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Effect of helium irradiation on deuterium permeation behavior in tungsten

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ABSTRACT

In this study, we measured deuterium (D) gas-driven permeation through tungsten (W) foils that had been pre-damaged by helium ions (He^+). The goal of this work was to determine how ion-induced damage affects hydrogen isotope permeation. At 873 K, the D permeability for W irradiated by 3.0 keV He^+ was approximately one order of magnitude lower than that for un-damaged W. This difference diminished with increasing temperature. Even after heating to 1173 K, the permeability returned to less than half of the value measured for un-damaged W. We propose that this is due to nucleation of He bubbles near the surface which potentially serve as a barrier to diffusion deeper into the bulk. Exposure at higher temperatures shows that the D permeability and diffusion coefficients return to levels observed for undamaged material. It is possible that these effects are linked to annealing of defects introduced by ion damage, and whether the defects are stabilized by the presence of trapped He.

1. Introduction

Tungsten (W) is a promising candidate for plasma facing materials (PFMs) in future fusion reactors [1,2] due to low hydrogen isotope retention and high melting temperature. A comprehensive understanding of the behavior of hydrogen isotopes in W is crucial for evaluating the safety of future fusion reactors [3]. Hydrogen diffusion and permeation in un-damaged W has been studied since the late 1960's (Frauenfelder: [4,5]) and early 1970's (Zaharakov: [6]). For further details on more recent developments, we refer the reader to Ref. [7–9]. Investigations of high-flux plasma-driven permeation using tritium [10] are also planned in the near-future. However, for a variety of reasons, tritium-based measurements of this nature are quite complex. Experiments that use deuterium gas-driven permeation provide complementary information, and can operate at relevant temperatures to provide useful comparisons between different plasma-facing materials (including advanced alloys, as described in Ref. [11].)

When considering permeation measurements, it is important to keep in mind the complexity of the fusion environment. In addition to exposure to high-flux, low energy D + T plasmas, the PFMs will also be exposed both energetic helium (He) and 14 MeV neutrons that escape from the plasma. The damage introduced by this irradiation will strongly affect how hydrogen migrates through the material. In particular, He^+ irradiation causes the formation of pressurized He bubbles. Prior temperature-programmed desorption and depth profiling measurements indicate that deuterium (D) diffusion into the W bulk is reduced considerably as a result of mixed species irradiation. The prevailing hypothesis is that the dense layer of He bubbles formed near the W surface [12,13] strongly influences this migration.

In our previous study [14], we examined the effect of He^+

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irradiation on D retention in W. We proposed that the higher thermal stability of He-trapped dislocation loops interfered with the complete recovery of damage at 1173 K. The growth of He bubbles was remarkable above 1073 K and damage recovery was not possible even at 1173 K. Given these retention results, the next logical step (in the context of the effect of fuel behavior on fusion reactor operation) is to evaluate what effect He bubbles have on hydrogen isotope permeation. This is the focus of the present study, which includes a comparison of hydrogen permeation in both He^+ irradiated and un-damaged W.

2. Experimental configuration

Fig. 1 shows a diagram of the gas-driven permeation instrument at Shizuoka University. This instrument includes a sample holder that can accommodate 10 mm diameter specimens secured with fittings for 6.35 mm VCR tubing. To create a tight seal, the specimen is sandwiched between two silver coated metal gaskets [15]. A thermocouple was introduced through the inner tubing and directly contacted the upstream side of the specimen to measure its temperature. The upstream D_2 gas pressure is regulated by a variable leak valve (V. L.V.) and monitored by capacitance manometers. The vacuum at the downstream side was maintained at ~10^{-6} Pa by a turbomolecular pump (TMP) backed by a rotary pump (RP). The sample holder was contained within an evacuated quartz tube to reduce oxidation and prevent any adventitious signals arising from the D permeation to the surrounding environment during heating. The TMP and RP maintained a vacuum within the quartz tube of less than 10^{-4} Pa. Thereafter, D_2 gas was introduced to the upstream side. The D permeation to the downstream side was quantified by a MKS Microvision quadrupole mass spectrometer (QMS) to monitor mass 4 (D_2) and mass 3 (HD) peaks [16]. The D_2 sensitivity of the QMA was calculated by two standard leaks with different leak rates. The same sensitivity was assumed for the D_2 and HD peaks.

The permeability (P) is expressed by:

$$P = \frac{xJ}{AP_D^n} \tag{1}$$

where J is steady-state permeation flux, x is sample thickness, A is surface area and P_D is the deuterium driving pressure. A pressure exponent n is calculated by measuring the permeation fluxes at different driving pressures as follows,

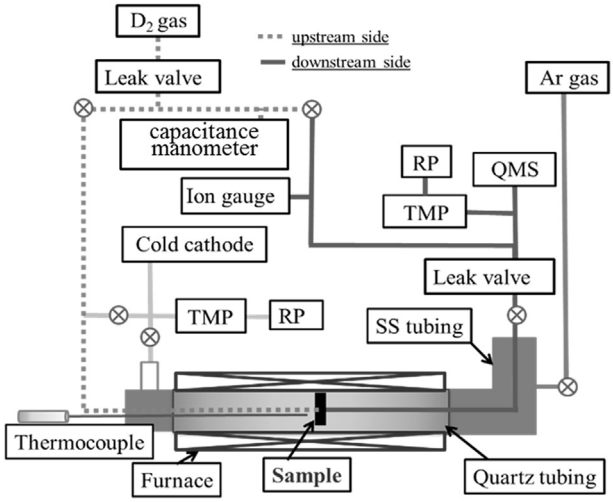

Fig. 1. The diagram of the gas-driven permeation measurement instrument.

$$J \propto P_D^n \tag{2}$$

The permeation controlled by diffusion-limited regime satisfies the equation with $n = 0.5$. Likewise, when the permeation rate is limited by surface processes, the exponent of the deuterium driving pressure P_D is unity ($n = 1$). In addition, the time evolution of the permeation flux can be used to determine the diffusion coefficient D analytically using the following equation:

$$t_{lag} = \frac{x^2}{6D} \tag{3}$$

Here t_{lag} is determined by where a line fit to the asymptotic region of the permeation flux intersects the time axis [17].

The test specimens consisted of rolled W foils (Nilaco Co. Ltd) cut into discs (10 mm dia. × 0.035 mm thick) with snips. They were cleaned in ultrasonic bath with ethanol and preheated at 1173 K for 30 min under ultrahigh vacuum (<10^{-6} Pa) to remove impurities and residual damage. Furthermore, an additional set of 3 mm dia. discs were thinned by pierce punch and electrolytic polishing for transmission electron microscopy (TEM) examination.

The W samples were then exposed to varying He^+ irradiation conditions to cover a range of energies and ion fluences. For the first set of experiments, 1.0 or 3.0 keV He^+ was implanted into the sample at room temperature (flux: 3.0×10^{17} He^+ m^{-2} s^{-1}, fluence: 3.0×10^{21} He^+ m^{-2}). Additional W samples were exposed to a fixed He^+ energy of 3.0 keV, with the fluence ranging between 0.03 and 9.0×10^{21} He^+ m^{-2}. Following the pre-irradiation step, each sample was then installed into the gas-driven permeation instrument and exposed to D_2 pressures ranging between 10.00 and 120.0 kPa. We then measured permeation fluxes over a temperature range of 873–1173 K in each case.

To correlate the permeation results with the defect structure created by He^+ damage (and ameliorated by heating), we used transmission electron microscopy (TEM) (JEM 2000EX, JASCO Inc.) In addition, D depth profiles of He^+ irradiated W after exposure to D_2 gas at 873 K and 1173 K for 2 h were also evaluated by the Glow Discharge-Optical Emission Spectroscopy (GD-OES) (GD-Profiler 2, HORIBA Ltd.) measurements at the University of Toyama.

3. Results and discussion

3.1. He^+ energy dependence

Fig. 2 shows D depth profiles of He^+ irradiated W after exposure

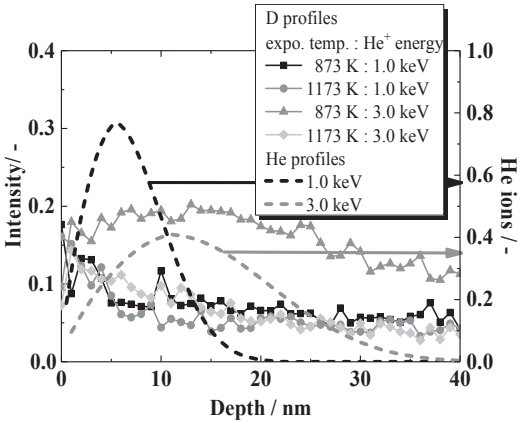

Fig. 2. D and He depth profiles for each sample by GD-OES measurement and SRIM calculation, respectively.