



Study on retention of hydrogen isotopes and helium in nickel using tritium beam test apparatus

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Abstract

In fusion reactors, a membrane of some metals, such as nickel, may be employed to enhance the pumping efficiency for helium ash because of its capacity to retain helium. In the present study, retention behavior of helium and hydrogen isotopes including tritium in nickel was investigated by means of thermal desorption spectroscopy (TDS), using a new beam irradiation system which is capable of handling tritium. In the thermal desorption spectra of helium (He), at least two helium desorption peaks were observed at 490–555 K and 715–740 K, where hydrogen isotope irradiations after helium irradiations were found to enhance the helium release and shift the desorption peak temperature to lower temperatures. However, the peak temperatures of helium desorption in helium irradiations after hydrogen isotope irradiation showed little difference from that without hydrogen isotopes irradiations. In the tritium experiments, tritium was supplied either directly from an ampoule or from a sorption pump to the vacuum chamber pumped by a cryopump. An ion beam of hydrogen isotopes containing tritium was irradiated to nickel at room temperature and the TDS spectra for DT and T₂ were obtained. It was revealed that both DT and T₂ have at least two desorption peaks, appearing at 410–425 K and 600–650 K for the former and at 500–520 K and 650–670 K for the latter. © 1997 Elsevier Science B.V.

1. Introduction

The behavior of hydrogen isotopes and helium in the plasma facing materials is one of the important factors that determine the performance of fuel plasma. Helium ash produced by fusion reaction makes it difficult to sustain the ignition condition owing to fuel dilution and enhancement of radiation loss. It is regarded that the helium concentration in the core plasma has to be within approximately 10% [1]. Additional pumping is necessary when the pumping capability of the divertor is not sufficient. Nickel has been suggested as a candidate material to selectively pump He [2–4]. In the limiter of TEXTOR, an enhanced pumping of helium ash by nickel has been demonstrated [5–8].

To date, the retention behavior of hydrogen and helium in nickel has been well studied. However, less attention has been paid to their mutual effect or difference in the

retention behavior of each hydrogen isotope in the presence of helium; namely, the isotope effect. Furthermore, in the light of future DT fusion, very little data on tritium retention are available. In the present study, we investigated the retention of ion-implanted helium and hydrogen isotopes including tritium in nickel by means of thermal desorption spectroscopy.

2. Experimental

A schematic view of the experimental setup is shown in Fig. 1. The experimental device was designed for the purpose of dealing tritium in a compact system to minimize tritium inventory due to sorption at the internal surface of the device. The chamber was pumped by a cryopump (Ulvac, Cryo-U6) and a sputter ion pump (Anelva, 913-7000) in the experiments dealing with only protium and deuterium (base pressure; 2×10^{-6} Pa). However, in the tritium experiments the chamber was pumped only by cryopump for fear that tritium pumped by a sputter ion pump may not be recoverable.

The sample employed in the present study was a 0.1

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mm thick polycrystalline nickel foil (99.7% in purity) and was cleaned in an ultrasonic bath with trichloroethylene. The sample was irradiated with 3 keV He^+ or H_2^+ (D_2^+) to a dose varying from 1.2×10^{20} ions m^{-2} to 6.0×10^{20} ions m^{-2} . In one experimental run, the procedure of irradiation by He^+ and H_2^+ (D_2^+) was reversed. The sample was irradiated at the pressure of $2.0\text{--}6.0 \times 10^{-3}$ Pa.

In the experiments dealing with tritium, tritium was first supplied directly from an ampoule containing 630 MBq of T_2 gas. The total fluence of ions irradiated to the sample was 3.5×10^{20} ions m^{-2} . According to the residual gas analysis by a quadrupole mass spectrometer (QMS), the ions were considered to be mainly composed of deuterium ions and the total amount of irradiated tritium was evaluated to be 1.7×10^{16} T m^{-2} (5.4 kBq). Following the above irradiation, the gas containing tritium was recovered by a sorption pump (SAES Getters, GP-50W2F) upon activation of the cryopump. The gas transported to the sorption pump was then introduced to the chamber for the succeeding irradiations.

In the thermal desorption spectroscopy (TDS) measurements, the total amount of desorbed gases was measured with QMS which had been calibrated beforehand. The sample temperature was increased from room temperature to as high as 1073 K at the ramp rate of 3 K s^{-1} . In the

experiments dealing with tritium, the sample temperature was increased to 823 K at the rate of $0.2\text{--}1.2 \text{ K s}^{-1}$.

3. Results

3.1. Co-irradiation by helium and hydrogen isotope ions

Table 1 summarizes the irradiation procedure and the peak temperature of helium desorption in each experiment. The precision of temperature measurement using a W-5%Re/W-26%Re thermocouple was $\pm 5 \text{ K}$. The thermal desorption spectrum measured for the sample which was irradiated with He^+ to a dose 1.2×10^{20} m^{-2} is shown in Fig. 2, corresponding to run No. 1. The total amount of desorbed helium was about 1.2×10^{19} He m^{-2} and helium desorption peaks were observed around 530 and 740 K. Although a peak is also seen around 400 K in Fig. 2, it was not always observable, so that it is not taken into consideration in the following discussion. This peak is probably due to helium adsorbed from the gas phase.

The nickel sample was irradiated with He^+ (1.2×10^{20} He^+ m^{-2}), then followed by H_2^+ irradiation (6.0×10^{20} H_2^+ m^{-2}) in run No. 2. The total amount of helium desorption in run No. 2 was 3.9×10^{19} He m^{-2} and the temperature of helium desorption peaks were around 525 and 720

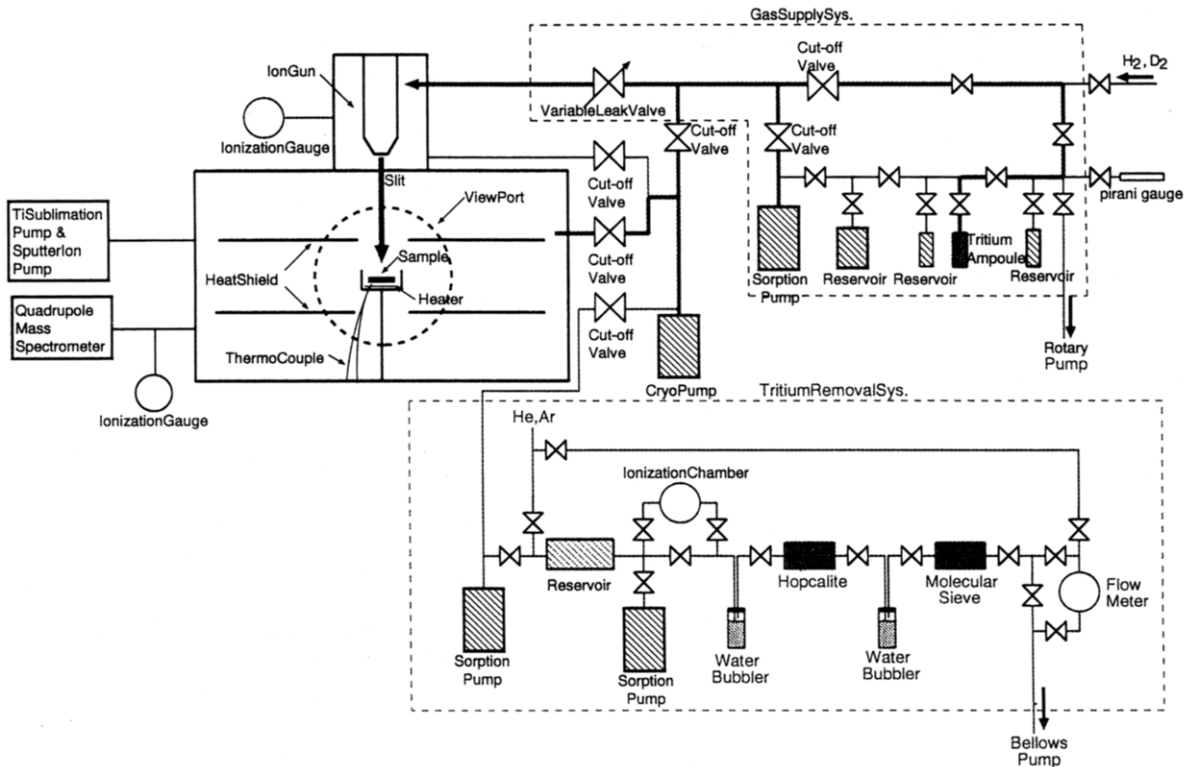


Fig. 1. Schematic diagram of tritium beam test system (TBTs).

Table 1

Summary of irradiation procedure and the corresponding results of thermal desorption experiments for He

No.	Irradiation conditions (m^{-2})	Temperature of desorption peak (K)	Temperature of desorption peak (K)	Amount of He desorption (m^{-2})
1	$\text{He}^+ 1.2 \times 10^{20}$	530	740	1.2×10^{19}
2	$\text{He}^+ 1.2 \times 10^{20} \rightarrow \text{H}_2^+ 6.0 \times 10^{20}$	525 ($\Delta 5 \searrow$)	720 ($\Delta 20 \searrow$)	3.9×10^{19}
3	$\text{He}^+ 1.2 \times 10^{20} \rightarrow \text{D}_2^+ 6.0 \times 10^{20}$	500 ($\Delta 30 \searrow$)	720 ($\Delta 20 \searrow$)	–
4	$\text{He}^+ 6.0 \times 10^{20} \rightarrow \text{H}_2^+ 6.0 \times 10^{20}$	440 ($\Delta 90 \searrow$)	720 ($\Delta 20 \searrow$)	8.8×10^{19}
5	$\text{H}_2^+ 6.0 \times 10^{20} \rightarrow \text{He}^+ 6.0 \times 10^{20}$	520–550	735 (\rightarrow)	2.4×10^{19}

K. In run No. 3, the hydrogen isotopes irradiated to the sample was changed from hydrogen ions to deuterium ions but to the same dose as in run No. 2. However, the amount of helium desorption in run No. 3 was not measured because it is difficult to distinguish helium from deuterium with QMS. The temperature of helium desorption peaks in run No. 3 were around 500 and 720 K.

Fig. 3 presents the thermal desorption spectra of helium and hydrogen desorbed from nickel in run No. 4. The irradiation dose of helium ions in run No. 4 was increased by a factor of five as compared with that in run No. 2. The total amount of desorbed helium was about 8.8×10^{19} He m^{-2} and the peak temperatures of helium desorption were around 490 and 725 K in run No. 4. The hydrogen irradiation after helium irradiation shifted the desorption peaks to a lower temperature. On the other hand, the sample was irradiated with He^+ after H_2^+ irradiation in

run No. 5, where the doses of He^+ and H_2^+ irradiated to the sample were equal to those in run No. 4. The total amount of desorbed helium in run No. 5 was 2.4×10^{20} He m^{-2} and its desorption peaks were around 520–550 K and 735 K. The shift of helium desorption peaks was not observed in this experimental run as compared with those of run No. 1. However, an increase of helium desorption was observed as compared with the case of the helium irradiation without hydrogen irradiation.

3.2. Tritium experiment

In the tritium experiments, the total ion fluence irradiated to the sample was 1.7×10^{20} ions m^{-2} . The ions were considered to be mainly composed of deuterium ions and tritium fluence irradiated to the sample was evaluated to be 1.7×10^{16} T m^{-2} (5.4 kBq). Fig. 4 shows the thermal desorption spectra of DT ($m/z = 5$) and T_2 (m/z

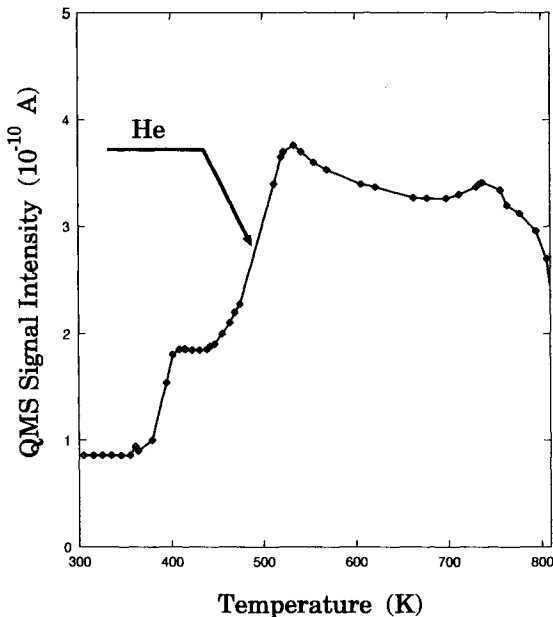


Fig. 2. Thermal desorption spectrum of He from Ni irradiated to 1.2×10^{20} $\text{He}^+ \text{m}^{-2}$, corresponding to run No. 1 in Table 1.

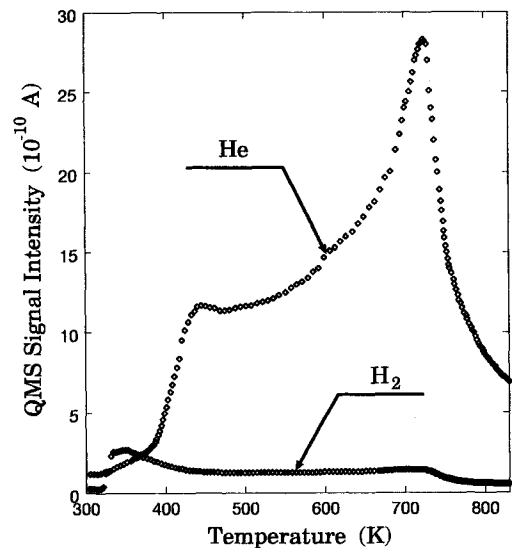


Fig. 3. Thermal desorption spectra of H_2 and He from Ni irradiated to 6.0×10^{20} $\text{He}^+ \text{m}^{-2}$ after irradiation to 6.0×10^{20} $\text{H}_2^+ \text{m}^{-2}$ (run No. 4).

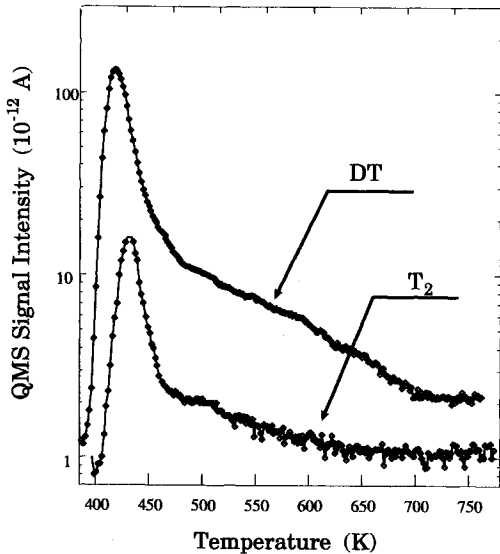


Fig. 4. Thermal desorption spectra of tritium-containing gas species from Ni irradiated by tritium beam.

= 6) for the sample irradiated by tritium containing gas which was directly supplied from the ampoule. The peak temperatures of desorption were observed around 435 K for DT and 440 K for T₂, although it seems that other desorption peaks may exist in the higher temperature portion of DT and T₂ spectra. The total amounts of DT and T₂ desorbed from nickel in this run were evaluated to be 2.0×10^{14} DT m⁻² (0.06 kBq) and 3.4×10^{13} T₂ m⁻² (0.02 kBq), respectively.

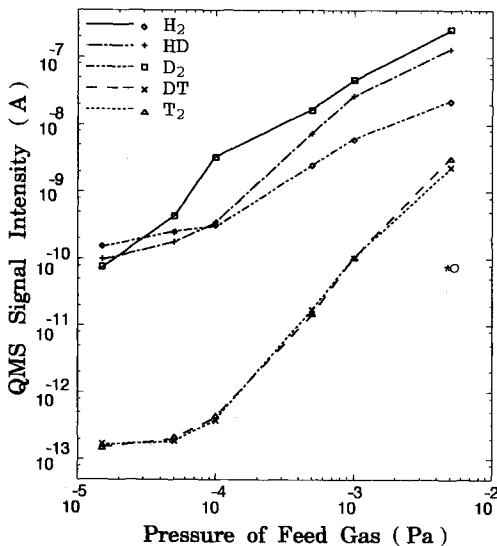


Fig. 5. The pressure dependence of hydrogen isotopes in the feed gas supplied by sorption pump. (*) and (O) are the signal intensities of DT and T₂ in a feed gas directly supplied from ampoule.

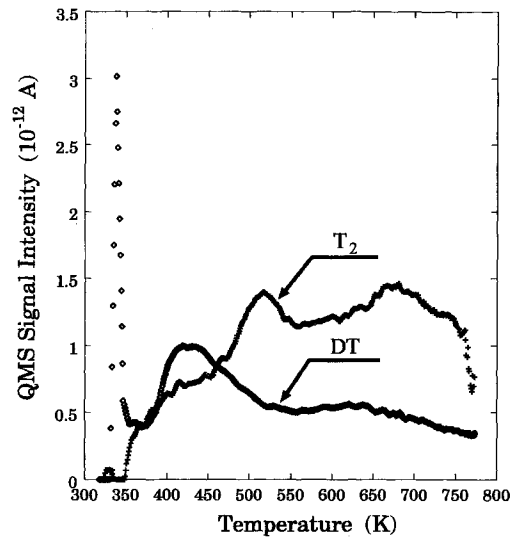


Fig. 6. Thermal desorption spectra of tritium-containing gas species Ni irradiated with tritium beam supplied from sorption pump and cryopump.

The cryopump was then activated and the gas desorbed from the pump was recovered by the sorption pump. The composition of the hydrogen isotopes in the gas from the sorption pump is shown Fig. 5. In the succeeding tritium experiments, the dose of ion beam irradiated to the nickel was 3.5×10^{20} ions m⁻². The fraction of H, D and T in the feed gas was measured by QMS. The thermal desorption spectra of DT and T₂ for the sample which was irradiated with hydrogen isotope gas of H:D:T ≈ 150:135:1 is shown in Fig. 6. The peak temperatures of desorbed DT were around 425 and 600–650 K. The peak temperatures of desorbed T₂ were around 515 and 670 K. The amounts of DT and T₂ desorbed from nickel were 2.5×10^{15} DT m⁻² (0.79 kBq) and 4.5×10^{15} T₂ m⁻² (2.9 kBq), respectively. In the experiments using hydrogen isotope gas of H:D:T ≈ 6:55:1, the peak temperatures of desorbed DT were around 410 and 600–650 K, while those of desorbed T₂ were around 505 and 650 K. The amounts of DT and T₂ desorbed from nickel were 8.4×10^{15} DT m⁻² (2.7 kBq) and 9.5×10^{15} T₂ m⁻² (6.0 kBq), respectively.

4. Discussion

4.1. Effect of co-irradiation by helium and hydrogen isotope ions

Helium irradiation to nickel under the conditions of the present study is likely to produce helium bubbles. The maximum density of helium inside a bubble is considered to be 2×10^{29} atoms m⁻³ [9,10]. In many studies [11–13], many small spherical bubbles in the range of 1–2 nm were

found in microstructure observed with transmission electron microscopy (TEM). Another study [14] shows that the pressure inside the bubbles was estimated to be about 3.5×10^{10} Pa and that the effective binding energy of hydrogen trapped near or around helium bubbles is in the range of 0.7–0.9 eV (atom)⁻¹. Unfortunately, in the present study, no observation on the microstructure evolution due to ion irradiation was performed, so that it is not possible to correlate each He desorption peak with irradiation effects. Nevertheless, the two peaks discussed in the previous section are quite contrasting. The results clearly showed that the lower temperature peak was quite susceptible to interaction with hydrogen isotopes, whereas such was not the case with the higher temperature peak. One possible explanation is that helium competes with hydrogen for same bulk sites in nickel, desorption from which the lower temperature peak is formed, whereas the higher temperature peak may be due to helium-related defect which is stable under the influence of hydrogen isotopes.

It is reported in the literature that no helium desorption was observed in nickel for helium doses less than 2.0×10^{21} He m⁻² with 40 keV [15]. However, it can be seen from the results obtained in the present study that helium desorption is enhanced due to co-irradiation in the presence of hydrogen. Of the two peaks observed in helium desorption, the lower ones, in particular, shifted to lower temperature in run Nos. 2–4 compared with He⁺ irradiation without hydrogen isotope irradiation (run No. 1). These results indicate that hydrogen atoms interact with the helium atoms in such a way as to decrease the effective binding energies of helium bubbles in nickel, thereby leading to reduction of apparent activation energy of desorption. It should be noted that in run Nos. 2 and 3, shift of helium desorption peak to much lower temperature was observed in run No. 3 (post-irradiation by deuterium) than in run No. 2 (post-irradiation by hydrogen); isotope effect was observed. It should be reasonable to assume that heavier isotope is more effective in interacting with helium in the solid.

Table 1 shows that the position of the lower temperature peak of helium in run No. 5 showed little difference from that in run No. 1. This seems to indicate that pre-irradiation by hydrogen has little effect on the succeeding helium irradiation. In other words, helium is more likely to be bound to nickel than hydrogen isotopes. This can also be seen in Fig. 3 that hydrogen desorption starts at much lower temperature than helium and that the retained fraction of hydrogen was generally less than several %, about an order of magnitude smaller than that of helium.

4.2. Behavior of tritium in nickel

630 MBq of tritium amounts to only 2.0×10^{21} T m⁻², under the conditions of the present study, so that the gas of other hydrogen isotopes, namely deuterium, had to be mixed to sustain ion beam to a sufficient flux density

level. Although the tritium beam irradiations were performed several times, the fraction of hydrogen isotopes varied to an extent described in Section 3.2.

The thermal desorption spectra of DT may resemble those of DT and T₂ in Fig. 4 and H₂ in Fig. 3. However, on some occasions, T₂ showed unique spectra such as shown in Fig. 6. Furthermore, it is shown in the figure that the amount of desorption is larger than that of DT. Considering the mass balance of hydrogen isotopes in the experimental system, such a phenomenon seems to be inexplicable. Additional measurement of tritium may be required to check the measurement made by QMS.

5. Conclusions

While the peak temperatures of helium desorption in He⁺ irradiation after H₂⁺ (D₂⁺) irradiation showed little difference from that without H₂⁺ (D₂⁺) irradiation, those observed in the He⁺ irradiation followed by H₂⁺ (D₂⁺) irradiation were shifted to lower temperature compared with He⁺ irradiation. The shift of the helium desorption peak to the lower temperature was greater in the case of deuterium post-irradiation than in the case of hydrogen post-irradiation.

It was demonstrated that tritium used in irradiation experiments can be recovered and supplied again for continuous operation of tritium irradiation by using sorption pump and cryopump. In the preliminary irradiation experiments using tritium, at least two desorption peaks appeared in thermal desorption spectra of DT and T₂, although the detailed analysis on the mass balance of tritium remained to be done.

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