Hydrogen Gas-Driven Permeation through F82H Steel Coated with Vacuum Plasma-Sprayed Tungsten*)

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The subject of hydrogen isotopes transport through tungsten coated reduced activation ferritic steels such as F82H has attracted increasing interest in the fusion engineering research community. This paper reports on laboratory-scale studies that have been done to assess the hydrogen permeation properties of vacuum plasma-sprayed tungsten (VPS-W) coatings at the temperature range of 200 - 500 °C. W coatings with thicknesses of 46 μm and 90 μm have been investigated. It has been found that the observed permeation rates through composite VPS-W/F82H specimen are reduced to ~7% compared to that of pure F82 H. VPS-W coating is porous and has an open system of connected pores, which density is evaluated to be ~7%. The main effect of the W coating on hydrogen permeation is to reduce the incoming flux at the W/F82H interface owing to pore diffusion in the coating and to reduce the effective surface area for hydrogen dissolution in the substrate.

Keywords: gas-driven permeation, tungsten coating, reduced activation ferritic steel

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1. Introduction

Reduced activation ferritic steels such as F82H are the candidate first wall materials for DEMO reactors [1]. For the blankets employing self-cooled breeder, the first wall is supposed to be subjected to bi-directional hydrogen isotopes permeation by two different mechanisms: in one direction by edge plasma-driven permeation (PDP) and in the other direction by bred tritium gas-driven permeation (GDP) [2]. In the previous work, hydrogen PDP and GDP through pure F82H were studied [3, 4]. It was found that for a DEMO reactor, with a first wall area of several thousand square meters, the uncontrollable gas fueling rate is to be of the order of 1000 Torr liter/s (133.3 Pa m^3/s) [3], which is undesirable from the viewpoint of achieving high-confinement via plasma-wall boundary control. Therefore, there is a need for surface coating as hydrogen permeation barrier. W has been proposed as a candidate plasma-facing material for advanced fusion devices because of its beneficial properties, such as high melting point, low sputtering yield and low hydrogen retention [5]. Often, the easiest way to produce a W armor is to deposit a W coating on some feasible substrate. This avoids the problems of manufacturing plasma-facing components from bulk W, which is difficult to machine due to its hardness and brittleness.

Vacuum plasma spraying is an efficient technique to get W coatings. As it has been investigated by Golubeva et al. [6], that the mechanism of hydrogen permeation through VPS-W coated graphite is molecular hydrogen gas flow through the system of connected porosity. The purpose of this paper is to report on measurements and analyses that have been made to determine the hydrogen permeation properties of VPS-W. Such information is important to evaluations of hydrogen isotopes retention and permeation safety issues for fusion devices that utilize VPS-W components.

2. Experimental

Hydrogen GDP experiments are carried out using the VEHICLE-1 facility [7]. VPS-W coated F82H specimens are prepared in the same dimensions as those commercially available conflat flanges with an outer diameter of 70 mm and the permeation membrane surface area is ~35 mm in diameter. Shown in Table 1 are the specimens with different thicknesses of W coating and F82H substrate. The W coatings are deposited at ~600 °C and blast treatment has been carried out for the F82H steel surface before VPS process. The average size of W powder particles is ~25 μm. Figure 1 shows a scanning electron microscopy (SEM) image of VPS-W coating surface. It is shown that VPS-W coating has an inhomogeneous microstructure, i.e., a mix-

Table 1 Specimen list.

Specimen	Thickness of F82H (mm)	Thickness of W coating (mm)
1	0.5	0.09
2	0.5	/
3	1	0.09
4	1	0.046

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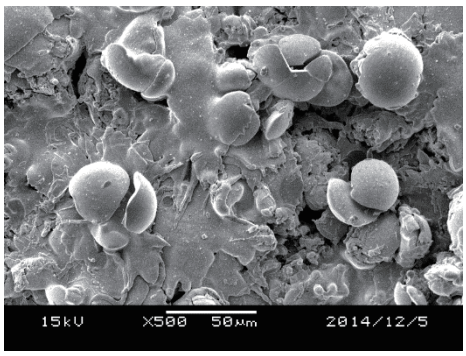

Fig. 1 SEM image of VPS-W coating surface.

ture of disorganized areas composed of large unmelted W particles, fine randomly melted W areas and void regions (or porosity) [8]. Void regions or pores are observed primarily next to the unmelted particles. The average density of VPS-W coatings is ~90% of bulk W.

The background pressures of GDP upstream and downstream are $\sim 10^{-5}$ Pa and 10^{-6} Pa, respectively. Hydrogen GDP flux can be measured by a quadrupole mass spectrometer on the downstream side. A resistive heater is positioned beneath the membrane to control the sample temperature, which is measured by a thermocouple. All the samples have been in-situ degassed at ~500 °C for 12 hours before experiments.

3. Results and Discussion

The effective diffusivity D_{eff} can be obtained by fitting the transient permeation flux [9], as shown in Fig. 2 (a), or from the so-called time lag method $t_l = L^2/6D$ [10], as shown in Fig. 2 (b).

The hydrogen D_{eff} for VPS-W coated F82H has been evaluated from 200 °C to 500 °C, as shown in Fig. 3. Literature data are shown for comparison. The evaluated hydrogen D_{eff} can be expressed as:

$$D_{\text{eff}} = 7.9 \times 10^{-4} \exp\left(\frac{-0.20\,\text{eV}}{\text{kT}}\right) \text{cm}^2\,\text{s}^{-1}. \qquad (1)$$

Shown in Fig. 4 are the steady-state hydrogen permeation fluxes through VPS-W coated F82H at the temperature range of 200 - 500 °C. A linear relationship between the GDP fluxes and the square-root of upstream pressure has been found, indicating that hydrogen GDP through the VPS-W coated F82H is diffusion-limited [13].

To investigate the effective upstream surface area effects, hydrogen permeation in both directions has been carried out. Figure 5 shows the hydrogen permeation rates through composite VPS-W/F82H specimen in both directions compared to that of pure F82 H. The upstream hydrogen pressure is set at 10^4 Pa. It shows that the GDP fluxes with the W side on the upstream are reduced to ~7% compared to that of pure F82 H. However, the GDP fluxes with the F82H side on the upstream are comparable with that of pure F82 H.

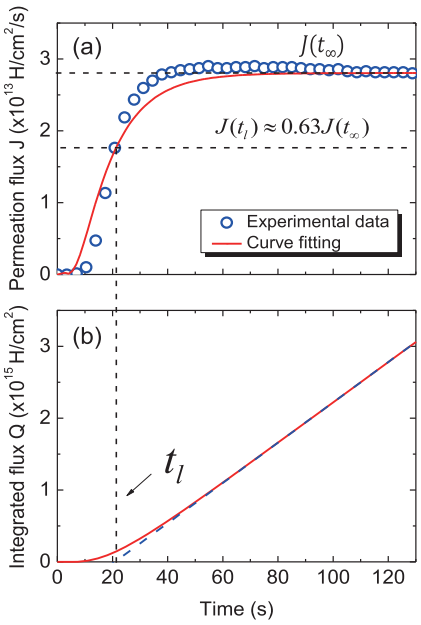

Fig. 2 (a) Curve fitting of the transient GDP flux and (b) time lag from time-integrated flux $Q(t)$. In this case, the thicknesses of W coating and F82H substrate are 0.09 mm and 0.5 mm, respectively and the temperature is 420 °C. The upstream hydrogen pressure is 760 Pa. The lag time is measured to be ~21 s.

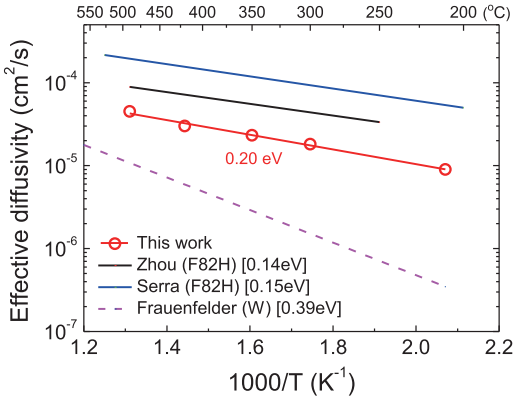

Fig. 3 Temperature dependence of hydrogen effective diffusivity for VPS-W coated F82 H. The thicknesses of W coating and F82H substrate are 0.09 mm and 0.5 mm, respectively. Zhou's [4], Serra's [11] and Frauenfelder's [12] data are shown for comparison.

Figure 6 shows the hydrogen permeation fluxes through 1 mm F82H coated with VPS-W coatings of different thicknesses. It is shown that the permeation fluxes for these two specimens are comparable with each other.

Usually, hydrogen permeation through materials is affected by both bulk and surface processes. In this study, F82H steel surface is covered by a layer of VPS-W coating. As it has been mentioned before, the typical VPS-W coating consists of partially melted W particles, melted W and substantial surface-connected porosity [8]. Even a

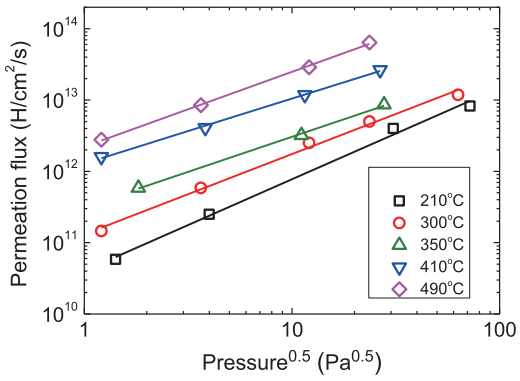

Fig. 4 Steady-state hydrogen GDP fluxes through a VPS-W coated F82H at the temperature range of 200 - 500 °C. The thicknesses of W coating and F82H substrate are 0.09 mm and 0.5 mm, respectively.

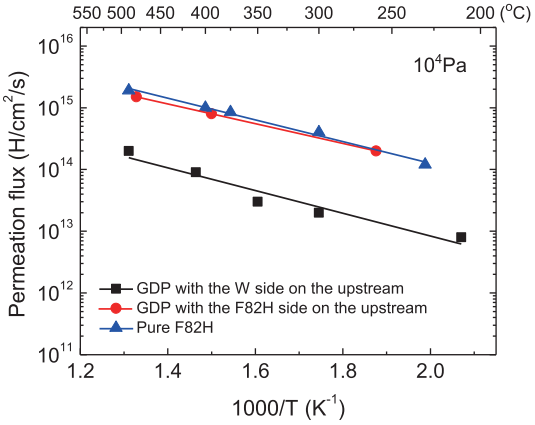

Fig. 5 Steady-state hydrogen permeation fluxes through VPS-W coated F82H (in both directions) and pure F82 H. The upstream hydrogen pressure is set at 10^4 Pa. The thicknesses of W coating and F82H substrate are 0.09 mm and 0.5 mm, respectively. A pure F82H specimen with thickness of 0.5 mm is used for comparison.

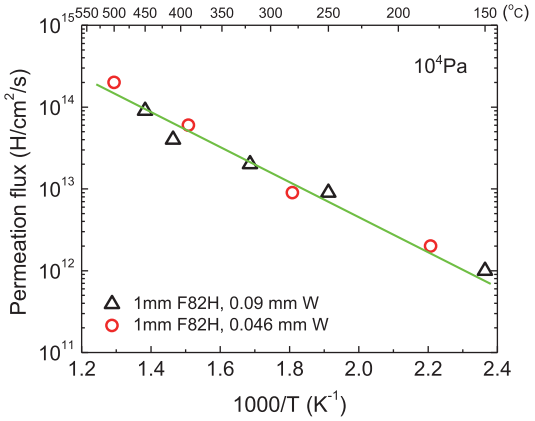

Fig. 6 Steady-state hydrogen permeation fluxes through 1 mm F82H coated with VPS-W coatings of different thicknesses (with the W side on the upstream).

200 μm thick layer of VPS-W has an open system of connected pores, which connects the front and rear surfaces of the deposited layer [6]. The density of the prepared VPS-W coating is ~90% of bulk W, which means the porosity is ~10%, including open (or connected) pores and closed pores. Thus, the results obtained in this study can be explained by molecular hydrogen permeation through open pores of VPS-W coating combined with permeation in a dissociated form through F82H substrate. That means, the open pores allow hydrogen gas reach to the W/F82H interface. Consequently, hydrogen dissolves at the interface and diffuses into the F82H substrate to allow hydrogen permeation after a certain period of time [14].

It is well known that the solubility of hydrogen in W is quite low compared to, for example, steels [15]. So one might assume that the bulk of W is practically impermeable for hydrogen under the given conditions. As it is shown in Fig. 5 that the permeation rates with the F82H side on the upstream are almost same with that of pure F82 H, which could be attributed to the same upstream surface area. Because the permeation rate is limited by the diffusion process in the F82H substrate, one might assume the hydrogen pressure at the W/F82H interface is ~0, i.e., hydrogen concentration at the interface is ~0. As a result, hydrogen GDP fluxes are not affected significantly by the 90 μm-thick porous VPS-W coating. But, the permeation rates with the W side on the upstream are ~7% compared to that of pure F82 H, which means the effective surface area of the F82H steel is reduced to ~7% by a porous W. In other words, the density of open pores is ~7% and ~93% of steel surface is covered by W (including closed pores). The main effect of the W coating on hydrogen permeation is to reduce the incoming flux at the W/F82H interface and the effective area for hydrogen dissolution in the substrate [16]. Therefore, the permeation rates through composite VPS-W/F82H specimen are independent of the W coating thickness, which is illustrated in Fig. 6.

Although the square-root dependence of permeation rates against driving pressure (Fig. 4) suggests the diffusion-limited permeation under the given conditions, the effective diffusivity shown in Fig. 3 is smaller than the values reported for F82H steel [4, 11]. These observations indicate that the permeation in transient states could be affected by also other factors such as surface contamination or trapping.

4. Conclusion

Hydrogen GDP experiments have been conducted for VPS-W coated F82 H. It has been found that the permeation rates with the W side on the upstream are reduced to ~7% compared to that of pure F82 H. However, the permeation rates with the F82H side on the upstream are comparable with that of pure F82 H. VPS-W coating has an open system of connected pores and the density is evaluated to be ~7%. For a fusion reactor, based on these data one

might predict that it is infeasible for VPS-W coatings to be used as hydrogen isotopes permeation barrier from the viewpoint of bred tritium gas-driven permeation through the first wall. In the future work, hydrogen permeation properties of physical vapour deposition W coatings will be studied.

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