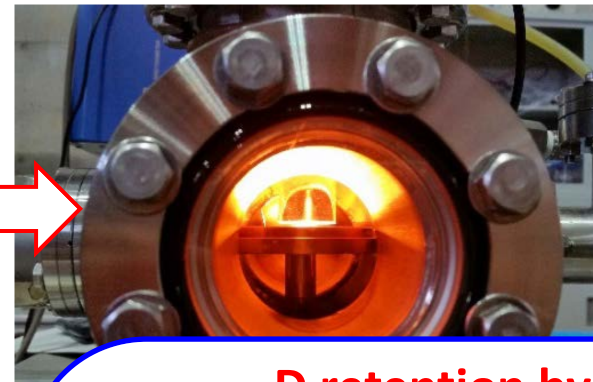
$$K_r(\mathrm{D-F82H}) = 3.8\times10^{-17}\exp\left(\frac{0.20\ eV}{kT}\right)\ \mathrm{cm^4\,s^{-1}}$$

[1] T. Nagasaki et al., J. Nucl. Mater. 202 (1993) 228–238.
[2] B. L. Doyle et al., J. Nucl. Mater. 123 (1984) 1523-1530.
[3] R. Anderl et al., Fusion Sci. Technol. 21 (1992) 745-752.

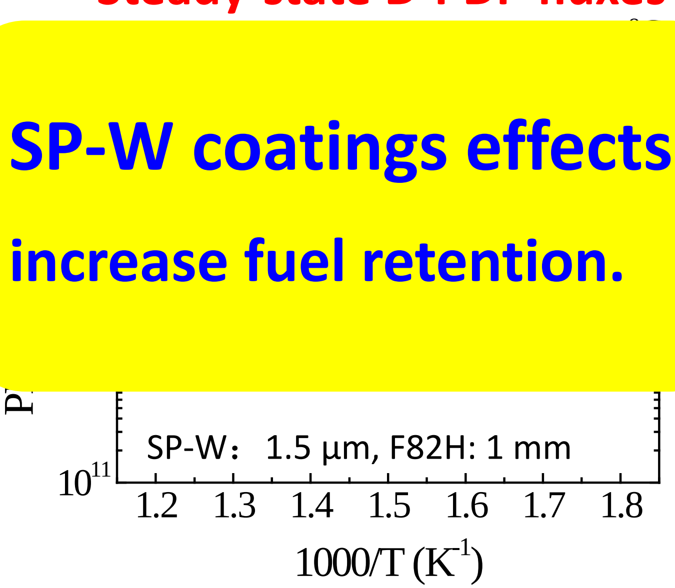
Increased D retention in SP-W coated F82H has been observed: trapping effects [1] ?

PDP (implantation)

TDS (desorption)



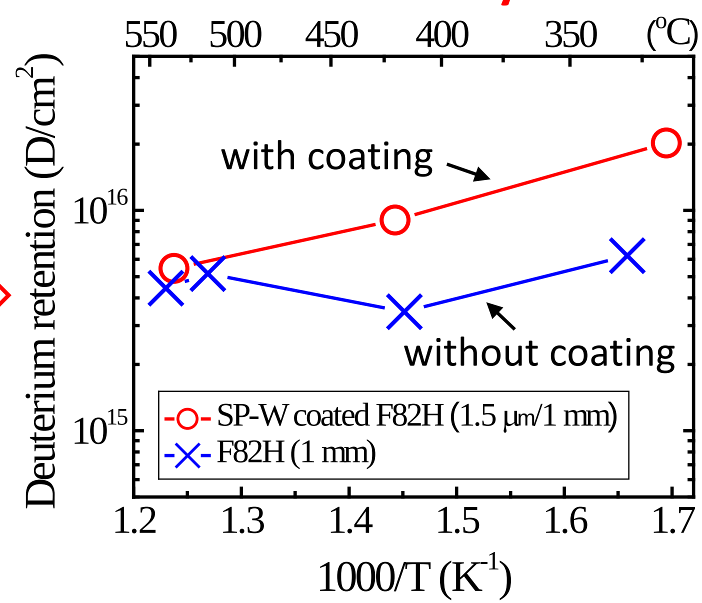
Steady-state D-PDP fluxes



SP-W coatings effects:

increase fuel retention.

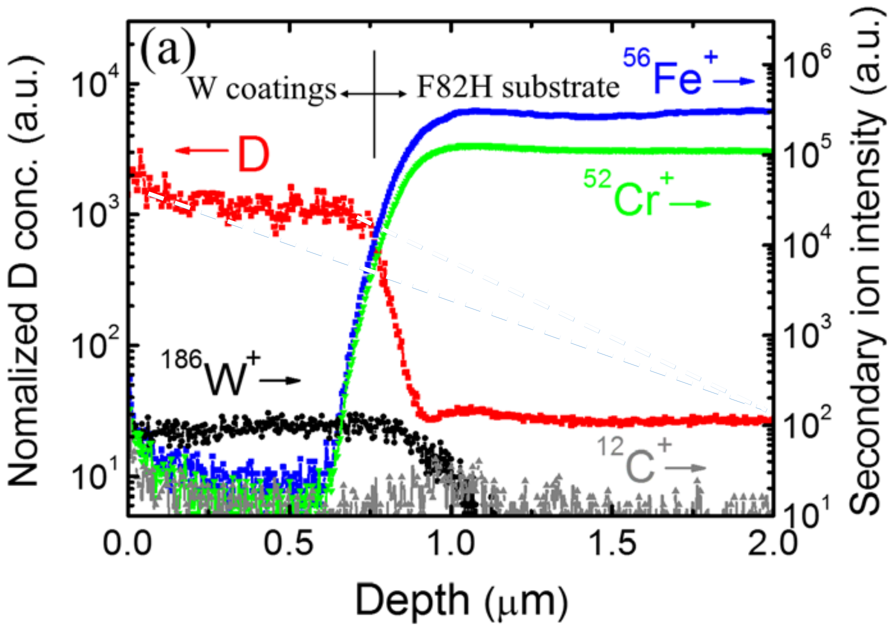
D retention by TDS



[1] B. Zajec, et al., J. Nucl. Mater. 412 (2011) 116-122.

SIMS deuterium conc. depth profile

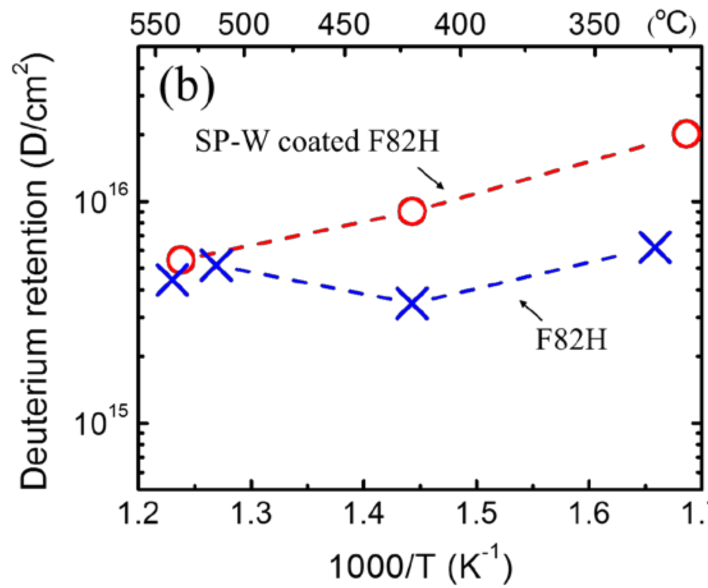
SIMS depth profile



D-PDP: 1×10^{16} D/cm^2/s, ~250 °C, 2 h
D retention: 1.2×10^{16} D/cm^2
SIMS: O_2^+, 5 keV, 250 nA

(Secondary Ion Mass Spectrometry)

D retention data by TDS

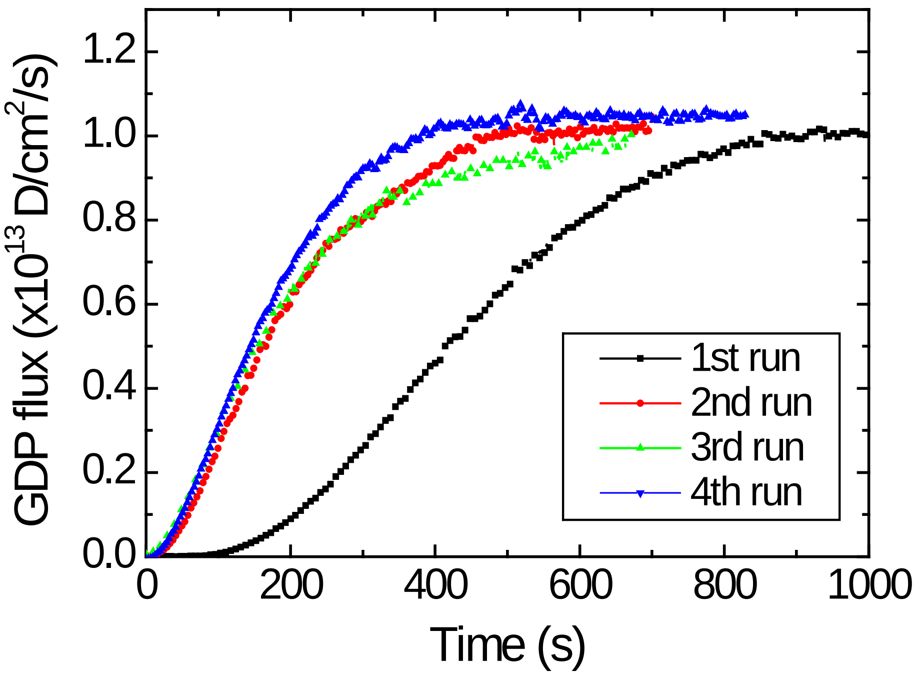


SIMS depth profile shows a good agreement with the D retention data and exhibits a sign of "uphill diffusion" of D in the W/F82H bi-layer structure.

$$\frac{C_{1,i}}{C_{2,i}} = \frac{S_1}{S_2}$$

S_{eff}

Evidence of trapping effects in SP-W



Hatano et al., data presented on PSI-22, Rome (2016).

ndary

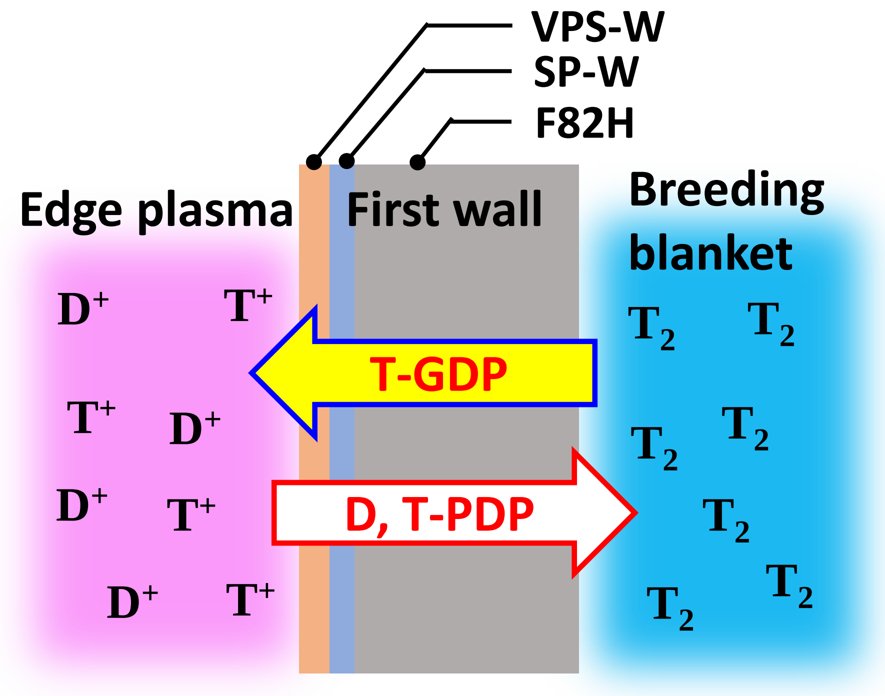
ion

Possible trapping sites:

1) **Grain boundaries;**
2) **Dislocations;**
3) **Vacancy and vacancy clusters…**

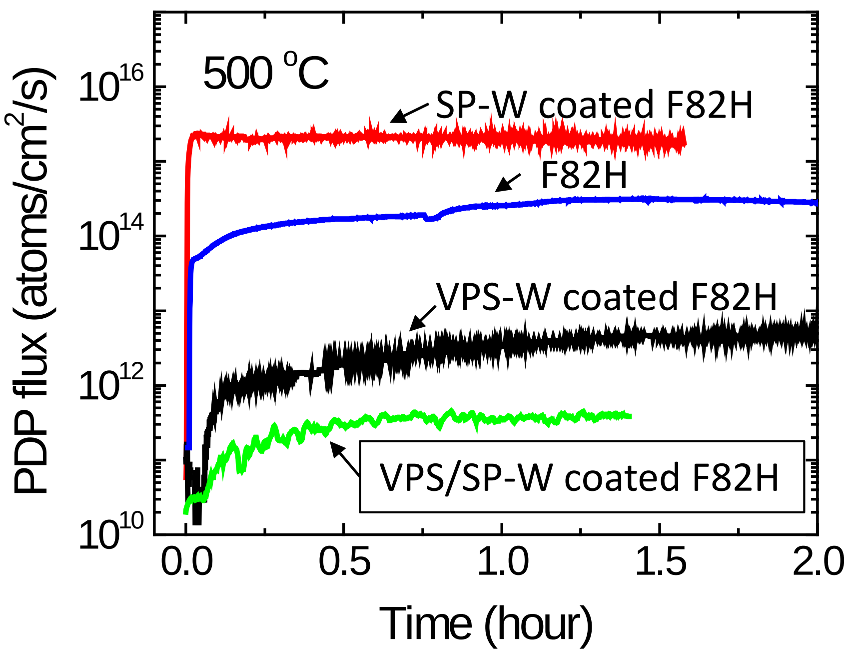
Y. Xu et al., Fusion Eng. Des. DOI: 10.1016/j.fusengdes.2017.07.021 (2017).

3.3. VPS+SP double-layer tungsten coated F82H

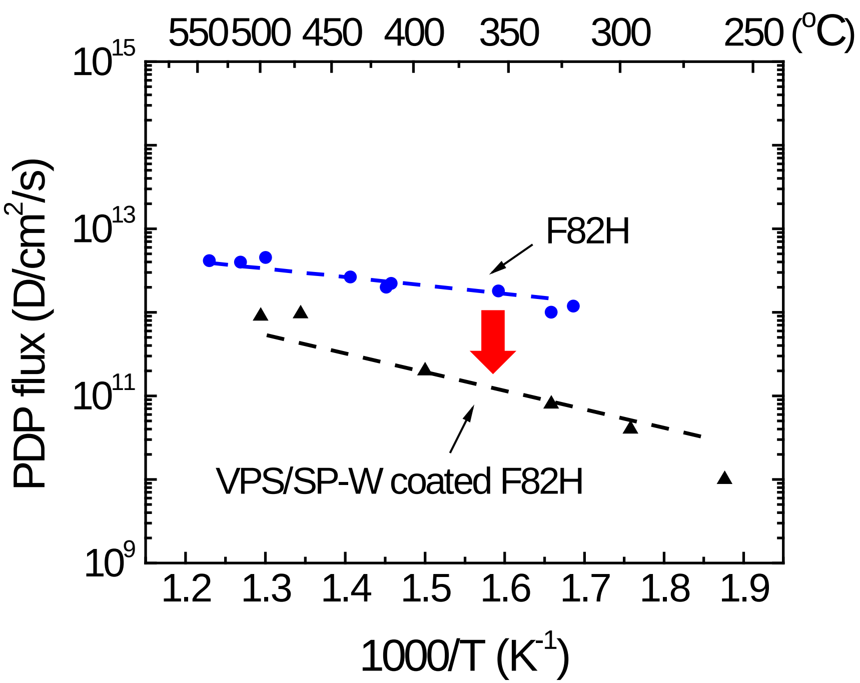


Double-layer W coatings suppress PDP

Hydrogen PDP at ~500 ℃



Steady-state PDP fluxes

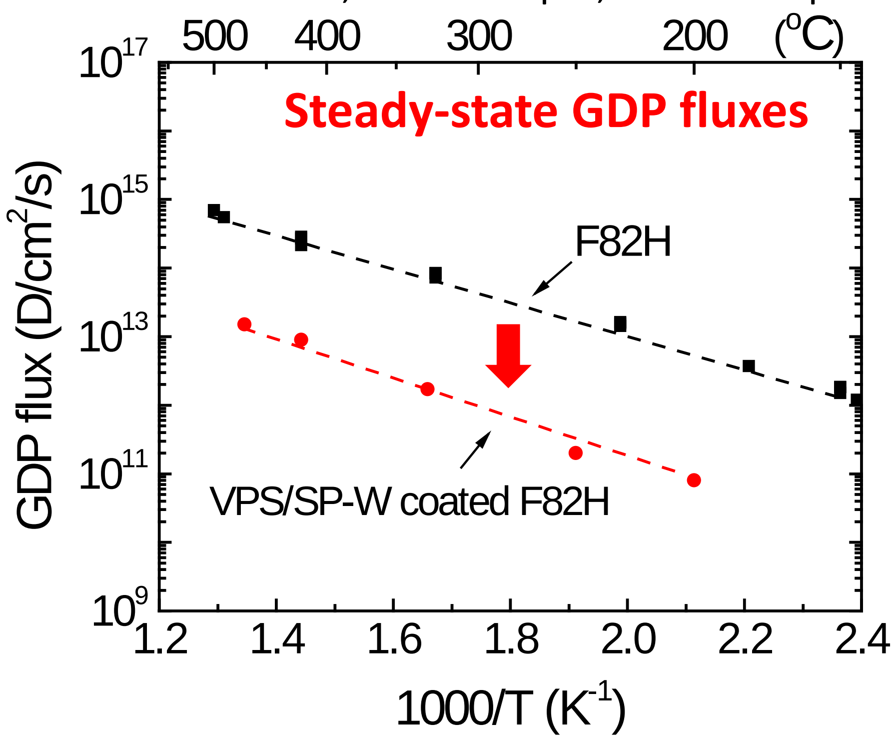


Bi-layer W coatings effects on PDP:

- **VPS/SP-W suppresses DT-PDP from the plasma side.**

Double-layer W coatings suppress GDP

F82H: 1 mm, SP-W: 1.5 µm, VPS-W: 90 µm



Bi-layer W coatings effects on GDP:

- **VPS/SP-W suppresses T-GDP from the blanket side.**

Summary

I. **VPS-W coatings have been found to have a low density structure with connected pores, while SP-W coatings have a dense and pore-free structure.**

II. **Effects of W coatings on GDP and PDP have been investigated:**

   1) **VPS-W coatings suppress PDP, but does not suppress GDP.**

   2) **SP-W coatings suppress GDP, but tend to enhance PDP.**

III. **Based on these observations, the use of overlaid VPS+SP coatings has been proposed for the first wall design.**

IV. **It has been demonstrated that VPS+SP double layer W-coatings can suppress both GDP and PDP of hydrogen isotopes through the first wall.**