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Study of the retention of hydrogen isotopes implanted in Mo

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Abstract

A thin foil of Mo was irradiated by ions of hydrogen isotopes including tritium using a tritium beam test apparatus at the University of Tokyo. Thermal desorption spectroscopy (TDS) technique was employed to evaluate the total amount of retained hydrogen isotopes in Mo. According to the results, deuterium retention increased monotonously as the irradiation fluence was increased. It was observed that thermal desorption of both hydrogen and deuterium occurred predominantly at low temperatures, below 400 K, and that the increase of desorption could be attributed to higher temperature peaks which became appreciable as the fluence was increased; but, the amount of hydrogen or deuterium desorption from these high temperature peaks were found to be very small. The tritium retention in Mo, on the other hand, was nearly the same as that found for Ni.

1. Introduction

The use of tritium is being envisaged in near-term fusion devices such as ITER. It is expected that the retention of hydrogen isotopes in plasma facing materials (PFM) will play an important role in determining tritium inventory as well as hydrogen recycling. The pioneering experience of DT experiments in JET [1] and TFTR [2], together with the laboratory-scale experiments [3–5], would certainly be beneficial to ITER. However, owing to radiation protection issues associated with usage of tritium, they are limited in number.

High Z materials such as Mo and W are considered as candidates for PFM because of their high melting points and low hydrogen solubility [6]. Although hydrogen retention data are available to date for various candidate PFM's, most of them are obtained for hydrogen (H) and deuterium (D). It is in urgent need that retention data for tritium should be obtained, especially from the standpoint of isotope effect.

In the present study, a thin foil of Mo was irradiated by hydrogen isotopes including tritium. The thermal desorption spectroscopy (TDS) technique was employed to evaluate the total amount of retained hydrogen isotopes in Mo. The obtained results are compared between the isotopes, as well as with previous results for Ni [7].

2. Experimental

Different types of hydrogen isotope ion beams namely, pure H, pure D and mixed D/T, were irradiated on to a thin foil of Mo at normal incident angle and at room temperature using the Tritium Beam Test System (TBTS) at the University of Tokyo [7]. The vacuum chambers of TBTS were pumped with a cryopump and/or sputter ion pump which were backed up by a turbomolecular pump and a rotary pump. Tritium gas was stored in a sorption pump from an ampule containing 630 MBq of tritium.

The sample employed in the present study was a 0.1 mm thick polycrystalline Mo foil (Nilaco, 99.95% in purity) and was cleansed in an ultrasonic bath with trichloroethylene. In the runs dealing with H and D, the sample was irradiated with 3.0 keV H_2^+ (D_2^+) at room temperature to a dose varying from 1.3×10^{19} to

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1.3×10^{21} ions m^{-2}. Two hot runs were performed using tritium, in which the gas supplied from the sorption pump was ionized in an ion gun through a variable leak valve and was irradiated to the sample with an incident energy of 3.0 keV and to a total incident fluence of 1.3×10^{20} ions m^{-2}. Although direct analysis of ion beam composition was not performed in the present study, according to the residual gas analysis by quadrupole mass spectrometer (QMS), the ions were considered to be mainly composed of hydrogen and deuterium ions, where the compositions of hydrogen isotopes were estimated to be H:D:T $\simeq$ 1:300:1 and 4:55:1.

In the thermal desorption spectroscopy (TDS) measurements, the temperature of the sample was increased to 1000 K at the rate of 1.6 K/s and the total amount of desorbed gas was measured by integrating the signal of QMS which had been calibrated beforehand. Since in these experiments only a limited amount of tritium was used, it was necessary to be able to supply a sufficient amount of tritium over the whole experimental run. For this purpose, the cryopump was activated after several TDS measurements, and the gas was stored in a sorption pump. Tritium gas was then supplied to the chamber from the sorption pump. It was confirmed through residual gas analysis that the peak signals of DT and T_2 did not decrease during ion beam irradiation.

3. Results and discussion

Table 1 summarizes the temperatures of desorption peaks and the desorbed amounts of gaseous species from Mo irradiated by hydrogen isotopes ions described in Section 2. The temperatures of the desorption peaks were determined by simply reading the peak temperatures in the raw spectra. The thermal desorption spectra of H_2 measured for the sample which was irradiated with H_2^+ ions to a dose varying from 1.3×10^{19} to 1.3×10^{21} ions m^{-2} are shown in Fig. 1. The figure shows that hydrogen retention increases as the irradiation fluence was increased, but tends to saturate at $>1.3 \times 10^{20}$ ions m^{-2}. It is also observed that the thermal desorption of hydrogen occurred predominantly at low temperatures, below 400 K, and that the increase of

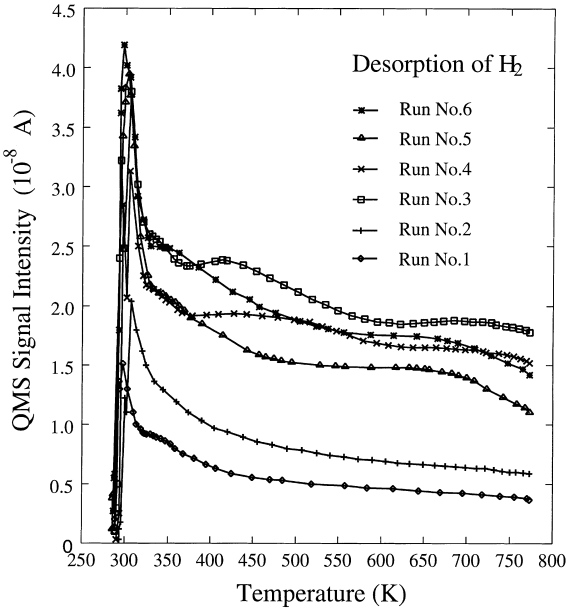

Fig. 1. Thermal desorption spectra of H_2 from Mo irradiated by hydrogen ions (H_2^+) to a fluence varying from 1.3×10^{19} to 1.3×10^{21} ions m^{-2}.

Table 1
Summary of TDS measurements on Mo

Run no.	Incident fluence (ions m^{-2})	Temperatures of desorption peak (K)	Amount of desorption (molecules m^{-2})
1	H_2^+: 1.3×10^{19}	300, 330	6.1×10^{18}
2	H_2^+: 6.5×10^{19}	305, 350	2.5×10^{19}
3	H_2^+: 1.3×10^{20}	305, 330, 410, 685	6.3×10^{19}
4	H_2^+: 3.9×10^{20}	305, 340, 460, 700	5.4×10^{19}
5	H_2^+: 6.5×10^{20}	305, 340, 600	5.1×10^{19}
6	H_2^+: 1.3×10^{21}	300, 350, 600	6.0×10^{19}
7	D_2^+: 6.5×10^{19}	310, 350	1.0×10^{19}
8	D_2^+: 1.3×10^{20}	300, 315	1.6×10^{19}
9	D_2^+: 3.9×10^{20}	310, 325, 390	2.2×10^{19}
10	D_2^+: 6.5×10^{20}	310, 390, 580	2.1×10^{19}
11	D_2^+: 1.3×10^{21}	295, 345, 590	3.9×10^{19}
12	1.3×10^{20}	DT: 315, 350, 550	1.3×10^{16}
	H:D:T = 1:300:1	T_2: 310, 360, 570	5.7×10^{15}
13	1.3×10^{20}	DT: 325, 350, 550	1.1×10^{16}
	H:D:T = 4:55:1	T_2: 310, 360, 570	5.6×10^{15}

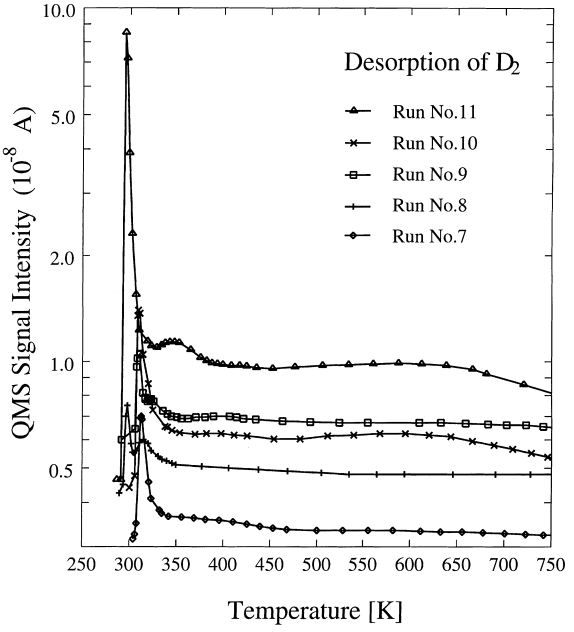

Fig. 2. Thermal desorption spectra of D_2 from Mo irradiated by deuterium ions (D_2^+) to a fluence varying from 6.5×10^{19} to 1.3×10^{21} ions m^{-2}.

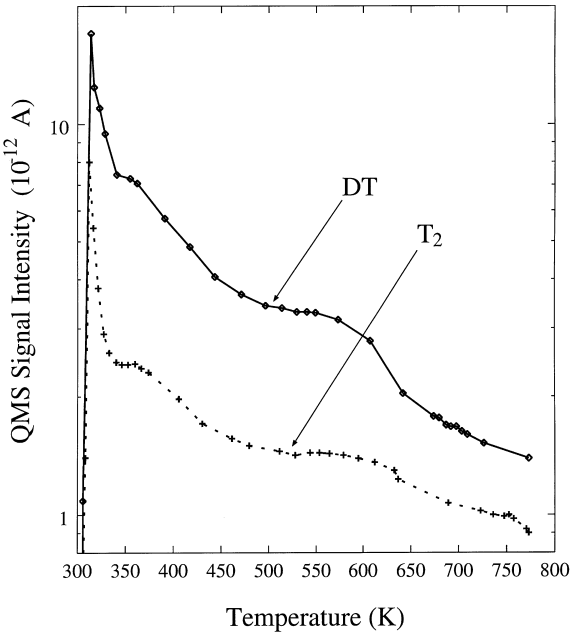

Fig. 3. Thermal desorption spectra of DT and T_2 from Mo irradiated by hydrogen isotope ions to a fluence of 1.3×10^{20} ions m^{-2}. The ratio of hydrogen isotopes during irradiation is estimated to be H:D:T = 1:300:1.

desorption could be attributed to higher temperature peaks which became appreciable as the fluence was increased, although the amount of hydrogen isotopes desorbed from these peaks was small. On the other hand, the thermal desorption spectra of D_2 from Mo are shown in Fig. 2. As in the case of hydrogen, a similar trend was observed for deuterium, but more clearly the deuterium retention increased monotonously with the irradiation fluence. It is further seen that the position of the low temperature peak was nearly independent of the incident fluence. This fact is considered to indicate that this peak is due to desorption of hydrogen which are dissolved in normal solution sites and/or adsorbed on the surface sites. The higher temperature peaks, on the other hand, were more sensitive to the irradiation fluence and so are considered to be due to hydrogen trapped at the irradiation damages which are produced by hydrogen bombardments. The peak at 600–700 K of hydrogen appeared at the fluence above 1.3×10^{20} H_2^+ m^{-2} (Run No. 3), where point defects are known to form clusters [8]. It can be considered that these defects provide additional sites for hydrogen retention in Mo.

In the tritium experiments three peaks of desorbed DT and T_2 were observed. An example of thermal desorption spectra is shown in Fig. 3. Although the incident fluence of tritium is about two orders of magnitude smaller than that of other isotopes, three peaks are already apparent at this low fluence. In the light of the above discussion, this implies that tritium in Mo is bound to the trap site produced by irradiation due to other isotopes (hydrogen and deuterium) ions. Also in the figure it is shown that the peak temperatures of T_2 were nearly the same or somewhat higher than those of DT, see Fig. 3 and Table 1.

The observed results are similar to those reported by Tanabe et al. [9], but the ion irradiations were performed using 25 keV D^+ ions as opposed to 3 keV D_2^+ ions in the present study. On the other hand, the experimental conditions and procedure of the present study are somewhat similar to those of Haasz and Davis [10], but only a single peak is observed in their study. Although the incident fluence is orders of magnitude smaller, low annealing temperature (≈1000 K) in the present study resulted in the high temperature trap site remaining unchanged. In fact, the retained amount of hydrogen isotopes appears to be larger than that reported by these authors. The dependence of the retained amount of various hydrogen isotopes on the incident fluence is shown in Fig. 4. The deuterium retention increased monotonously with fluence, similar to the results reported by Haasz and Davis [10]. On the other hand, it appears that the amount of hydrogen desorption is nearly constant in the fluence range between 1.3×10^{20} and 1.3×10^{21} H ions m^{-2}. However, since the retained amount in this fluence region is $\approx 10^{20}$ H m^{-2}, still below the saturation value of $\approx 10^{21}$ H m^{-2}, prolonged irradi-

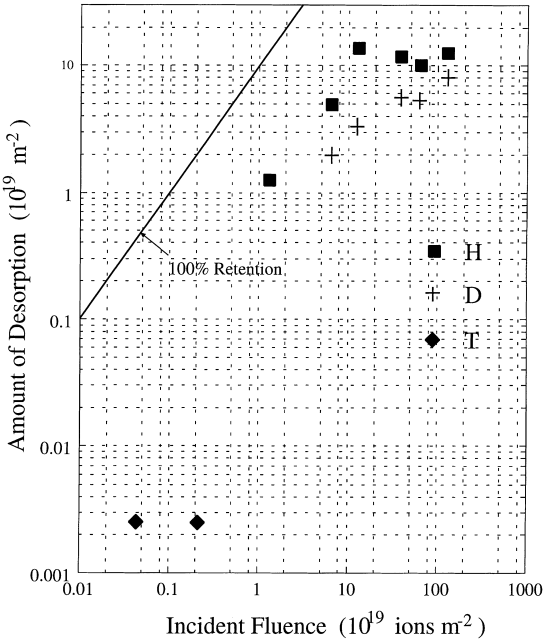

Fig. 4. Relation between retention of hydrogen isotopes in Mo and incident ion fluence. The incident fluence of tritium is estimated from residual gas analysis of the feed gas.

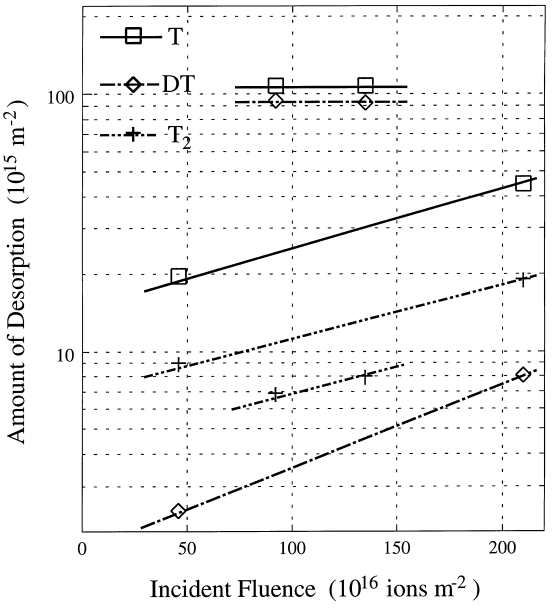

Fig. 5. Retention of tritium in Ni as a function of incident ion fluence. T denotes the total retention of tritium obtained by taking the sum of DT and T_2 retention.

ation to much higher fluence is necessary to check whether the retention has really achieved saturation. One of the reasons for saturation, if any, may be attributed to rapid diffusive release from Mo, resulting in the prompt loss of implanted hydrogen after the termination of the ion beam [9]. It is interesting to note that, according to Table 1, the temperature of desorption peaks at Run Nos. 3 and 4 are different from those at Run Nos. 5 and 6. Since the incident fluence in the former runs were smaller than that in the latter runs, the obtained results may indicate some fluence dependence of peak temperature which should reflect the behavior of hydrogen in Mo, the details of which are not clear at present.

It appears that in the fluence range investigated the retention of hydrogen is larger than that of deuterium. In addition, when the retention curve for hydrogen or deuterium is extrapolated to lower fluence tritium retention was smaller than that for other hydrogen isotopes. More data on tritium retention is required at higher fluence to further discuss the isotope dependence of the hydrogen retention in Mo. However, the use of tritium is limited to 2.0×10^{21} T m^{-2}, under the conditions of the present study.

The retained amount of tritium in Ni was evaluated using the same apparatus and is shown in Fig. 5 as a function of the incident fluence of tritium. The tritium retention in Ni is further complicated by the fact that two patterns of tritium desorption have been observed. As shown in the figure, the values for Mo are close to the lower values of Ni, although the retained amount of hydrogen or deuterium was generally smaller for Ni. The diffusive release of the retained hydrogen may also be possible in Ni [9], but it is not clear at present whether this has any effect on the obtained data.

4. Summary

The retention of various hydrogen isotopes including tritium in Mo has been measured by means of post-irradiation TDS technique. The deuterium retention increased monotonously as the irradiation fluence was increased. It was observed that the thermal desorption of both hydrogen and deuterium occurred predominantly at low temperature, below 400 K, and that the increase of desorption could be attributed to higher temperature peaks which became more appreciable as the fluence was increased. Similar dependence on the incident fluence as reported in the literature [10] was observed for deuterium desorption. The tritium retention was nearly the same for Mo and Ni at low fluence, although at higher fluence hydrogen or deuterium retention was larger for Mo. Data on tritium retention at higher fluence are needed to compare the obtained results with the existing retention data from the standpoint of isotope effect.

Acknowledgements

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