9.4 Deuterium Retention Properties of $Be_{12}Ti$

H. Iwakiri[1], N. Yoshida[1], M. Uchida[2] and H. Kawamura[2]

[1]Research Institute for Applied Mechanics, Kyushu University
6－1 Kasugakoen, Kasuga, Fukuoka 816－8580, Japan
[2]Japan Atomic Energy Research Institute,

Abstract

Thermal desorption of deuterium from its ion irradiated $Be_{12}Ti$ and microstructural change during irradiation were examined to understand deuterium retention and desorption property. Total retention of deuterium in $Be_{12}Ti$ is much smaller than that of beryllium at the wide temperature range. For implantation of $2x10^{21}D/m^2$ at room temperature, 10% of implanted deuterium is retained and most of them are desorbed around 400K, where a high-density of cavities were formed. For the case of beryllium, 83% of implanted deuterium is retained. These results indicate that deuterium trapping efficiency of cavity in the $Be_{12}Ti$ is much lower than that of beryllium.

1. Introduction

For the neutron multiplier, beryllium metal (Be) is a reference material in the blanket design. However, it may not be applicable to the DEMO blanket, which requires high temperature and neutron dose, because of high reactivity and large swelling. Therefore, it is necessary to develop an advanced material for a neutron multiplier that has high temperature resistance and high radiation resistance. Berylides such as $Be_{12}Ti$ and $Be_{12}V$ have been suggested as promising candidates for advanced neutron multipliers from the view points of high melting point, high beryllium content, fast decay of gamma dose rate and good chemical stability, etc[1].

The behaviour of hydrogen isotopes in the neutron multiplier will influence the safety of the reactor. Furthermore, the generated hydrogen isotope causes problems for the neutron multiplier with respect to tritium retention. In this study, therefore, thermal desorption experiments and microstructure observation in deuterium irradiated $Be_{12}Ti$ were performed complementary.

2. Experimental procedures

$Be_{12}Ti$ specimens used in the present work were fabricated with hot isostatic pressing (HIP) process by NGK Insulators, Ltd [2]. Irradiations of 8keV-D_2^+ ions were carried out in an ultra-high vacuum evacuation apparatus equipped with a small duo-plasmatron type ion gun. After irradiation, the specimens were transferred into the TDS apparatus, where the thermally desorbed deuterium gas was measured with a high resolution quadruple

mass spectrometer. The desorption rate of deuterium was quantitatively calibrated by comparing with a data of He standard leak with correction of the relative ionization efficiency. The temperature of the specimens was increased up to 1700K with a ramping rate of 1K/sec.

Pre-thinned samples for transmission electron microscope observation were obtained by twin-jet electro-polishing. The in-situ observation under D_2^+ ion irradiation was conducted using a 200kV- transmission electron microscope equipped with a low energy ion accelerator. Details of the facility were described in elsewhere [3].

3. Results

3.1 Retention and thermal desorption of deuterium

Fig.1 shows thermal desorption spectra of D from $Be_{12}Ti$ irradiated with 8keV-D_2^+ ions up to $6 \times 10^{21}D/m^2$ at room temperature. The desorption stage can be classified into two groups; a sharp stage at low temperature side (350K － 450 K) and broad stage at high temperature side (450K－800K). The former and the later desorption stages are named here the low temperature and high, respectively. The both desorption stage increase with increasing dose up to $2.0x10^{21}D/m^2$. However, both desorption stage is shown decrease tendency between $2 \times 10^{21}D/m^2$ and $6 \times 10^{21}D/m^2$.

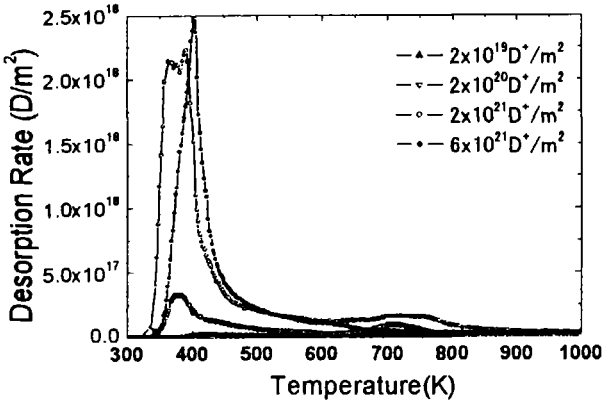

Fig. 1. Thermal desorption spectra of D from $Be_{12}Ti$ irradiated with 8keV-D_2^+ ions up to $3 \times 10^{21}D_2^+/m^2$ at room temperature.

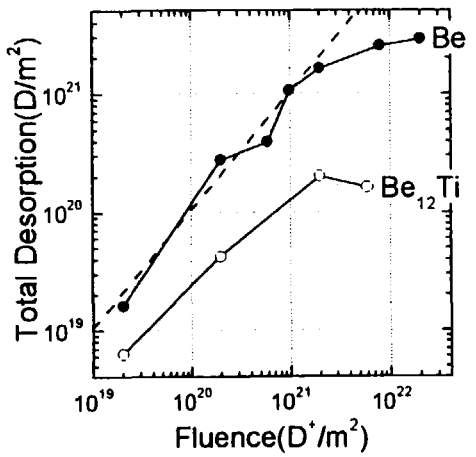

Fig. 2. Total amount of desorbed D from the $Be_{12}Ti$ and Be irradiated with 8keV-D_2^+ at room temperature.

Total amount of desorbed D from the $Be_{12}Ti$ and Be irradiated with 8keV-D_2^+ at room temperature is plotted in Fig. 2 as a function of ion dose. The straight dashed line shows 100% retention. It is clear that the trapping efficiency of deuterium in the $Be_{12}Ti$ is much lower than that of Be.

Fig. 3 shows thermal desorption

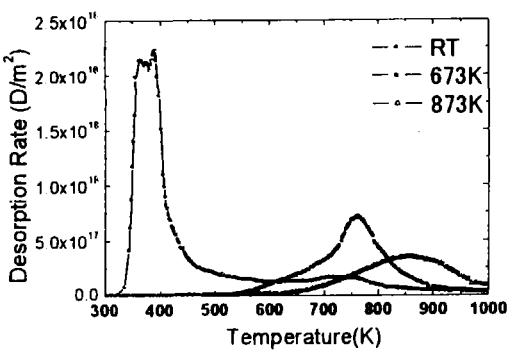

Fig. 3. Thermal desorption spectra of D form the $Be_{12}Ti$ irradiated with 8keV-D_2^+ at several temperatures up to 873K.

spectra of D form the $Be_{12}Ti$ irradiated with 8keV-D_2^+ at several temperatures up to 873K. The total amount of desorption decrease with increasing temperature. In the case of deuterium irradiation at 673K, the desorption at the low temperature stage is diminished, but the desorption at the high temperature stage increases and majority of the trapped deuterium is desorbed between 600K and 900K. With increasing irradiation temperature, the high temperature stage shifts toward the higher temperature side.

3.2 Microstructure evolution during deuterium ion irradiation

Fig. 4 shows microstructural evolution at room temperature under irradiation with 8keV-D_2^+ ions. Any defect was not observed at low dose less than 2×10^{20}D/m^2. Fine cavities of 2－3nm in size were observed densely at 6×10^{20}D/m^2. With increasing dose, the cavity size grows gradually at first and grows further by coalescing each other. An interstitial-type defect, e. g., an interstitial loop, was scarcely observed at any fluence. These facts may show that the nucleation of interstitial loops is very difficult in $Be_{12}Ti$.

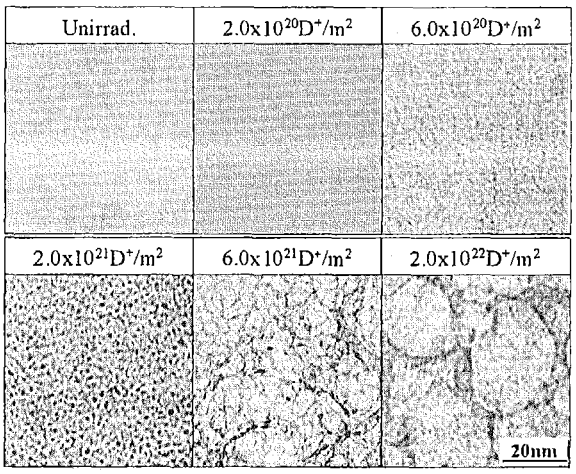

Fig. 4. Microstructural evolution of $Be_{12}Ti$ at room temperature under irradiation with 8keV-D_2^+ ions.

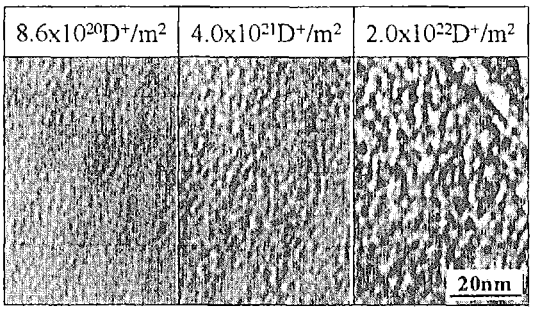

Fig. 5. Microstructural evolution of Be at room temperature under irradiation with 8keV-D_2^+ ions.

Fig.5 shows transmission-electron micrograph of Be irradiated at room temperature by D_2 ions at 8keV with doses of (a) 8.6×10^{20}D/m^2, (b) 4.0×10^{21}D/m^2, (c) 2.0×10^{22}D/m^2. It differs from $Be_{12}Ti$, the cavity size does not grow more than several nm even at very high dose.

Fig.6 shows isochronal annealing (100K-step, 30min) of microstructure formed by the irradiation at the dose of 2×10^{21}D/m^2. No remarkable change occurred up to 473K, and they start to coalesce between 473K and 573K. Large cavities exceed 20nm by the annealing up to 573K－673K. The squarish cavities could be observed above 873K. Relatively small cavities (<10nm) disappeared between 873K and 1073K, and then larger cavities disappeared at 1273K.

4. Discussion

Fig. 7 shows thermal desorption spectra of D released from $Be_{12}Ti$ and Be irradiated to 2×10^{21}D/m^2 at room temperature and 673K. In the case of $Be_{12}Ti$, 10% of implanted deuterium is retained and most of them are desorbed around 400K. For implantation at 673K, retention becomes smaller, about 5% for 2×10^{21}D/m^2, and the majority of deuterium is desorbed between 600K and 900K. In the case of Be, however, deuterium retention is much

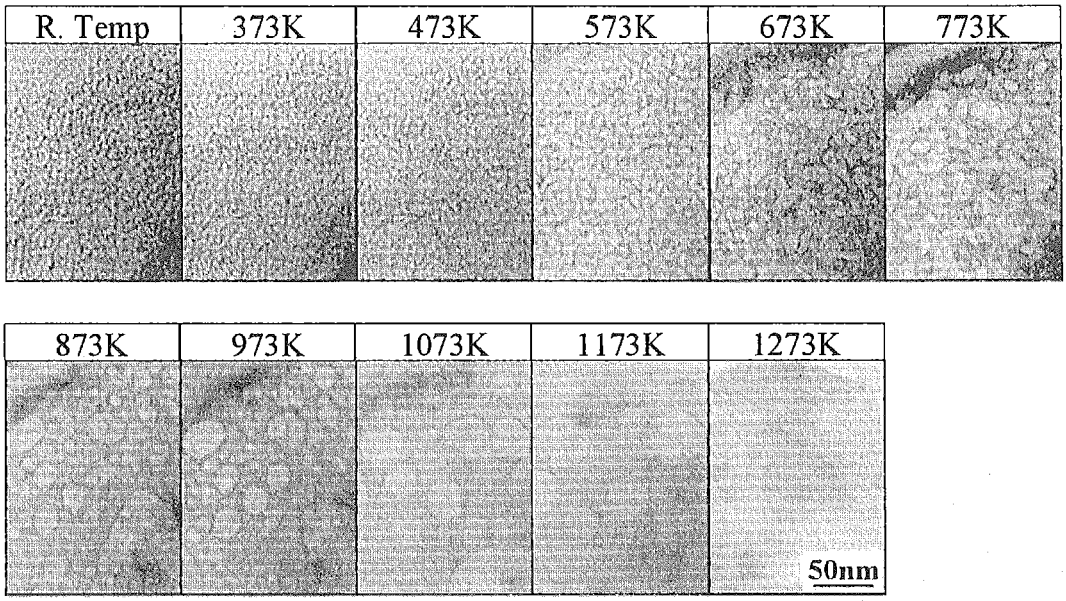

Fig. 6. Isochronal annealing (100K-step, 30min) of microstructure formed by the irradiation at the dose of $1 \times 10^{21} D_2^+/m^2$.

larger than that of $Be_{12}Ti$; 83% of implanted deuterium is retained at room temperature and 66% of implanted deuterium is retained at 673K. Some authors also clarified that two large desorption stage were formed in Be under D-ion irradiation [4,5,6,7], and they suggest the higher temperature side is related to cavities filled in the D_2 molecules. By using SIMS and RGA (residual gas analysis), the existence of D_2 molecules in irradiated Be was confirmed by Almov et al. [8]. As well as Be, fine cavities were formed densely in $Be_{12}Ti$. By repeating migration and coalescing, as it was shown in Fig. 5, cavities disappear gradually from the surface by the annealing up to 473K－1273K. Such phenomenon may be related to broad desorption stage at high temperature side. However, partial amount of desorbed deuterium from the high temperature stage in $Be_{12}Ti$ is about 1/15 in comparison with the thing in the Be. These results indicate that deuterium trapping efficiency of cavity in the $Be_{12}Ti$ is very lower than that of Be. In other words, though highly pressurized D_2 cavity can be formed in the Be, the pressure of the cavity in the $Be_{12}Ti$ is very low.

On the other hand, there is no influential evidence on low temperature stage in the $Be_{12}Ti$. In the case of Be, multiple hypotheses have been advocated. Wilson et al. [9] interpreted that this stage corresponded to the detrapping from amorphous beryllium hydride which decomposed at 400K. Another interpretation was suggested by Zakharov et al. [10], deuterium atoms may be trapped at the chemisorption site on the walls of the D_2 cavities. However, these hypotheses can not give reasonable explanation in present case since the amount of desorbed deuterium from

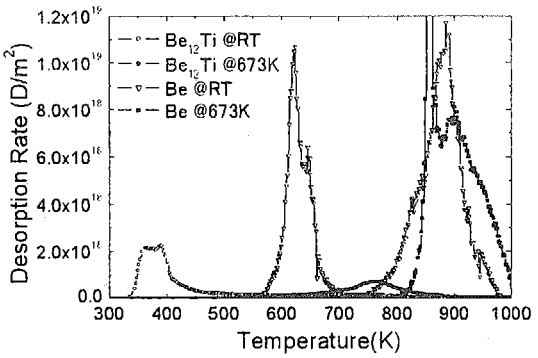

Fig. 7. Thermal desorption spectra of D released from $Be_{12}Ti$ and Be irradiated to $1 \times 10^{21} D_2^+/m^2$ at room temperature and 673K.

the low temperature stage in $Be_{12}Ti$ is much smaller than that of Be. We need more experiments to understand the details of the low temperature stage in $Be_{12}Ti$. As one possibility, comparably week trapping site at vacancy is guessed like endothermic hydrogen occluding metal [11] such as Mo, Ni and W.

These results indicated that the deuterium retention properties of $Be_{12}Ti$ for the neutron multiplier material are superior to that of Be. It is obvious that the tritium inventory from $Be_{12}Ti$ is much smaller than that for Be.

5. Conclusion

Thermal desorption deuterium from 8keV D_2^+ ions irradiated $Be_{12}Ti$ and microstructural change during irradiation and annealing were examined to understand deuterium retention and desorption of the implanted deuterium and to identify responsible traps. The results are summarized as follows.

(1) The deuterium desorption stage of $Be_{12}Ti$ can be classified into low temperature side and high temperature side at room temperature. In the case of deuterium irradiation at 673K, the desorption at the low temperature stage is diminished, but the desorption at the high temperature stage increases.

(2) Total retention of deuterium in $Be_{12}Ti$ is much smaller than that of Be at the wide temperature range. These results indicated that the deuterium retention properties of $Be_{12}Ti$ for the neutron multiplier material are superior to that of Be. It is obvious that the tritium inventory from $Be_{12}Ti$ is much smaller than that for Be.

(3) As well as the Be, fine cavities were also formed densely. With increasing dose, however, the cavity size grows gradually at first and grows further by coalescing each other unlike the Be.

(4) Possible trapping site of high temperature stage is cavities as well as Be. However, deuterium trapping efficiency of cavity in the $Be_{12}Ti$ is much lower than that of Be.

References

[1] M. Uchida, E. Ishitsuka, H. kawamura, Fusion Engineering and Design 69 (2003) 499
[2] H. Kawamura, M. Uchida, V. Shestakov, J. Nucl. Mater. 307-311 (2002) 638
[3] T. Muroga, R. Sakamoto, M. Fukai, N. Yoshida, T. Tukamoto, J. Nucl. Mater., 196-198 (1992) 1013
[4] W. R. Wampler, J. Nucl. Mater. 122-123 (1984) 1598
[5] R. A. Langley, J. Nucl. Mater. 122-123 (1979) 1123
[6] N. Yoshida, S. Mizusawa, R. Sakamoto, T. Muroga, Fusion Technology 30 (1996) 798
[7] A. P. Zakharov, A. E. Gorodetsky, V. Kh. Alimov, S. L. Kanashenko, A.V. Markin, J. Nucl. Mater. 241-243 (1997) 52
[8] V. kh. Alimov, V. N. Chernikov, A.P. Zakharov, J. Nucl. Mater. 241-243 (1997) 1047
[9] K. L. Wilson, R. A. Causey, W. L. Hsu, B. E. Mills, M. F. Smith, J. B. Whitley, J. Vac. Sci. Technol. A 8 (1990) 1750
[10] A. P. Zakharov, A. E. Gorodetsky, V. Kh. Alimov, S. L. Kanashenko, A.V. Markin, J. Nucl. Mater. 241-243 (1997) 52
[11] T. Kiriyama, T Tanabe, J. Nucl. Mater. 220-222 (1995) 873