The Atom-driven Transport of Deuterium in Nb under the Influence of Surface Impurities

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Abstract

Atom-driven permeation behavior of deuterium in Nb was studied experimentally. The measurements were performed under two different vacuum conditions which resulted in contrasting surface compositions: one resulted in a surface rich in carbon (C) after atom beam irradiation and the other rich in sulfur (S) which was unaffected by the irradiation. The result of permeation measurements revealed that when the specimen surface was covered by sulfur the deuterium permeation rate weakly depended on the temperature, whereas when the surface was rich in carbon and oxygen it tended to decrease sharply for temperatures below $\approx$ 700 K.

1 Introduction

The interaction of atoms with solid surfaces is an area where the current database for fusion applications is lacking [1]. Hydrogen atoms produced through thermal dissociation process in an atomizer heated up to temperatures around 2350 K, have translational energies around 0.3 eV. Hence, in contrast to energetic ion beams, such a low energy of incidence will not produce defects in the specimen, and their transport may be affected by additional factors such as chemical interactions between adatoms and surface atoms.

The purpose of the present study is to investigate the interaction of ionic and atomic deuterium with the surface of Nb, with particular emphasis on the effect of surface impurities on the observed deuterium transport.

2 Experimental

As shown in Fig.1, an atomic beam source (ABS) has been installed in our existing hydrogen permeation experimental device. This facility has two quadrupole mass spectrometers (QMS) for dynamic measurement of the deuterium fluxes (HD and D_2) in both the upstream-side and downstream-side chambers (indicated as C1 and C2 in the figure, respectively), together with AES (Auger electron spectroscopy) or SIMS (secondary ion mass spectrometry) analyzing systems.

The deuterium was employed as a source of the atomic beam, and the deuterium atomic flux, ϕ_a (D m^{-2} s^{-1}), to the specimen was thus evaluated to be 2.5×10^{17} D m^{-2} s^{-1} at a nozzle temperature 2350 K. The specimen employed in the present study is a Nb foil (supplied by Nilaco Co. Ltd., 0.1 mm$t$, 14 mmϕ, 99.9 at% in purity), which was welded on the top of a cylindrical sample holder made of type 304 SS. The specimen was cleaned ultrasonically in acetone before being installed in the vacuum vessel. The AES or SIMS analyses were performed occasionally in order to monitor the surface impurities on the specimen, while the specimen temperature was varied from 600 to 1000 K.

3 Results and Discussion

The temperature dependence of steady state deuterium permeation rate was measured in the temperature range between 600 and 1000 K, and the results are shown in Fig.2. In this figure, the results of measurements performed under different vacuum conditions are compared. A weak temperature dependence of the permeation rate, with an apparent

activation energy of 16.5 kJ mol^{-1}, which tends to saturate at higher temperatures, was observed in the case of the sulfur-rich surface (open symbols). In the case of the carbon-rich surface (closed symbols), a rather sharp decrease of permeation rate, with an apparent activation energy of 84.5 kJ mol^{-1}, was observed below 700 K.

The observed phenomena may be explained as follows. Under steady state conditions, when the transport of hydrogen is limited by surface recombination at both sides, the molecular permeation rate, $\Phi_{\rm P}$ (D_2 m^{-2} s^{-1}), may be expressed by

$$\Phi_{\rm P} = \xi \frac{\alpha_2}{\alpha_1 + \alpha_2} \left(\frac{\phi_{\rm a}}{2} \right) \ , \tag{1}$$

where $\phi_{\rm a}$ is the atomic flux, ξ is the "retention" fraction of atoms on the surface, and α_1 and α_2 are the probabilities of dissociative adsorption at the upstream-side and downstream-side, respectively [2]. Since ξ is considered to be independent of temperature, T, the above equation would predict the permeation rate to be independent of T, assuming that the surface conditions are the same; i.e. $\alpha_1 \approx \alpha_2$.

Although Eq.(1) may be considered to be consistent with the experimental results in general, it does not predict the decrease of the permeation rate at lower temperatures in the case of the carbon-rich surface (closed symbols in Fig.2). In order to be consistent with the experimental result one must assume that the deposition of carbon during atom beam irradiation has resulted in the decrease of ξ. Either due to the increase of surface potential barrier by carbon deposition, or to the increased retention by impurity layer, which is likely to become pronounced at lower temperatures, the entry of atomic deuterium into the bulk of Nb may have been limited.

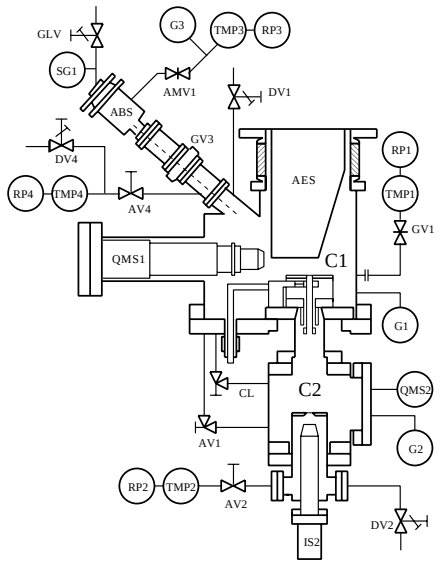

Fig.1: Schematic drawing of experimental device (HYPA-IV).

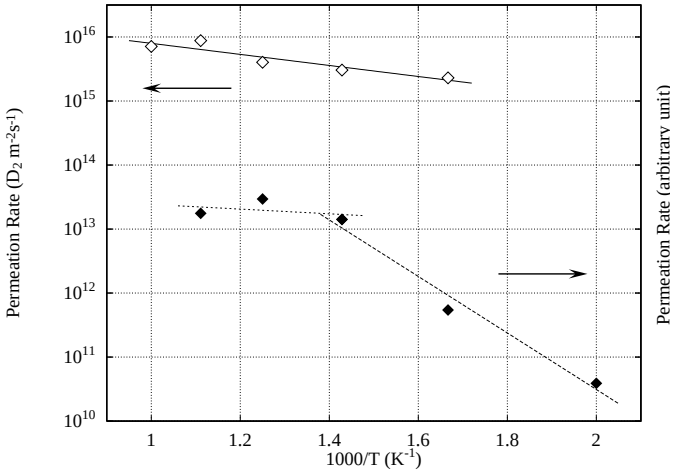

Fig.2: Temperature dependence of atom-driven deuterium permeation rate from Nb. The measurements performed under different vacuum conditions are compared.

References

[1] W. Eckstein, J. Nucl. Mater. 248 (1997) 1.

[2] A.I. Livshits, M.E. Notkin, A.A. Samartsev, J. Nucl. Mater. 170 (1990) 79.