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Experiment on atomic hydrogen reflection by use of a permeation probe

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Abstract

Experiments on reflection behavior of atomic hydrogen with lower energies were conducted by use of a palladium membrane as a probe for detecting atomic hydrogen. The palladium membrane was exposed to a deuterium rf-plasma, and reflectors were located between them. Hydrogen atoms easily permeated through palladium, which acted as a window for hydrogen atoms. By changing the arrangement of the reflectors of nickel, graphite, glass, tungsten and polytetrafluoroethylene, the corresponding permeation flux was observed, and from which the relative value of the incident atoms to the membrane was estimated. The incident atoms reflected at least once by nickel were nearly the same as those which were reflected more than twice. This indicated that an atomic reflection was very significant. The other reflector materials also showed high atomic reflection. These results may mean that atomic hydrogen is transported for a long distance in openings of structures in fusion devices.

1. Introduction

For evaluation of hydrogen recycling in fusion devices, it is important to know reflection behavior of atomic hydrogen particles from plasma-facing materials. Many experimental works [1–3] on the behavior of energy and particle reflection have been conducted in the energy of keV region and the computer codes such as MARLOWE [4] and TRIM [5] have been successful to reproduce the experimental data [6]. However, these codes would not be applied in lower energy region [7], where few experimental data are available.

Baskes [8,9] has simulated the chemical interactions and shown that, in lower energy region, typically a few eV, the particle reflection coefficient of hydrogen from a clean nickel surface is considerably lower than that by the TRIM code, and almost no hydrogen particles are reflected as atoms but desorbed as molecules when the surface is covered with hydrogen. These results are generally considered to be more plausible.

Some experimental results of the plasma-driven permeation [10,11], however, suggest that some hydrogen particles are reflected as being atoms even in lower energies. So, in the present work, we experimentally studied the reflection behavior of atomic hydrogen using a technique of the permeation probe and show that a degree of atomic reflection is higher than that generally considered.

2. Experimental

2.1. Permeation probe

One of the difficulties of the reflection experiment in lower energy region would be the way of detecting atomic hydrogen. In the present work, a palladium membrane is used as a probe for atomic hydrogen, of which technique has been developed by Winter et al. [12,13]. Hydrogen atoms significantly permeate through some metals such as palladium, iron and nickel, which is known as the superpermeation [14] or the plasma-driven permeation while thermalized hydrogen molecules do not. The reason for palladium is that the diffusion coefficient of hydrogen isotopes is relatively high and hence the permeation is expected to be limited by the recombination process. Another reason is that palladium is one of noble metals and does not need cleaning

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treatment such as ion bombardment and hydrogen flushing [13] before experiments.

In the recombination-limited permeation, from particle balance, the permeation flux J_f at the steady state is expressed by

$$J_f = \frac{k_d}{k_u + k_d} F_f, \tag{1}$$

where F_f is the incident hydrogen flux, k_u and k_d are the recombination coefficients of hydrogen on the upstream (hydrogen-incident side) and the downstream (hydrogen-permeation side), respectively. Although the values of k_u and k_d are unknown in the present work, the relative changes in F_f can be known from the observed value of J_f.

Prior to a reflection experiment, characteristics of palladium with respect to hydrogen permeation were studied. As the experimental setup has been described elsewhere [11], the procedure will be briefly explained. A palladium membrane with a thickness of 0.1 mm was set between two vacuum chambers and the upstream side was exposed to a deuterium rf-plasma. A mechanical shutter with large and small holes was located between the membrane and the plasma, and time-dependent change in the permeation flux through the membrane was monitored when the shutter was moved from the small to the large hole or vice versa.

When the initial flux of the incident hydrogen F_i is abruptly changed to F_f at $t = 0$, the following differential equation is obtained in the recombination-limited permeation [15]:

$$L \frac{dC(t)}{dt} = F_f - (k_u + k_d) C^2(t), \tag{2}$$

where $C(t)$ is the hydrogen concentration in the membrane and L the membrane thickness. Under the boundary conditions of $F_i = (k_u + k_d) C_i^2$ and $F_f = (k_u + k_d) C_f^2$, where C_i and C_f are the hydrogen concentration at $t \leqslant 0$ and $t \to \infty$, respectively, the solution of Eq. (2) is

$$J(t) = J_f \left(\frac{A(t) - 1}{A(t) + 1} \right)^2, \tag{3}$$

$$A(t) = \frac{1 + \sqrt{J_i/J_f}}{1 - \sqrt{J_i/J_f}} \exp(\lambda t), \tag{4}$$

$$\lambda = \frac{2(k_u + k_d)\sqrt{J_f}}{L\sqrt{k_d}}, \tag{5}$$

where J_i is the steady-state permeation flux at $t \leqslant 0$. To derive the solution, the relations of $J_i = k_d C_i^2$ and $J_f = k_d C_f^2$ are taken into account. Note that J_f is always larger than zero, the solution in the case of $J_f = 0$ is not considered.

Between the membrane temperature of 398 and 568 K, the time-dependent change in the permeation flux was fairly reproduced with a fitting parameter of λ, as typically shown in Fig. 1 where the membrane temperature was 398 K and $\lambda = 4.1 \times 10^{-3}$ s^{-1}. It was confirmed that the permeation was limited by the recombination process under our experimental conditions and the membrane could be used as the probe for atomic hydrogen.

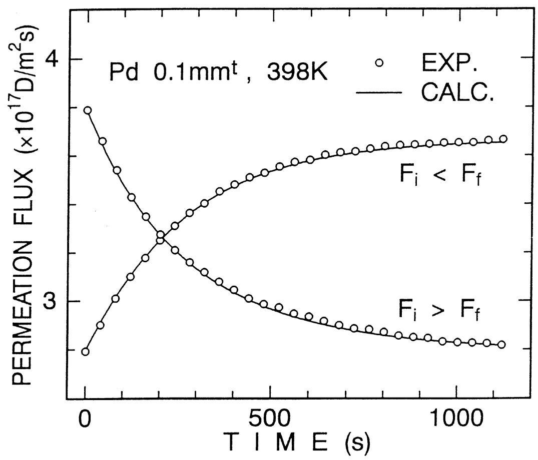

Fig. 1. Transient curves of deuterium permeation through the palladium membrane when the incident flux increased ($F_i < F_f$) and decreased ($F_i > F_f$).

2.2. Reflection experiment

The essential part of the setup for the reflection experiment is the same as that for the permeation experiment explained in Section 2.1. Instead of the mechanical shutter, some reflectors of a cylinder and/or disks are located with several arrangements as schematically shown in Fig. 2. The permeation probe of the palladium membrane is set between two vacuum chambers. After the probe is heated up, the upstream side of it is exposed to a deuterium rf-plasma produced in a discharge tube with a nozzle, and deuterium permeated to the other side is monitored by a quadrupole mass analyzer. By changing the arrangements of the reflectors, the corresponding permeation flux at the steady state is observed.

The thickness, the diameter and the temperature of the palladium membrane are 0.1 mm, 16 mm and 452 K, respectively. Conditions of the plasma are kept constant during the whole experiments. The mean energy of atomic deuterium from the plasma is difficult to be measured but considered to be around a few eV.

3. Results and discussions

3.1. Reflector arrangements

In order to demonstrate a degree of atomic reflection, the arrangement of nickel reflectors was sequentially

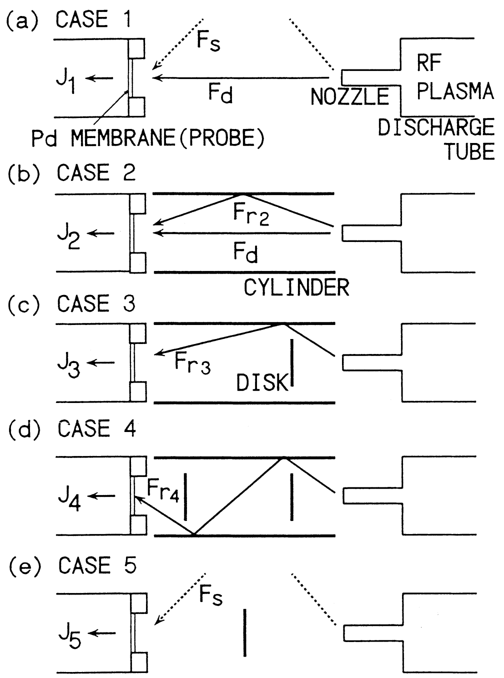

Fig. 2. Schematic showings of reflector arrangements between the palladium probe and the deuterium plasma in the discharge tube.

changed from case 1 to 4 (see Fig. 2). First of case 1, there were no reflectors between the probe and the top of the nozzle, of which distance was 99 mm. The permeation flux J_1 through the probe at the steady state was 2.0×10^{18} atoms m^{-2} s^{-1}. The direct flux F_d and the scattered flux F_s, which will be explained later, would contribute to J_1. Next, in case 2, a nickel cylinder with a inner diameter of 32 mm and a length of 86 mm was inserted between the probe and the nozzle. The reflection flux F_{r2} as well as F_d contributed to the permeation flux J_2, which was 4.4×10^{18} atoms m^{-2} s^{-1} and 2.2 times larger than J_1.

In case 3, a nickel disk with a diameter of 18 mm was set into the cylinder so that F_d was fully blocked and only the reflection flux F_{r3} impinged to the probe. The distance between the disk and the nozzle was 13 mm. J_3 was 3.1×10^{18} atoms m^{-2} s^{-1}, not became zero. Another disk was added to a position of 75 mm from the nozzle in case 4. The deuterium atoms in the reflection flux F_{r4} were reflected at least twice before impinging to the probe. J_4 was 2.5×10^{18} atoms m^{-2} s^{-1}, still larger than J_1. A series of case 1–4 was repeated and a good reproducibility of the permeation flux was observed. When the discharge was turned off, the permeation flux decreased to zero.

These results indicated that a large number of deuterium atoms was reflected many times as being atoms by the cylinder and the disks, since deuterium molecules, thermally desorbed from the surface, would not contribute to the permeation flux under our experimental conditions. In fusion devices, atomic hydrogen isotopes may be transported for a long distance in openings of structures even if they are not faced to the plasma. The nickel surface, continuously exposed to the deuterium plasma, would be covered by deuterium atoms [16], so the above results might contradict to the Baskes' calculation [9], in which the incident hydrogen atom tends to recombine with a hydrogen atom on the surface and to be desorbed as a molecule.

The atomic reflection coefficient is defined here as a probability that hydrogen atoms are reflected as being atoms. If it was low enough, for example 0.2, a probability of reflection for atoms which were reflected twice is 0.04 and the permeation flux J_4 would have been close to zero. Hence the atomic reflection coefficient should be much higher value, may possibly be close to unity. Quantitative estimation of the coefficient is, however, difficult since there is lack of information such as angular distributions of the emitted atoms from the nozzle and of the reflected atoms. So further experiments are needed.

3.2. Reflection from some materials

Similar reflection experiments were conducted with some reflector materials of nickel, graphite, borosilicate glass, tungsten and polytetrafluoroethylene (PTFE) in the arrangement of cases 1–3 and 5. The size and position of reflectors were different from those in Section 3.1, that is, the distance between the probe and the nozzle 38.5 mm, an inner diameter and a length of a cylinder 32 and 25 mm, respectively, a diameter of a disk 18 mm, and the distance between the disk and the nozzle 16.5 mm. The thickness of each material was 0.1 mm for nickel and tungsten, 1 mm for PTFE and 2 mm for graphite and glass.

Two series of the arrangements were sequentially changed and the permeation flux observed in each case of the second series was almost the same as that in the first series, as typically shown in Fig. 3. The data are summarized in Table 1. For the sake of simple description, the permeation flux is expressed by arbitrary unit in Fig. 3 and Table 1.

When the proportional constant of $k_d/(k_u + k_d)$ in Eq. (1) is denoted by a, the following equations are derived:

$$J_1 = a(F_d + F_s), \quad (6)$$

$$J_2 = a(F_d + F_{r2}), \quad (7)$$

$$J_3 = aF_{r3}, \quad (8)$$

$$J_5 = aF_s, \quad (9)$$

where J_i represents the permeation flux in the ith case. Using Eqs. (6)–(9), the incident fluxes were estimated

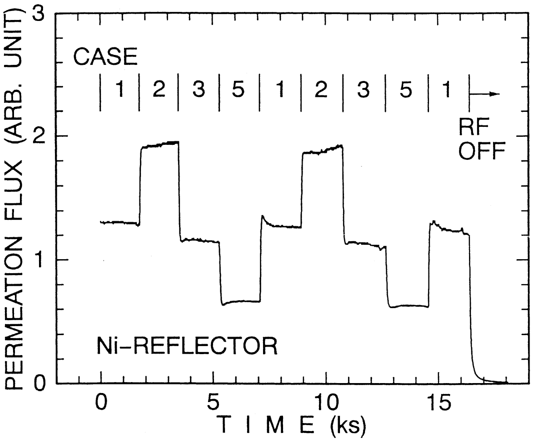

Fig. 3. The observed permeation flux for the nickel reflectors when the arrangement was sequentially changed.

Table 1
The permeation flux in some reflector arrangements and the ratio of the reflection flux to the direct flux estimated

Material	Ni	Graphite	PTFE	W	Glass
J_1	1.30	1.00	1.40	2.07	2.08
J_2	1.93	1.55	1.90	2.74	3.09
J_3	1.17	0.85	1.21	1.70	2.03
J_5	0.67	0.50	–	–	–
F_{r2}/F_d	2.06	2.10	1.79	1.74	2.06
F_{r3}/F_d	1.86	1.70	1.78	1.70	2.01
F_s/F_d	1.06	1.00	(1.06)	(1.06)	(1.06)

with the unknown parameter of a. As the direct flux F_d was considered to be constant during the whole experiment, the ratio of the reflection flux to F_d is listed in Table 1.

In case 5, only the disk was located and the incident flux F_s would be attributed to reflection by the inside of the vacuum chamber, made of stainless steel [11]. For nickel and graphite, F_s was comparable to F_d, which means significant reflection of deuterium atoms by stainless steel. The permeation flux J_5 was not observed for glass, tungsten and PTFE so the incident fluxes were estimated assuming that the ratio of F_s/F_d was equal to 1.06 for nickel reflectors since F_s and F_d would not be affected by the reflector materials.

The fact of F_{r3}/F_d for each material being nearly the same suggested that a degree of atomic reflection was almost independent of the reflector materials. One of possible reasons may be that the surface was covered with deuterium so that the impinging atoms with lower energies was not affected by the original surface property with respect to reflection.

After the experiment, the deuterium concentration near the surface of the graphite disk was observed by the nuclear reaction analysis (NRA) with the $D(^3He,p)^4He$ reaction [17]. An incident energy of a probing beam of $^3He^+$ ions was 1.3 MeV and a scattering angle was 135°. A normalized yield of protons, which represented a relative deuterium concentration, from the plasma-exposed surface (8.09×10^3 $\mu C^{-1} sr^{-1}$) was almost the same as that from the back surface (9.23×10^3 $\mu C^{-1} sr^{-1}$). As deuterium molecules are not effectively absorbed on graphite at lower temperatures, the NRA measurement indicated that deuterium atoms were reflected many times as being atoms, just as the case of nickel described in Section 3.1.

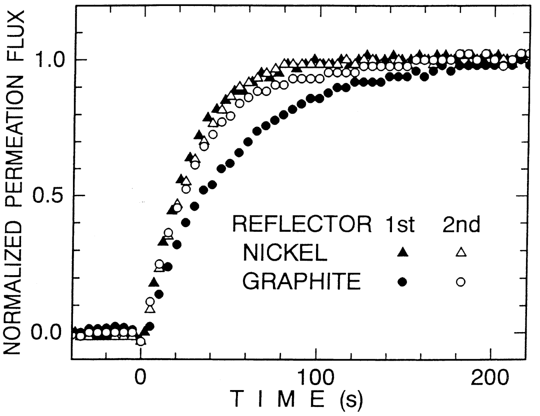

Fig. 4. Transient curves of deuterium permeation for the nickel and the graphite reflectors when the arrangement was changed from case 1 to 2.

Fig. 4 shows transient curves of the normalized permeation flux for the nickel and the graphite reflectors when the arrangement was changed from case 1 to 2. The curves for nickel and the second series for graphite corresponded to the transient permeation curve described in Section 2.1 The curve of the first series for graphite was, however, slowly rising compared with the other curves. This would be because that the graphite surface in the first series was free from deuterium and the atomic reflection was very low while the surface was covered with deuterium in the second series, as observed by the NRA measurement, and the atomic reflection became higher.

4. Summary

Experimental studies on atomic reflection of deuterium by some materials were performed using a palladium membrane as a probe for detecting atomic deuterium. It is generally considered that a hydrogen atom with lower energies, typically a few eV, tends to recombine and subsequently be desorbed as a molecule on solid surfaces which are covered with hydrogen. The results, however, indicated that the atomic reflection was very significant because a large amount of deuterium atoms survived recombination although they were reflected at least twice. The atomic reflection of nickel, graphite, PTFE, tungsten and borosilicate glass seemed to be similar but it was relatively low for graphite when the surface was free from deuterium. Further experiments are needed to study more quantitatively on atomic reflection of hydrogen isotopes with lower energies.

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