Studies on bi-directional hydrogen isotopes permeation through the first wall of a magnetic fusion power reactor

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Plasma-Wall Interaction with Reduced Activation Steel Surfaces in Fusion Devices Dec. 9-11th 2015



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- **1. Background and motivation**
- 2. Experimental facility and setup
- 3. Hydrogen permeation experimental data and modelling
 - **3-1.** Implications for reactor fuel loop operations
 - 3-2. Hydrogen transport parameters in the first wall

6. Summary

Background (1)

Definition of the first wall:

- ✓ All the fusion experimental devices up to ITER: the first wall is a vacuum chamber wall to separate plasma from the environment.
- Power reactors: the first wall is the plasma-facing surfaces of breeding blanket units.



FFHR reactor

Background (2)

Ι.

mechanical stress.



A. Sagara et al., J. Nucl. Mater. 258-263 (1998) 2079-2082. 4

Background (3)

In most of the recent reactor studies, the first wall is designed to be 5 mm or even less, although these concepts employ various first wall materials.



Potential Issues with the "thin" wall design



Potential issues, but not quite well addressed, with the "thin" wall design are:

Tritium equilibrium pressures in breeders



Solubility of gases in metals (Sieverts' law):

$$C_{\text{T in wall}} = S(T) \cdot \sqrt{p_{\text{T}_2}}$$

Assuming 0.1 ppm, the total tritium inventory is of the order of 10 g in the FLiBe blanket.

For blanket employing FLiBe, the tritium thermodynamic equilibrium pressure is ~10⁴ Pa at 527 °C at a tritium concentration of ~0.1 ppm.

S. Fukada, Y. Edao, Y. Maeda and T. Norimatsu, Fusion Eng. Des. 83, 747 (2008).

DIFFUSE-code predictions on FLiBe-blankets

(PDP: 100eV, e16 H⁺/cm² \rightarrow 5mm α -Fe, GDP: 10⁴Pa H₂ \rightarrow 5mm α -Fe at 300 \rightarrow 800K)

1000

900

800

700

600

500

400

300

200

100

0

1000

 \mathbf{X}

Temperature

Permeation fluxes

PDP

GDP

600

800

GDP~1 Torr liter/s/m²

Time(s)

400

—— Temperature(K)

1.0E+16

(\$1.0E+14 1.0E+12 1.0E+10 1.0E+08 1.0E+04 1.0E+04 1.0E+02 1.0E+00

1.0E+00

1.0E-02

1.0E-04

1.0E-06

1.0E-08

1.0E-10 **1.0E-12** 1.0E-14

1.0E-16

1.0E-18

1.0E-20

0

200

GDP fluxes (

PDP



Concentration profiles

F82H: Hydrogen PDP and GDP fluxes

Hydrogen PDP and GDP fluxes measured in VEHICLE-1 for a 5 mm thick F82H membrane (under the FLiBe-blanket conditions)



- Hydrogen transport in the first wall is dominated by the flow from the blanket.
- Using the experimental data, the hydrogen recycling rate has been estimated to be R= 1.025.

Evaluation of bi-directional permeation



The tritium release flux at the plasma-facing surface is a total flux from GDP and re-emission. 10

PDP and GDP under reactor-relevant conditions



Assuming a particle reflection coefficient of 0.5, the total incident flux is 2×10^{16} D&T·cm⁻²·s⁻¹ and the first wall recycling rate has been estimated to be R= 1.018.

Motivation

Potential issues associated with bi-directional DT permeation:

- PDP–D lowers the recovery efficiency of T from the breeder.
- Gas-T permeation increases recycling on the first wall side.

What is necessary to address the bi-directional hydrogen permeation and its associated reactor operation issues?

- Understandings of hydrogen PDP and GDP individually in detail;
- Evaluation of the tritium equilibrium pressure in the fuel recovery loop system;
- Determination of missing hydrogen isotopes transport parameters (solubility, diffusivity and surface recombination coefficient).

No literature data available for the surface recombination coefficient (for F82H), which is important for recycling and retention.

- I. To understand the mechanisms driving hydrogen isotopes transport processes.
- II. To demonstrate experimentally hydrogen transport phenomena that are predicted for the first wall of a fusion power reactor.
- III. To establish a database on hydrogen transport parameters for designing fusion power reactors.

2. Experimental facility and setup

Experimental facility

VEHICLE-1





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

Plasma characteristics in VEHICLE-1



Experimental setup



Thickness: 0.5-5.0 mm Diameter: 35 mm

Materials:

- F82H (Fe-8Cr-2W)
- SUS304 (Fe-19Cr-11Ni)



- The plasma- and gas-driven permeation fluxes are measured by two H₂ partial pressure gauges, respectively.
- Ion bombardment energy is provided by a negative bias (-100 V or -50 V).
- Temperature: ~200 520 °C

3. Bi-directional hydrogen permeation experiments and modelling

Bi-directional permeation

 For the self-cooled breeder blankets, hydrogen isotopes will penetrate through the first wall by plasma-driven permeation (PDP) in one direction and gas-driven permeation (GDP) in the opposite direction.



Important parameters

GDP:

- Solubility
- Diffusivity
- External pressure

PDP:

- Surface recombination coefficient
- Diffusivity
- Implantation flux
- Reflection coef.

PDP GDP: Bi-directional permeation

Hydrogen bi-directional permeation experiment



- H plasma: Te: ~10 eV Ne: ~1.0×10¹⁰ cm⁻³ Bias: -50V
- H₂ pressure: ~7x10⁴ Pa
- Membrane: 0.6 mm thick F82H
- H GDP flows in the counter direction to H PDP flow and affects the upstream plasma.
- The GDP flux has been measured to be 9.9x10¹⁵ H/cm²/s.

Hydrogen bi-directional permeation modelling

- GDP-T₂ pressure:
 ~7x10⁴ Pa
- Ion flux:
 ~8.5×10¹⁵ H·cm⁻²·s⁻¹
- Membrane:
 0.6 mm thick α-Fe
- Temperature: Gas-facing side: ~580 °C Plasma-facing side: and ~550 °C
- Boundary conditions: Gas side: Sieverts' law Plasma side: recombination
- Intrinsic trap density: 1%
- Trapping energy: 0.62 eV



The experimental result is in relatively good agreement with the prediction by modelling.

F82H: Hydrogen PDP and GDP fluxes

Hydrogen PDP and GDP fluxes measured in VEHICLE-1 for a 5 mm thick F82H membrane (under the FLiBe-blanket conditions)



- Hydrogen transport in the first wall is dominated by the flow from the blanket.
- Using the experimental data, the hydrogen recycling rate has been estimated to be R= 1.025.

4. Modelling on the reactor fuel loop operation with hydrogen isotopes bi-directional permeation through the first wall

4-1. Re-evaluation of hydrogen bidirectional permeation fluxes through the first wall for FLiBe blankets

Re-evaluation of the tritium pressure for FLiBe blankets

Re-evaluation of the tritium flows in a FLiBe loop has been performed, taking into account tritium leakage from the first wall.



 M_{T} : tritium inventory in the loop;

*J*₁: tritium production rate;

- J₂: tritium PDP flow into blanket;
- J₃: tritium GDP flow from blanket and GDP leak from pipes;

*J*₄ : recovered tritium;

J₅: tritium GDP leak from heat exchanger

Overall tritium inventory:

$$\frac{dM_T}{dt} = J_1 + J_2 - \sum J_3 - J_4 - J_5$$

Re-evaluation of the tritium pressure for FLiBe blankets

Conditions for analysis:

*Song et al., Plasma Fusion Res. 7 (2012) 2405016.

	This work	*Song et al.
Fusion power	3 GW (Sagara)	1 GW
Tritium breeding ratio	1.3 (Sagara)	1.25
Blanket surface area	3000 m ² (Tanaka)	489 m ²
FLiBe flow rate	2.2 m ³ /s	2.2 m ³ /s
Tritium recovery rate	0.99 (Sagara)	0.98
First wall	5 mm thick F82H	10 mm thick F82H with coatings
Plasma flux	5 × 10 ¹⁵ D cm ² /s 5 × 10 ¹⁵ T cm ² /s	No PDP assumed
Calculated tritium pressure	1.1×10 ³ Pa	4.3×10 ³ Pa

4-2. Isotopes effects on hydrogen permeation

Modelling of bi-directional permeation involving multiple hydrogen isotopes

Bi-directional GDP of the same hydrogen isotope



- Temperature: 527 °C
- Boundary condition: Sieverts' law
- Intrinsic trap density: 1%
- Trapping energy: 0.62 eV

The tritium concentration profiles interact with each other in the two counter flows, finally reaching a flat profile with no net directional flow (quasi-thermodynamic equilibrium).

Modelling of bi-directional permeation involving multiple hydrogen isotopes

Bi-directional PDP-D/T and GDP-T₂ flows.



- T₂ pressure: ~1 Pa
- Implantation flux: 5×10¹⁵ D·cm⁻²·s⁻¹ 5×10¹⁵ T·cm⁻²·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.

4-3. Re-evaluation of hydrogen bidirectional permeation fluxes through the first wall for FLiBe blankets

Re-evaluation of the bi-directional permeation process

Conditions for DIFFUSE calculation:

- T₂ pressure: 1.1×10³ Pa
- Implantation flux assumptions:
 - $\Gamma_{\rm D}$ = 5×10¹⁵ D·cm⁻²·s⁻¹
 - $\Gamma_{\rm T} = 5 \times 10^{15} \, \text{T} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$
- Membrane: 5 mm thick α -Fe

D+T Plasma

Intrinsic trap density: 1%

Trapping energy: 0.62 eV

The tritium release flux at the plasma-facing surface is a total flux from GDP and re-emission. 30

D/T release fluxes at the plasma-facing surface



PDP and GDP under reactor-relevant conditions



Assuming a particle reflection coefficient of 0.5, the total incident flux is 2×10^{16} D&T·cm⁻²·s⁻¹ and the first wall recycling rate has been estimated to be R= 1.006.

5. Experimental work on hydrogen isotopes transport parameter

5-1. Gas-driven permeation (GDP) experiments for the evaluation of solubility and diffusivity

Gas-driven permeation



Permeability for GDP: $P(T) \equiv D(T)S(T)$

Diffusion coefficient from GDP

3 methods to evaluate the diffusion coefficient, D:



[2] MAV Devanathan *et al.*, Proc. R. Soc. Lond. A270 (1962) 90-102.

SUS304: Validation of the experimental setup and data analysis



F82H: pressure and thickness dependence of GDP flux



• A linear relation between GDP flux and the square-root of upstream pressure has been found at all the temperatures examined in this work.

GDP is diffusion-limited model:
$$J_{\text{GDP}} = -D(T)\frac{dC}{dx} = \frac{D(T)S(T)}{L}\sqrt{p_{\text{H}_2}}$$

 Even for a 5 mm thick membrane, reactor-relevant, hydrogen GDP has been found to be diffusion limited.

F82H: hydrogen diffusion coefficient evaluation

The diffusion coefficient of H through F82H has been evaluated from the transient permeation behavior.



Measured diffusion coefficient:

>~250 °C
$$D = 7.5 \times 10^{-4} \exp\left(\frac{-0.14 \text{ [eV]}}{kT}\right) \text{ [cm}^2 \cdot \text{s}^{-1}\text{]}$$

< ~250 °C $D = 1.9 \exp\left(\frac{-0.50 \text{ [eV]}}{kT}\right) \text{ [cm}^2 \cdot \text{s}^{-1}\text{]}$

V. Shestakov et al., J. Nucl. Mater. 307-311 (2002) 1494.

A breaking point has been found for the diffusion coefficient data, which we attribute to the trapping effect.

In the presence of traps:



T.V. Kulsartov et al., Fusion Eng. Des. 81 (2006) 701.

E. Serra et al., J. Nucl. Mater. 245 (1997) 108.

F82H: hydrogen permeability and solubility

The permeability and solubility of H in F82H have been evaluated from the steady state temperature dependent GDP data.



5-2. Plasma-driven permeation (PDP) experiments for the evaluation of surface recombination coefficients

Steady state plasma-driven permeation models



Estimation of the net implantation flux for PDP experiments



- H_3^+ is the dominant ion species in the electron temperature range of the experiments.
- The concentration of H⁺ increases as the increase of electron temperature and becomes the dominant species when the electron temperature is higher than 4 eV.

F82H: membrane thickness effects on steady state PDP

Steady state PDP data for F82H membranes at ~220 °C and ~500 °C (-100 V bias)



• The steady state permeation flux is inversely proportional to the membrane thickness, meaning that the hydrogen transport process is in the recombination-diffusion limited regime.

F82H: surface recombination coefficient measurements

The surface recombination coefficient of H on F82H has been measured from the steady state temperature dependent PDP data.



The surface recombination coefficient:

$$K_r = 4.8 \times 10^{-21} \exp(\frac{0.48 \text{ [eV]}}{kT})$$

[cm⁴·s⁻¹]

• The recombination coefficient for F82H has been estimated for the first time from the experimental data.

5-3. Surface condition effects on PDP

Earlier studies related to surface condition effects

Surface condition effects interpretations in earlier studies:

- (1) changes in surface recombination coefficient due to sputtering or deposition of contaminations (Causey et al.);
- (2) changes in surface recombination coefficient and diffusivity due to ion-induced surface defects (Winter et al.).

This work will:

- (1) propose a new hypothesis to interpret the surface contamination effects and;
- (2) propose a new model to describe the surface area/roughness effects.

Surface contamination effects on PDP (1)

(1) Surface contamination effects



Surface contamination effects on PDP (2)

The contamination effects have been investigated by the surface oxidization method.



L

d

Surface contamination effects on PDP (2)

Surface composition of the samples are examined by X-ray photoelectron spectroscopy (XPS).

0

0

20

Polished surface



After 45 min oxidization and PDP



Hydrogen implantation profile

Depth (nm)

40

60

80

After 45 min oxidization

С

0

Fe

Cr

W

100



The thickness of the impurity layer is larger than the implantation range.



Surface area effect on PDP behavior



Surface area effects on PDP (1)

To verify surface area effect, PDP experiments have been performed using samples with well controlled surface morphology.



Surface area ratio: $A_1/A_0=6.4$ Effective thickness:

4 mm< $L_{\rm eff}$ < 5 mm

Three samples:

- Flat surface
- Modified surface-1
- Modified surface-2

Modified surface-2



Surface area ratio: $A_1/A_0=3.2$ Effective thickness:

4 mm< **L**_{eff} < 5 mm

Surface area effects on PDP (2)

The measured steady state permeation flux has been found to be inversely proportional to the square root of surface area, which is in good agreement with the model prediction.



Surface area effects on PDP (3)



- Compared with the polished surface, the steady state PDP flux for the plasma modified surface has been found to decrease by a factor of ~1.7, indicating a surface area ratio of $A_1/A_0 = ~2.8$.
- The surface area is increased due to the surface modification.

Combined two surface condition effects



• A combined effect has been observed as follows:

~3.2

Combined surface effect ~ oxidization effect × surface area effect

Summary and future plans

- I. Bi-directional hydrogen permeation has been demonstrated for the first time in a laboratory-scale steady state plasma facility.
- II. Revaluation of the dynamic tritium pressure in a fuel recovery loop has been performed.
- III. Hydrogen PDP and GDP through F82H have been investigated under some of the reactor-relevant conditions.
- IV. Hydrogen transport parameters have been evaluated for F82H, including the recombination coefficient.
- V. Surface condition effects on hydrogen permeation have been examined and a new model has been proposed to interpret the data.
- VI. Future plans include: deuterium PDP and GDP experiments to evaluate the isotope effects and also surface coatings effects (i.e. W-coatings).