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Implantation driven permeation behavior of deuterium through stainless steel type 316L

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Abstract

Implantation driven permeation (IDP) behavior has been investigated for deuterium implanted into 316L SS. The experimental results showed that the IDP behavior at the steady state was controlled by the RD regime at low temperature and seemed to change to the RR regime at high incident ion energy region with increasing the temperature. The significant incident ion energy dependence of IDP was also observed at the steady and transient state. This appears to result from change of conditions such as effective diffusion and/or recombination coefficient at implanted region by the irradiation damages caused by deuterium implantation. The experimental results were compared with those of 304 SS reported previously. The results show the advantage of 316L SS for tritium permeability under the low tritium ion implantation compared to 304 SS.

1. Introduction

From a viewpoint of safety evaluation of fusion reactors, estimation of permeation amounts of tritium to coolant through plasma facing components is one of the critical issues. To simulate tritium permeation, a number of experimental studies on Implantation Driven Permeation (IDP) phenomenon using hydrogen isotopes have been carried out extensively for candidate metals of first walls by ion beam [1–4] or discharge method [5–8]. The typical energy region of incident particles used in those experiments was greater than a few keV in the ion beam method or lower than a few 10 eV in the discharge methods. From design studies for ITER (International Thermonuclear Experimental Reactor), however, incident energies of tritium implanted into the first wall are supposed to be distributed below 1 keV [9]. This means that IDP data below 1 keV should be more important to estimate amounts of tritium permeated into the first wall coolants. Although a number of experimental data for IDP phenomenon have been acquired, data observed below 1 keV are limited until now. Therefore, it would be desired to accumulate IDP data of tritium in incident energy region below 1 keV for candidate materials for first walls of fusion reactors. Authors have investigated the IDP behavior of deuterium implanted into various metals with low incident energy below 2 keV and relatively high flux deuterium ions in order to obtain knowledge for mechanism of IDP in a view point of estimation of tritium permeation [10–12].

This paper reports the IDP behavior of deuterium implanted into 316L SS, which is a candidate material as the vacuum vessel of fusion reactors, at the steady state and transient state. From the experimental results, the control regime of the IDP behavior is determined. The results are also compared with those for 304SS.

2. Experimental

The experimental apparatus for the IDP experiments consists of the following five sub-systems: (1) an ion source for production of a hydrogen isotope ion beam with high flux and variable low energies, (2) a main chamber system for ion implantation, (3) a measuring system of the permeated gases through the target, (4) hydrogen isotope gas supply and recovery system, and (5) a vacuum pump system.

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The ion source was a modified quartz capillary duoPIGatron designed by Isoya [13] which can produce low energy ions with high intensity. More than 90% of the ion species in the extracted beams was found to be D^+ ion by means of mass spectrometry. The other species were D_2^+ and D_3^+. The monochromatic D^+ ion energy could be varied from 100 eV to 2 keV. The effective implantation area of the target is the central region of 25 mm in diameter. More details of this apparatus are described in a previous paper [14].

A 316L SS sheet of 0.05 mm thickness was cut into a disc of 34 mm diameter as a target of IDP experiment. The sample disc was clamped on the target flange with a thermocouple, fixed in the main chamber and heated up to about 670 K for more than several hours under vacuum for degassing. After degassing of the sample, D^+ ions with energy of 1 keV and a flux of over 1×10^{19} D^+/m^2 s were implanted onto the target at about 670 K for more than 20 h. After the above sample treatment, the series of implantation experiments were performed to determine the dependence of the permeation flux on the target temperature, incident ion flux and incident ion energy.

The permeated fluxes were measured by a quadrupole mass spectrometer ($M/e = 4$). During these experiments, the pressure of the upstream (incident) side in the main chamber was controlled at 4×10^{-4} Pa, though the base pressure was below 1×10^{-6} Pa. The down stream base pressure was also below 1×10^{-6} Pa at the room temperature.

3. Results and discussions

3.1. IDP behavior at steady state

The dependence of IDP flux at steady state on incident ion energy was measured under the conditions of incident ion energy ranging 200–1000 eV, incident ion flux of 5.7×10^{18} D^+/m^2 s and the temperature of 588 K. Fig. 1 shows the result of the incident ion energy dependence of IDP. The IDP flux had a tendency to increase with increasing the incident ion energy, and it was saturated around 1000 eV.

The temperature dependence of IDP flux at steady state was measured under the temperature range of 476–625 K, the incident ion energy range of 400–1000 eV and the incident ion flux of 1.0×10^{19} D^+/m^2 s. Fig. 2 shows the experimental results of the temperature dependence of the IDP flux at several incident ion energies. The ratio of the IDP fluxes against incident ion flux was found to be about $1.5–3 \times 10^{-5}$ at 1000 eV. The activation energies of permeation process calculated from Fig. 2 are summarized in Table 1. The temperature dependence of IDP behavior was found to be classified into two categories, one is a region of above 700 V with small activation energy of IDP and another is a region of below 700 eV with activation energy of averaged about 24 kJ/mol of IDP from Fig. 2 and Table 1.

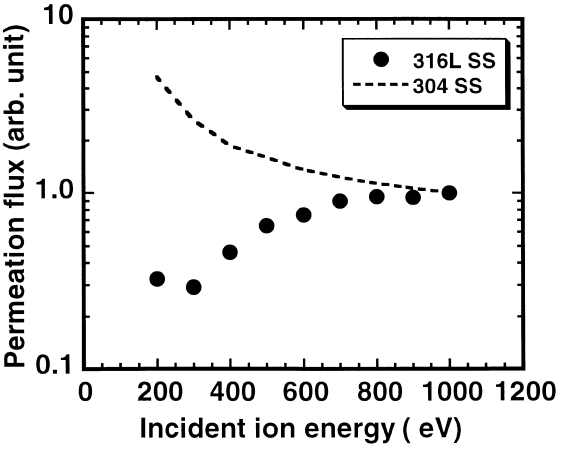

Fig. 1. Incident ion energy dependence of IDP of deuterium implanted into 316L SS at steady state. (Dashed line means IDP flux for 304 SS reported previously.)

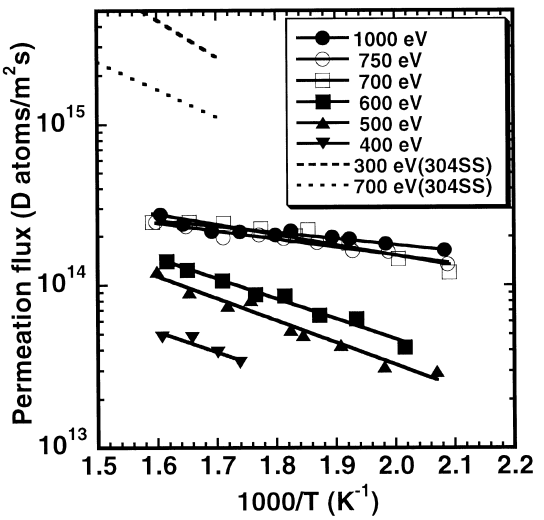

Fig. 2. Temperature dependence of IDP of deuterium implanted into 316L SS at steady state. (Dashed lines mean IDP flux for 304 SS which normalized to 316L SS conditions.)

Table 1
Activation energies of IDP of deuterium implanted into 316L SS

Ion energy (eV)	400	500	600	700	750	1000
E_P (kJ/mol)	23.9	25.8	23.6	12.2	9.5	7.2

Incident ion flux dependence of IDP at a steady state was measured to determine the n value defined from the $\phi_p = \alpha(\phi_i)^n$ relation, which is one of the key parameters

for identifying the control regime of IDP. Where, ϕ_p is a permeation flux, ϕ_i is an incident flux, and α is a constant. The relation was derived by Doyle and Brice [15,16], and Kerst et al. [6], assuming a steady state model.

The measurements were carried out under the conditions of 500, 750 and 1000 eV, the incident ion flux range of 2.5×10^{18}–1.1×10^{19} D^+/m^2 s and the temperature range of 493–633 K. Fig. 3 summarizes the n value derived from the incident ion flux dependence of IDP for each experimental condition. The figure shows the tendency that the n value increases with increase of temperature from 0.5 to nearly 1 at higher incident ion energies (750 and 1000 eV). On the other hand, the n value maintained almost 0.5 at the lower energy (500 eV) even at high temperature.

According to the steady state model, the IDP process of hydrogen isotopes implanted into metals can be classified into four categories depending on whether the rate-determining step is the recombination reaction on the surface or diffusion process for hydrogen isotopes, at the front or back side of the metal. These are (1) recombination reaction both side (RR regime), (2) the recombination at the front surface and diffusion at the back side (RD regime), (3) diffusion at both side (DD regime) and (4) diffusion at the front side and recombination at back side (DR regime). The n value is predicted to be 0.5 for RD regime, 1 for DD and RR regime, and 2 for DR regime from this model. The observed n value increased from almost 0.5 to nearly 1 with increasing temperature at higher energy, and maintained almost 0.5 at lower energy as mentioned above. Therefore, it was suggested that IDP behavior at the lower energy was controlled by RD regime and that at higher energy changed from RD regime to DD or RR regime with increasing the temperature.

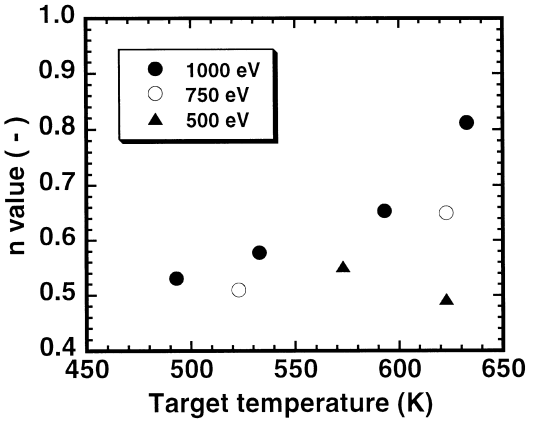

Fig. 3. Temperature dependence of n value derived from incident ion flux dependence of IDP for 316L SS.

Assuming IDP implanted by lower ion energy (below 700 eV) was controlled by RD regime, the activation energy of IDP could be estimated by the relation of $E_P = E_D - E_k/2$ from the above transport model, where, E_P, E_D and E_k mean activation energies of permeation, diffusion and surface recombination at front surface, respectively. The E_D and especially E_k for 316 SS are varied by the experimental conditions or surface conditions in previous reports, where, the reported activation energies are E_D: 52–54 kJ/mol [3,17], E_k: 58–96.4 kJ/mol [3,17]. Although it is difficult to compare the E_P observed in this experiment with the predicted value directly, it could be considered that the observed activation energy, which is about 24 kJ/mol at the low energy region in this experiment, is not far from the predicted activation energy of permeation assumed the IDP controlled by the RD model.

From this experimental result, the tendency of the n value to increase corresponded to the IDP flux increase in the higher incident ion energy region. According to Doyle's model, it would be reasonable to consider that the control regime of IDP at high temperature changed from RD regime to RR regime, because IDP would be limited by recombination at back surface with increasing the IDP flux. The experimental result that observed small activation energy of IDP at higher incident ion energy above 700 eV as shown in Fig. 2 and Table 1 also supports this assumption.

There are, however, some difficulties on understanding the incident ion energy dependence of IDP, because the RD and RR regime should be independent to incident ion energy according to Doyle's model. It is difficult to explain the observed incident ion energy dependence of IDP except considering implantation effect such as irradiation damages etc. Concerning this implantation effect, Tanabe et al. [2] and Wilson et al. [5] have pointed out the possibility that the defects produced by hydrogen ion implantation could change the effective diffusion coefficient and/or recombination coefficient in the implanted region in their previous reports. It would be reasonable that the production of such defects has a incident ion energy dependence. Therefore, the observed incident ion energy dependence of IDP could be also explained by the results of incident ion energy dependence of production of the irradiation defects which could change effective diffusion and/or recombination coefficient at implanted region.

3.2. IDP behavior at transient state

In order to discuss the implantation effects on IDP behavior mentioned at 3.1 in another aspect, transient behavior of IDP was also studied. The change of IDP flux which was in a steady state at 1000 eV of incident ion energy was measured by changing the incident ion energy immediately to lower energy while keeping the

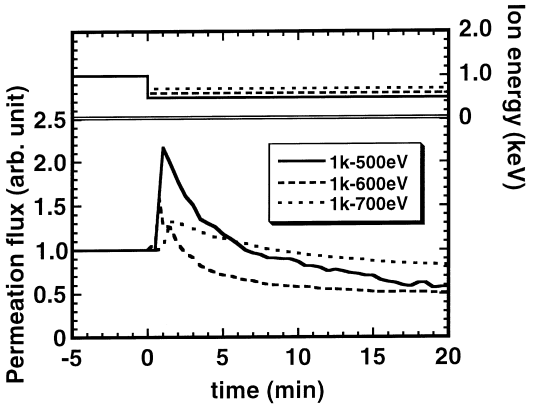

Fig. 4. The examples of the transient behavior of IDP by changing the ion energy from 1000 eV to 500, 600 and 700 eV, respectively.

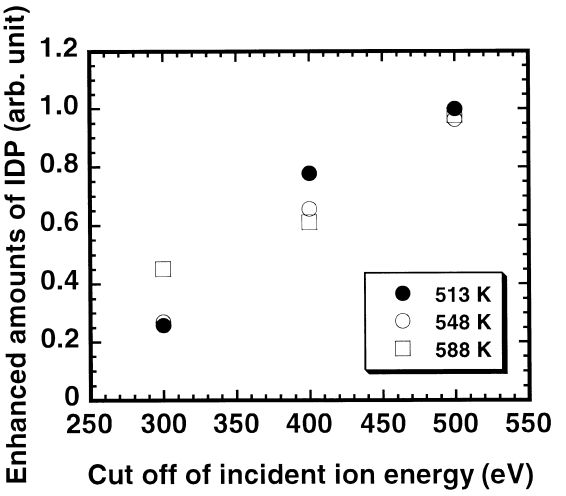

Fig. 5. Enhanced amounts of IDP by changing incident ion energy against to the cut off of incident ion energy calculated from Fig. 4.

incident ion flux constant. Fig. 4 shows the examples of transient behavior of IDP under the conditions of incident ion energy changed from 1000 eV to 500, 600 and 700 eV, respectively, at the temperature of 588 K and incident ion flux of 1×10^{18} D^+/m^2 s. It can be seen in Fig. 4 that the transient curves of IDP showed peaks looked like "permeation spike", which was observed by Perkins and Noda [1], while the IDP showed a simple decay curve when implantation was shut off. It was observed that the peak area of transient curve was enhanced with increasing the difference of energy between 1000 eV and the energy changed to (defined as "cut off" hereafter) as shown in Fig. 4. Therefore, the transient behavior of IDP was analyzed by comparing the enhanced amounts of permeation, which was determined by the difference of observed IDP and a transient curve estimated from a decay curve of IDP, with "cut off". Fig. 5 summarizes the enhanced amounts of permeation as a function of the "cut off". The result indicated that the enhanced permeation amount depended on "cut off" of incident ion energy, i.e., difference of implantation depth of deuterium ions, and hardly on temperature as shown in Fig. 5. Therefore, the enhancement of permeation could be considered to be caused by implantation effects of deuterium ions.

Although this behavior is not so called "permeation spike" reported previously [1,2,5,7], it may be possible to explain the transient behavior by applying the two reasons, which have been proposed for explanation of "permeation spike". One is the surface impurities effect [1,7], and the other an irradiation effect [2,5]. Considering these explanations to apply to this case, it is difficult to apply the impurities effect because continuous ion implantation was carried out during a series of experiment. Assuming that deuterium ion implantation forms weak irradiation defects which interact with deuterium atoms, it would be reasonable that spike like IDP peak would be considered as a result of migration and permeation of mobile deuterium atoms generated by annealing irradiation defects in region where the implantation was discontinued by the result of implantation depth shortening.

3.3. Comparison of IDP behavior between 316L SS and 304 SS

The characteristic differences of IDP behavior between 316L SS obtained in this experiment and 304 SS reported in the previous paper [10] were observed in the incident ion energy dependence and in the magnitude of permeability of IDP as shown in Figs. 1 and 2, respectively. The other parameter dependencies (temperature and ion flux) for 304 SS were similar to those of this result. The IDP flux for 316L SS was found to be smaller than that for 304 SS by about one order of magnitude for 700 eV at 625 K at normalized to 316L SS condition. Additionally, the ratio of IDP flux between 304 SS and 316L SS was found to be growing larger with decreasing incident ion energy as shown in Fig. 2. This result revealed the advantage of 316L SS from the viewpoint of tritium permeation under low energy tritium ion implantation.

Concerning to the incident ion energy dependence of IDP, the IDP behavior of 316L SS for incident ion energy showed opposite behavior to that for 304 SS as shown in Fig. 1. There are not clear explanations for this, however, these results imply the existence of different types of defects caused by ion implantation which could enhance or prevent the diffusion and/or recombination at the implanted face for each material.

4. Conclusions

The IDP behavior of deuterium through 316L SS was investigated at steady and transient state. The IDP at steady state was found to be controlled by the RD regime at low temperature, and seemed to change to RR regime at higher energy with increasing temperature.

The result of incident ion energy dependence of IDP at steady state and transient behavior of IDP suggested the existence of irradiation damage which could interact with deuterium atoms and could change such as effective diffusion or recombination coefficient at implanted region.

As the result of comparison of IDP behavior between 316L SS and 304 SS, it was found 316L SS had the advantage from the viewpoint of tritium permeation under low energy tritium ion implantation.

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