Hydrogen Isotopes Plasma-Driven Permeation through Tungsten Coated Reduced Activation Ferritic Steel F82H

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Hydrogen isotopes plasma-driven permeation (PDP) through F82H coated with two different types of tungsten coatings, i.e., sputter-deposited tungsten (SP-W) and vacuum plasma-sprayed tungsten (VPS-W) has been studied in the temperature range of 300 - 550 °C. It has been found that hydrogen isotopes PDP fluxes through VPS-W coated F82H are reduced compared to that through bare F82H. However, the PDP fluxes through SP-W coated F82H are enhanced compared to bare F82H. Reduced or enhanced PDP fluxes are related to the different microstructure of tungsten coatings and its surface recombination characteristics.

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Tungsten (W) has been proposed as a candidate plasma-facing material for ITER divertor because of its beneficial properties such as high melting point, high thermal conductivity and low sputtering yield [1]. For a DEMO reactor, surface coatings made of W are necessary to protect the plasma-facing wall made of reduced activation ferritic steels such as F82H [2]. The characterization of hydrogen isotopes transport through W coated F82H is of crucial importance to evaluate major reactor design issues including tritium retention, breeding feasibility and first wall particle recycling. In this work, hydrogen isotopes PDP through F82H coated with two different types of W coatings are investigated and the effects of W coatings on hydrogen PDP are discussed. Such information has important implications for the use of W coatings as plasma-facing material.

Hydrogen isotopes PDP experiments are performed using a linear plasma device VEHICLE-1 [3]. The plasma density is $\sim 10^{10}\,\mathrm{cm}^{-3}$ and the electron temperature is ~5.5 eV. The incident ion energy is controlled by biasing the sample. A bias of 100 V has been used for PDP in the present work. Taking into account the ion species mix (H^+ : H_2^+ : H_3^+) and surface particle reflection, the net implantation flux is estimated to be $\sim 1 \times 10^{16}\,\mathrm{H/cm^2/s}$. SP-W and VPS-W coated F82H membranes are used as samples. The permeation area is 35 mm in diameter. The thickness of F82H subtract is 0.5 mm, and the thicknesses of SP-W and VPS-W coatings are 0.5 μm and 90 μm, respectively. A 0.5 mm thick bare F82H is used for comparison. Small samples with a size of 12 × 12 × 1 mm are also prepared for ex-situ analyses such as scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDS) and X-ray diffraction (XRD). Notice that the thicknesses of W coatings of the small samples are different from that of the permeation membranes.

Figure 1 shows the SEM images of the investigated samples. SP-W coatings are prepared by argon plasma sputtering at temperature of ~300 °C. The density of SP-W coatings is evaluated to be ~19.2 g/cm^3, ~99.5% of bulk W. As shown in Fig. 1 (a) that the SP-W coating is well organized with an average crystal size of ~100 nm. EDS analyses yield the as-received surface elemental composition of: C (0.7 wt%), O (0.1 wt%), and W (99.2 wt%). Columnar W grains are observed from the cross-section view shown in Fig. 1 (b) and the W/F82H interface is sharp without voids, cracks or other defects. XRD analyses indi-

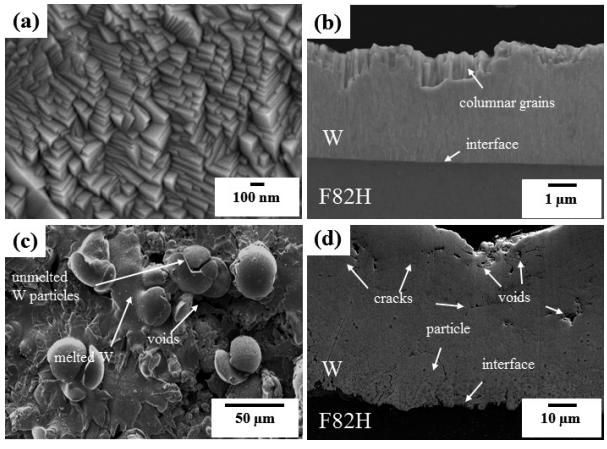

Fig. 1 SEM images of surface (a) and cross-section (b) of SP-W coatings, surface (c) and cross-section (d) of VPS-W coatings.

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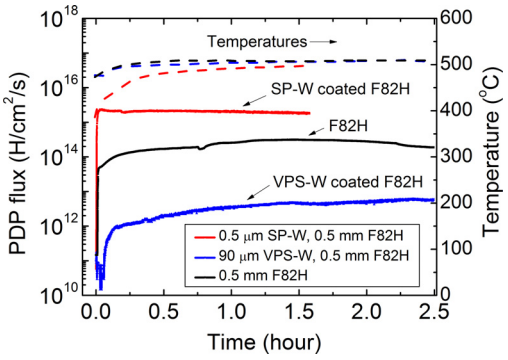

Fig. 2 Hydrogen PDP breakthrough curves through F82H with and without W coatings at ~500 °C.

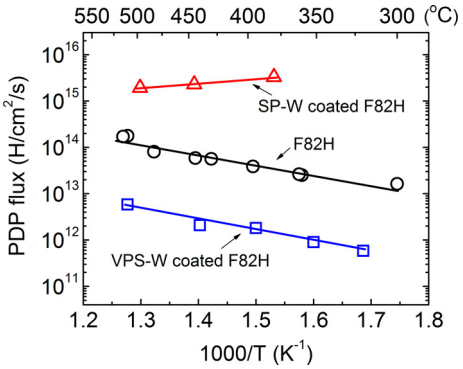

Fig. 3 Temperature dependence of steady-state hydrogen PDP fluxes through F82H with and without W coatings.

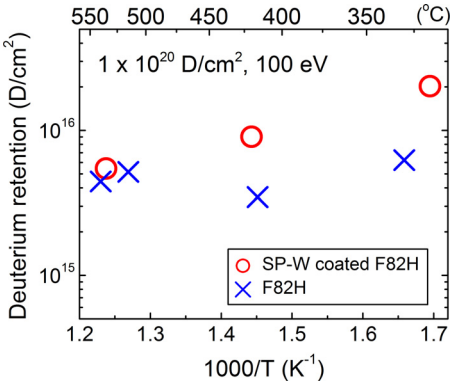

Fig. 4 Deuterium retention in SP-W coated F82H and bare F82H measured by TDS.

cate a strong (110) preferred orientation. VPS-W coatings are deposited at ~600 °C. The average size of W powder particles is ~25 μm. As shown in Figs. 1 (c, d) that VPS-W coatings have an inhomogeneous microstructure, i.e., a mixture of disorganized areas composed of large unmelted W particles, fine randomly melted W areas and void regions. Voids or pores are observed primarily next to the unmelted particles. The "as-fabricated" VPS-W surface is dull-colored and EDS analyses indicate surface impurity elemental composition is dominated by carbon. VPS-W coatings have a relatively low density structure with connected porosity [4]. The average density is evaluated to be ~90% of bulk W.

Shown in Fig. 2 are hydrogen PDP breakthrough curves through F82H with and without W coatings at ~500 °C. Not presented here are similar data taken on deuterium PDP experiments. The observed PDP flux through VPS-W coated F82H is more than one order of magnitude lower than that of bare F82H. Hydrogen permeation is observed shortly after the plasma-on time and the permeation flux continued to rise gradually until the implantation is terminated. In contrast, hydrogen PDP flux through SP-W coated F82H is significantly high and the permeation transient is much faster than those observed for VPS-W coated F82H or bare F82H.

Shown in Fig. 3 are the temperature dependence of hydrogen PDP fluxes from 300 to 550 °C. It is shown that VPS-W coatings reduce hydrogen PDP fluxes relative to that of bare F82H. The readers are referred to reports by Anderl [5], Otsuka [6] and Golubeva [7] for the permeation properties of hydrogen through VPS-W. Reduced permeation fluxes are attributed to the complex microstructure and a substantial surface-connected porosity.

The results shown in Fig. 3 for SP-W coated F82H indicate enhanced permeation fluxes. Notice, however, that the PDP flux decreases with increasing temperature. There are a number of experimental studies [5, 8] showing that coatings can also increase the permeation flux when exposed to ions or plasma. Enhanced PDP fluxes are believed to be attributed to its surface recombination characteristics. Hydrogen ions or plasma reaching either surface are not guaranteed immediate release. At the plasma-facing surface, with the exception of extremely high temperatures, recombination of the atoms into molecules must occur before release is possible. The hydrogen recombination process is expressed as [9]:

$$J_{-} = K_r C^2, \tag{1}$$

where J_{-} is the recombination flux, K_r is the recombination coefficient and C is the hydrogen concentration at the near surface. Recombination coefficient is extremely sensitive to the surface condition, such as surface morphology and contamination etc. For example, the reported data for stainless steel span several orders of magnitude [10]. A material with a high recombination coefficient promotes rapid release from the surface, keeping the overall hydrogen concentration low throughout the material. A material with a low recombination coefficient prevents rapid release, elevating the hydrogen concentration throughout the material. The low recombination coefficient leads to higher hydrogen inventory in the material with potentially higher permeation fluxes through the plasma-facing material.

Shown in Fig. 4 is the temperature dependence of deuterium retention in SP-W coated F82H and bare F82H

exposed to 100 eV deuterium plasma, with a fluence of 1×10^{20} D/cm^2. The amount of retained deuterium in SP-W coated F82H is a factor of 3 higher than that of bare F82H in the temperature range of 320 - 420 °C and the differences become smaller with increasing temperature. As has been reported that the evaluated hydrogen solubility in SP-W coatings [11] is nearly two orders of magnitude higher than that in F82H [12]. It is believed that the significantly high hydrogen solubility accounts for the enhanced PDP fluxes. Nevertheless, further investigations are still needed to address these issues.

In this study, hydrogen isotopes PDP behavior through W coated F82H has been studied. The PDP fluxes through VPS-W coated F82H are reduced. However, SP-W coatings tend to enhance hydrogen PDP fluxes. Characterization analyses indicate that microstructure and surface condition of the coatings significantly alter hydrogen permeation fluxes. Surface recombination is an important process determining the PDP flux.

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