Effects of surface conditions on the plasma-driven permeation behavior through a ferritic steel alloy observed in VEHICLE-1 and QUEST

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ABSTRACT

Effects of surface conditions on the plasma-driven permeation of hydrogen through a ferritic steel alloy F82H have been studied in a laboratory-scale plasma device: VEHICLE-1 and the medium-sized spherical tokamak: QUEST. Both of the surface contamination and area effects have been examined and discussed. Thick surface impurity film has been found to act as a second layer for diffusion and affect the permeation behavior in laboratory-scale and tokamak experiments. Hydrogen diffusion coefficient in the impurity layer has been estimated using the multi-layer diffusion model. A decrease in steady state permeation flux has been measured when increasing the plasma-facing surface area, which is in agreement with theoretical prediction.

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1. Introduction

In magnetic fusion power reactors, hydrogenic particles will escape from the confinement region and then migrate through the first wall by plasma-driven permeation (PDP) [1–10]. Hydrogen isotopes (deuterium and tritium) flowing into the blanket by PDP will hinder the recovery of tritium and will probably necessitate isotope separation [2]. Tritium permeation through the first wall may raise reactor safety issues as well.

Under reactor operational conditions, the plasma-facing side of the first wall would not be as smooth and clean as polished samples used in laboratories. The wall will be either covered by contaminations, or eroded by plasma bombardment, depending upon the local plasma conditions [11]. Although it is known that plasma-driven permeation is affected by the surface conditions of membranes [3–10], more detailed study on this issue is still needed, especially concerning the surface area effects.

In our previous studies, some surface condition effects on PDP through a ferritic steel alloy F82H have been observed [8]. The present work is a further investigation of these effects, in which two aspects are separately examined: one is the influence of surface contamination; and the other is the surface area effects. The experimental results presented in this paper cover the PDP data from the laboratory-scale steady state plasma device: VEHICLE-1 [12] as well as from the medium-sized spherical tokamak QUEST [13].

2. Models on surface modification effects on PDP

Some possible mechanisms related to the surface condition effects on plasma-driven permeation are briefly reviewed in this section, including both contamination and area effects. The validity of these theoretical models is shown in Section 4.

2.1. Surface contamination effects on PDP

In the steady state plasma-driven permeation model, three regimes are considered [1]: the diffusion–diffusion (DD) limited regime, the recombination–diffusion (RD) limited regime and the recombination–recombination ( RR) limited regime. Our previous experiments indicated that hydrogen PDP through F82H under current experimental conditions takes place in the RD limited regime, i.e., recombination-limited at the front surface and diffusion-limited inside the bulk, as shown in Fig. 1(a). Steady state permeation can be expressed by the following equations:

\[ J_0 = J_+ + J_- \]  \hspace{1cm} (1)

\[ J_+ = K_r \cdot C_1^2 \]  \hspace{1cm} (2)

\[ J_- = D \cdot C_1/L = D/L \cdot \sqrt{J_0/K_r} \]  \hspace{1cm} (3)
where \( J_0 \) is the net implantation flux, \( J_- \) is the recombination release (i.e., reemission) flux from the upstream surface, \( J_+ \) is the permeation flux, \( C_i \) is the front surface hydrogen concentration and \( K_i \) is the recombination coefficient of the plasma-facing surface.

Some literature data suggest that the hydrogen PDP flux \( J_+ \) is enhanced when the plasma-facing surface is contaminated [4–7]. That is because recombination release \( J_- \) is suppressed by the presence of impurity film, as shown in Fig. 1(b). However, it is also true that if the contaminated layer is thick enough to act as a second layer for diffusion, hydrogen PDP will be suppressed (Fig. 1(c)) [8,9]. The latter case is of interest from the fusion engineering point of view as it suggests a potential measure to reduce hydrogen PDP through the first wall in a reactor.

### 2.2. Surface area effects on PDP

The surface of plasma-facing walls can be modified significantly by plasma bombardment. Surface morphologies such as bubbles [14], coral-like structures and cones [11] may be formed, depending on the plasma conditions, material property, temperature and impurity seeding, etc. Those structures will change the plasma exposure area and result in higher hydrogen release at the front surface. Considering particle conservation and surface reflection, the net implantation fluxes can be expressed as

\[
J_1 = J_p \cdot (1 - R_1)
\]

for a polished surface and

\[
J_2 = J_p \cdot A_1/A_2 \cdot (1 - R_2)
\]

for a modified surface, where \( J_p \) is the ion incident flux, \( A_1 \) and \( A_2 \) are the surface areas for a polished surface and a modified surface, respectively, and \( R_1 \) and \( R_2 \) are the corresponding particle reflection coefficients. Using Eq. (3), the steady state permeation flux ratio \( J_2/J_1 \), of the two cases can be given as:

\[
J_2/J_1 \propto \sqrt{A_1/A_2}
\]

i.e., the permeation flux is inversely proportional to the square root of surface area.

### 3. Experimental

Details of the PDP experimental setup in VEHICLE-1 and QUEST have already been presented elsewhere [2,3,13]. In this section, some of the important features of the PDP experiments will be described.

For the PDP experiments in VEHICLE-1, the hydrogen permeation flux is measured by a quadrupole mass spectrometer (QMS) at the downstream side. The plasma density is of the order of \( 10^{19} - 10^{20} \text{ cm}^{-3} \) and the electron temperature is \( \sim 3.5 \text{ eV} \). The particle bombarding energy is controlled by a negative bias voltage applied on the membrane flange. A bias of \(-50 \text{ V} \) or \(-100 \text{ V} \) is used in the present experiments. A resistive heater is set beneath the membrane to control the temperature, which is measured by a thermocouple mechanically attached to the sample.

By taking into account particle reflection [15] and the ion species mix [3,12], the net implantation fluxes in the PDP experiments can be estimated from the electron and density data measured by a Langmuir probe in VEHICLE-1. The particle reflection coefficients are calculated from the Monte Carlo programs, such as SRIM [15]. Although the Binary Collision Approximation (BCA) models such as SRIM may be not necessarily accurate at low implantation energy of several eV, the particle reflection coefficients given by Monte Carlo programs are relatively consistent (\( \sim 0.5 \)) in the energy range of 50–100 eV [2]. The ion species mix of the hydrogen plasmas is estimated using a zero-dimensional model. This model includes not only the rate balance equations for \( \text{H}^+, \text{H}_2^+, \text{H}_3^+ \) and \( \text{H} \) atoms, as Hoffmann did in his work [16], but also the rate balance equation for \( \text{H}^+ \), whose concentration should not be ignored for plasmas with an electron temperature lower than 3 eV [17]. Hydrogen molecules are regarded as the particle source in this model, which is more relevant to the electron cyclotron resonance (ECR) discharge conditions in VEHICLE-1. The modelling results indicate that \( \text{H}^+_2 \) is the dominant ion species at the electron temperature and neutral hydrogen gas pressure (\( \sim 10^{-3} \text{ Torr} \)) in our experiments. The concentration of \( \text{H}^+ \) increases as the increase of electron temperature and becomes the dominant species when the electron temperature is higher than \( \sim 4 \text{ eV} \).

Surface oxidization effects on PDP have been studied in VEHICLE-1 because oxide would be one of the possible contaminations for the first wall of fusion reactors. The plasma-facing side of the F82H membrane is oxidized at \( \sim 450 \text{ °C} \) with an oxygen gas pressure of \( 10^{-4} \text{ Torr} \). For the area effects experiments, the surface morphology of the samples is modified by plasma bombardment or machining.

![Fig. 1](image-url) Plasma-driven permeation takes place in the RD-limited regime for (a) a clean surface, (b) a contaminated surface with thin film and (c) a contaminated surface with thick impurity layer.
QUEST [18] is a medium-sized spherical tokamak with a full metal chamber made of stainless steel. A permeation probe made from F82H has been installed near the mid-plane in QUEST and the permeation membrane is 35 mm away from the outboard wall in the radial direction. During plasma discharges, the membrane temperature is kept in a range of 200–300 °C, which is also measured by a thermocouple.

Samples made of F82H are prepared in the same dimensions as those commercially available conflat flanges, except that the circular area inside the knife-edge is machined down to thicknesses between 0.14 mm and 5 mm. The “standard” (i.e., not intentionally modified) F82H membranes are mechanically polished and then cleaned in an ultrasonic bath. The membrane diameters are 35 mm for the VEHICLE-1 samples and 16 mm for the QUEST ones, respectively.

Surface analysis such as scanning electron microscopy (SEM), optical microscopy and X-ray photoelectron spectroscopy (XPS) are utilized to evaluate the surface conditions before and after experiments.

4. Results and discussion

4.1. Surface contamination effects observed in VEHICLE-1 and QUEST

The hydrogen PDP behavior with/without surface oxidation is shown in Fig. 2. After oxidation, it takes a longer time for the permeation flux to reach steady state, suggesting a lower effective diffusion coefficient for the oxidized samples.

Fig. 3 shows the XPS analysis results for (a) a polished surface before PDP, (b) a surface after 0.75 h oxidation and (c) a surface after 0.75 h oxidation and 3 h plasma exposure. Notice that the implantation range of hydrogen particles in iron is less than several nm at an implantation energy of <50 eV, as calculated by SRIM [15], while the thickness of the oxide layer is tens of nm, which should be thick enough to act as a second layer for diffusion, as illustrated in Fig. 1(c). Plasma exposure can reduce the impurity layer thickness by chemical sputtering, which has been confirmed by measuring the water partial pressure in the plasma side during the PDP experiments (Fig. 3(d)). However, the XPS results indicate that the surface oxides would never be depleted completely under current experimental conditions (as shown in Fig. 3(c)), and as a result, the steady state PDP flux through these oxidized membranes is lower than a polished one.

Surface contamination effects on permeation through F82H have been observed in tokamak PDP experiments as well. It has been found that the diffusion coefficients estimated from the QUEST data are lower by a factor of 3–4 than those taken in VEHICLE-1, although the sample membranes are essentially the same [13,19]. A ~12 nm thick impurity layer containing carbon, tungsten and oxygen has been detected on the membrane surface by XPS. A membrane composed of two sheets of thicknesses \( L_1 \), \( L_2 \) and diffusion coefficients \( D_1 \), \( D_2 \) has an effective diffusion coefficient \( D_{\text{eff}} \), given by [20]:

![Fig. 2. Hydrogen PDP through clean and oxidized F82H membranes at a temperature around 520 °C.](image)

![Fig. 3. XPS results for (a) a polished surface, (b) a surface oxidized at ~450 °C by oxygen gas at 1 × 10⁻⁴ Torr for 0.75 h and (c) a surface after 0.75 h oxidation and 3 h plasma exposure. (d) Water partial pressure measurements at the plasma side in the PDP experiments for oxidized and clean samples.](image)
where $L$ is the total thickness of the membrane. Using the diffusion coefficient data for F82H from previous experiments [19]:

$$D = 7.5 \times 10^{-4} \exp \left( -\frac{0.14}{kT} \right) \text{cm}^2 \text{s}^{-1},$$

the hydrogen diffusion coefficient in the impurity layer has been estimated to be $\sim 2 \times 10^{-10} \text{cm}^2 \text{s}^{-1}$, which is close to the hydrogen diffusion coefficient for tungsten measured in the same temperature range [21].

### 4.2. Surface area effects on PDP

To verify the area effects model proposed in Section 2.2, PDP experiments have been performed using samples with well controlled surface morphology. The sample membranes are heated up to a steady state temperature of $\sim 500 \, ^\circ\text{C}$ by resistive heater radiation and plasma bombardment.

Fig. 4(a) shows a schematic diagram of the surface-modified permeation membrane. The first 1 mm or 0.4 mm of the 5 mm thick F82H membrane is machined into V-shape grooves to increase the surface area. From the dimension measurements by a microscope (as shown in Fig. 4(b)), the areas of the modified surfaces are measured to be larger by a factor of 6.4 and 3.2 than a flat one, respectively. The effective thickness of the modified sample membrane is between 4 and 5 mm. Eq. (4) suggests that when plasma-driven permeation is in the RD limited regime, the steady state permeation flux is inversely proportional to the square root of surface area.

Shown in Fig. 4(c) are the PDP flux data as a function of the square root of surface area ratio. The measured steady state permeation flux has been found to be inversely proportional to the square root of surface area, which is in excellent agreement with the model prediction.

### 4.3. Compound surface condition effects on PDP

Notice that the two surface effects are independent on each other, which means these two effects can be multiplied. The surface area modified sample (1 mm deep groove) shown in Fig. 4(a) has been oxidized at $\sim 500 \, ^\circ\text{C}$ for 4.5 h and then exposed to hydrogen plasma again.

Fig. 5 shows that a further reduction of hydrogen permeation has been observed. The steady state permeation flux has been found to be lower than that of the sample before oxidation by a factor of 1.4, which is in good agreement with the separate surface contamination effect data shown in Fig. 2. Compared with the clean and polished surface, the overall permeation flux has been found to be reduced by a factor of 3.2.

### 5. Conclusion

Hydrogen plasma-driven permeation behavior has been found to be affected both by surface contamination and area. The steady state hydrogen PDP flux can be reduced by a factor of 2 or 3 by surface treatment. However, such amount of reduction in hydrogen isotopes permeation is still not acceptable for a reactor operation and tens of g/day of tritium may still penetrate through the first wall by PDP [6,19]. The compound effects of surface contamination and area may be a possible solution, which warrants further investigation.

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### References