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Modification of tungsten coated carbon by low energy and high flux deuterium irradiation

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Abstract

Plasma sprayed tungsten coated carbon fiber composites (VPS-W/CX-2002U) and powder metallurgy tungsten (PM-W) have been exposed to a high deuterium flux ($\simeq 10^{22}$ m^{-2} s^{-1}) with low energy (100 eV) in a range from 708 to 843 K. Surface modification and deuterium retention after the exposure have been investigated to prove the suitability of such materials in fusion devices. Blisters are formed on the PM-W by deuterium irradiation with a fluence of 7.5×10^{25} m^{-2}. The amount of blisters and their average size increase with an increase of a fluence to 3.00×10^{26} m^{-2}. On the other hand, no modification is observed on VPS-W/CX-2002U. Desorption of VPS-W/CX-2002U irradiated by deuterium is different from that of PM-W. The peak temperature of D_2 and HD release from PM-W is about 703 K. However, the desorption curve of VPS-W/CX-2002U gradually increases with increasing temperature up to 1273 K.

1. Introduction

Some of the disadvantages of tungsten as a plasma facing material are related to its heavy weight and poor workability. One way to circumvent these disadvantages is to deposit tungsten on carbon materials that have shown good heat shock resistance in the present plasma confinement devices. Tungsten coatings on graphite, by means of plasma spray or physical vapor deposition (PVD), have been produced and their performance under high heat flux loading has been examined [1,2]. From the viewpoints of thermal conductivity and mechanical strength, it seems that carbon/carbon fiber composites (CFC) are preferable as a substrate material for high heat flux loading. Thick tungsten coatings on CFC and isotropic fine grained graphite have successfully been produced by vacuum plasma spray (VPS) technique and their good thermal and adhesion properties has been confirmed by high heat flux tests [3–5].

Plasma facing materials of fusion reactors are subject to a high hydrogen isotope particle and heat flux. It is well known that modification on tungsten due to hydrogen isotope irradiation depends on the fabrication process. In the present study, plasma sprayed tungsten coated CFC and powder metallurgy tungsten (PM-W) were exposed to a high deuterium flux with a low energy relevant to the boundary plasma of fusion devices. Surface modification and deuterium retention property after the exposure have been investigated to prove the suitability of such materials in fusion devices.

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2. Experimental

The carbon/carbon fiber composite CX-2002U made by Toyo Tanso was coated using VPS [3,4]. The CX-2002U received PVD multilayer diffusion barrier layers of rhenium and tungsten prior to the VPS tungsten (VPS-W) coating in order to inhibit uncontrolled brittle carbide formation. Heat treatments were performed to stabilize the microstructure of the sample. The thickness of the tungsten coating layers was 0.5 and 1.0 mm. The density of VPS-W was 92.5% of theoretical density. Details of the fabrication method were described in the previous papers [3,4]. The sizes of VPS-W/CX-2002Us were 20 mm length, 14.5 mm width and 2.0 mm thickness. Pure tungsten fabricated by powder metallurgy (PM-W) supplied by Tokyo Tungsten Co. Ltd., was also used and compared with the results of VPS-W/CX-2002U. The purity of PM-W was 99.99 wt%. A disk of PM-W has a size of 24.5 mm diameter and 2 mm thickness. The surface of the PM-W was electrically polished. All samples were degassed by heating up to 1273 K in high vacuum before installation in a plasma facility.

The facility used in the experiments is the PISCES-B which is a liner plasma simulator device at the University of California, San Diego. Details of the PISCES-B were described in Ref. [6]. In the present experiments, deuterium is used as working gas. The ion flux (D^+) to the sample is varied from 4.79×10^{21} to 1.20×10^{22} $m^{-2}\,s^{-1}$ corresponding to the plasma exposures. The energy of the impinging ions is 100 eV. The plasma parameters remain constant throughout the plasma exposure. The sample is clamped to a water or air cooled sample holder with a tantalum cap. The temperature is measured at the center of the sample on the side opposite to the plasma with a thermocouple. The samples are heated by the plasma and reach their final steady-state temperatures within about 1 min. The sample temperatures were in a range from 708 to 843 K depending on the ion fluxes.

After the plasma exposures, the sample is removed from the PISCES-B facility and microstructure is studied by scanning electron microscopy (SEM). In addition, retention of deuterium after the exposure is examined by thermal desorption spectroscopy (TDS). The sample is linearly ramped up to 1273 K for 65 min (0.25 $K\,s^{-1}$) and remain at 1273 K for 20 min, and is cooled down linearly to 25 K for 90 min (0.18 $K\,s^{-1}$).

3. Results

3.1. Surface modification

Fig. 1 shows SEM images of the surface of PM-W before and after deuterium irradiation. No modification was observed for PM-W by deuterium irradiation to a fluence of 7.20×10^{24} m^{-2}. However, blisters with a size of 0.1 to a few μm were formed after deuterium irradiation with a fluence of 7.50×10^{25} m^{-2} as shown in Fig. 1(b). The amount of blisters and their average size increased with an increase of a fluence to 3.0×10^{26} m^{-2} as shown in Fig. 1(c). Fig. 2 shows SEM images of the surface of VPS-W/CX-2002U before and after deuterium irradiation. It can be seen that spherical particles (≃10 μm) were melted or partially melted, joined each other and accumulated on the surface. Before irradiation, very small fragments (≃0.1 μm) were observed on the surface of VPS-W/CX-2002U. However, they disappeared and no other changes such as blisters were observed after irradiation.

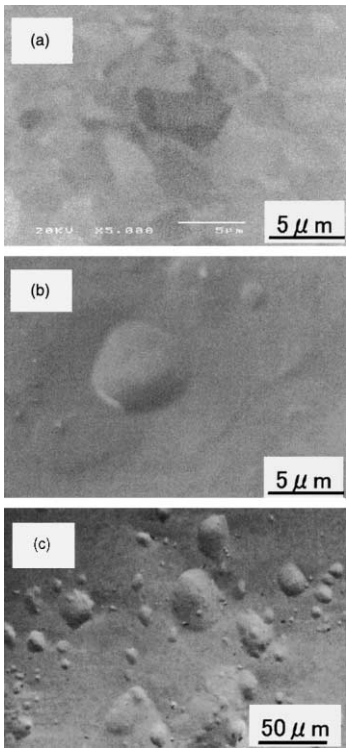

Fig. 1. SEM images before and after exposure to plasma: (a) PM-W before exposure, (b) PM-W after exposure to deuterium plasma (100 eV, 7.50×10^{25} $D\,m^{-2}$, 823 K), (c) PM-W after exposure to deuterium plasma (100 eV, 3.00×10^{26} $D\,m^{-2}$, 823 K).

3.2. Retention property

Fig. 3 shows the thermal desorption spectrum of HD and D_2 from PM-W(a) and VPS-W/CX-2002U(b) exposed to deuterium plasmas. The peak temperature of the D_2 and HD release from PM-W was about 703 K. In addition, a small desorption peak of HD at 873 K appeared. The total amount of D_2 was larger than that of

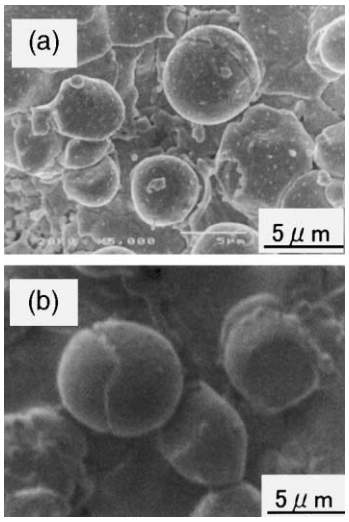

Fig. 2. SEM images before and after exposure to plasma: (a) VPS-W/CX-2002U before exposure, (b) VPS-W/CX-2002U after exposure to deuterium plasma (100 eV, 3.75×10^{25} D m^{-2}, 838 K). The thickness of VPS-W is 1.0 mm.

HD. On the other hand, the desorption curve of VPS-W/CX-2002U gradually increased with increasing temperature up to 1273 K and decreased at 1273 K for 20 min. On the contrary to PM-W, the total amount of HD released from VPS-W/CX-2002U was larger than that of D_2. Table 1 shows the total amount of desorption as a function of incident fluence. It can be seen that deuterium retention in VPS-W/CX-2002U was larger than that in PM-W.

4. Discussion

4.1. Blister formation

The maximum energy transfer from the incident ions to the target atom, $E_{p,max}$, is given by

$$E_{p,max} = 4M_1M_2E/(M_1 + M_2)^2,$$

where, E, M_1, M_2 are the energy and mass of incident ions and the mass of target atoms, respectively. According to the displacement threshold energy of 40 eV

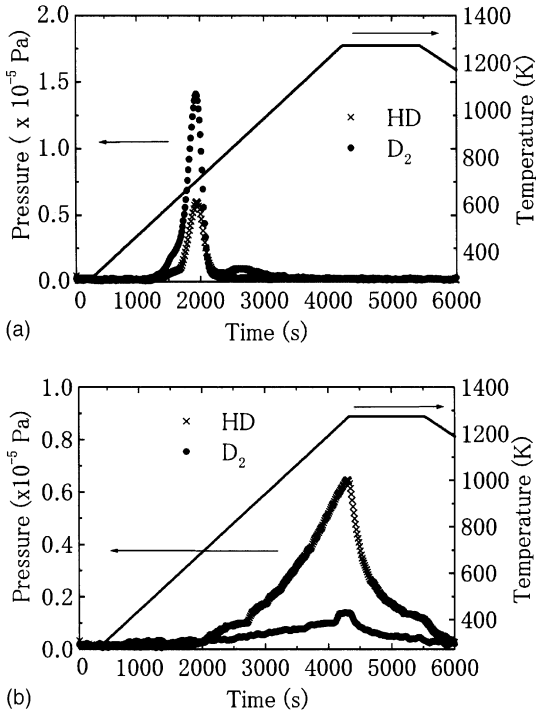

Fig. 3. Thermal desorption spectrum of HD and D_2: (a) PM-W after exposure to deuterium plasma (100 eV, 7.50×10^{25} D m^{-2}, 823 K), (b) VPS-W/CX-2002U after exposure to deuterium plasma (100 eV, 3.75×10^{25} D m^{-2}, 838 K). The thickness of VPS-W is 1.0 mm.

for tungsten [7], the minimum energy of deuterium ions for the displacement damage production are calculated to be 1236 eV, using the above equation. This implies that a displacement damage is not formed at the present experimental condition. A lot of research about blister formation due to gas ion irradiation has been carried out, but only for energies high enough to cause elastic damage [10]. These studies have shown that one of the necessary conditions for blister formation in metals is the agglomeration of implanted gas atoms and vacancies which are formed by displacement damage to form to gas bubbles in near-surface region. Therefore, the blister formation in the present experiment under parameters more relevant for fusion devices is different from that of

Table 1
Total desorption and incident deuterium

Sample	Irradiation temperature (K)	Fluence (D m^{-2})	Total desorption (D_2 m^{-2})
PM-W	843	7.20×10^{24}	6.35×10^{18}
PM-W	823	7.50×10^{25}	2.35×10^{20}
VPS-W/CX-2002U[a]	708	3.48×10^{25}	2.53×10^{20}
VPS-W/CX-2002U[b]	838	3.75×10^{25}	5.09×10^{20}

[a] The thickness of the VPS-W layer is 0.5 mm.
[b] The thickness of the VPS-W layer is 1.0 mm.

the previous studies. One of the possible mechanisms is that implanted deuterium diffuses deeper into PM-W, and agglomerates at grain boundaries with parallel direction of the surface, which were produced by rolling process during the fabrication of PM-W. Recent observations of the modification of tungsten surfaces exposed to low energy, high flux plasmas [8] and ion beam bombardment [9] have shown the formation of blisters. Experiments are underway to try to investigate and identify systematically the mechanisms responsible for this behavior. Since blister may result in formation of flaking and exfoliation at higher fluence due to further deuterium exposure, the surface modification by low energy and high flux hydrogen isotope irradiation may be an important erosion process for surfaces exposed to the plasma.

On the other hand, in the case of VPS-W, no modification such as blistering was observed. This reason is expected to be that the surface is hard to modify and that deuterium is hard to accumulate due to surface unevenness and pores near surface of VPS-W/CX-2002U. From this viewpoint, it is concluded that VPS-W is superior to PM-W.

4.2. Retention property of deuterium

The deuterium release peak occurs at a temperature that is lower than the exposure temperature as shown in Fig. 3(a). One possible reason is that tungsten absorbs deuterium which cools down. A test in a TDS oven have been performed but this effect has not been observed. Therefore, it is expected that as the sample cooled after the plasma exposure, the diffusing deuterium atoms already in the sample may become trapped in sites that remain unoccupied during the high temperature in the present plasma exposure experiment.

The deuterium desorption behavior between PM-W and VPS-W/CX-2002U was different. Judging from comparison with TDS results [11,12], the implanted deuterium on the surface of VPS-W/CX-2002U diffuses through the tungsten layer and the PVD Re/W multilayer, and is trapped in the CX-2002U substrate. These results indicate that the deuterium retention is influenced by substrate materials and surface materials. It is required to evaluate not only surface materials but also substrate materials.

5. Conclusion

Plasma sprayed tungsten coated CFC (VPS-W/CX-2002U) and PM-W have been exposed to a high deuterium flux with low energy. Surface modification and deuterium retention property after the exposure have been investigated to prove the suitability of such materials in fusion devices.

(1) Blisters are formed on the PM-W by deuterium irradiation with a fluence of 7.5×10^{25} m^{-2}. The amount of blisters and their average size increase with an increase of a fluence to 3.00×10^{26} m^{-2}. On the other hand, no modification is observed on the VPS-W/CX-2002U. This difference expected to be caused by surface morphology and structure.

(2) Desorption property from VPS-W/CX-2002U irradiated by deuterium is different from that of PM-W. The reason of this behavior is expected that implanted deuterium on the surface of VPS-W/CX-2002U diffused CX-2002U through the VPS-W and trapped in the CX-2002U.

Acknowledgements

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References

[1] R. Neu, K. Asmussen, S. Deschka, A. Thoma, M. Bessenrodt-Weberpals, R. Dux, W. Engelhardt, J.C. Fuchs, J. Gaffert, C. Garcia-Rosales, A. Herrmann, K. Krieger, F. Mast, J. Roth, V. Rohde, M. Weinlich, U. Wenzel, J. Nucl. Mater. 241–243 (1997) 678.
[2] C. Garcia-Rosales, S. Deschka, W. Hohenauer, R. Duwe, E. Gauthier, J. Linke, M. Lochter, W. Mallener, L. Plochl, P. Rodhammer, A. Salito, and the ASDEX-Upgrade Team, Fus. Technol. 32 (1997) 263.
[3] K. Tokunaga, N. Yoshida, N. Noda, T. Sogabe, T. Kato, J. Nucl. Mater. 258–263 (1998) 998.
[4] K. Tokunaga, N. Yoshida, N. Noda, Y. Kubota, S. Inagaki, R. Sakamoto, T. Sogabe, R. Plochl, J. Nucl. Mater. 266–269 (1999) 1224.
[5] K. Tokunaga, N. Yoshida, Y. Kubota, N. Noda, Y. Imamura, T. Oku, A. Kurumada, T. Sogabe, T. Kato, L. Plochl, Fus. Eng. Des. 49&50 (2000) 371.
[6] Y. Hirooka, R.W. Conn, T. Sketchley, W.K. Leung, G. Chevalier, R. Doerner, J. Elverum, D.M. Goebel, G. Gunner, M. Khandagle, B. Labombard, R. Lehmer, P. Luong, Y. Ra, L. Schmitz, G. Tynan, J. Vac. Sci. Technol. A 8 (1990) 1790.
[7] P. Lucasson, in: M.T. Robinson, F.W. Young Jr. (Eds.), Fundamental Aspects of Radiation Damage in Metals, vol. 1, CONF-751006-P1, USERDA 1075, p. 42.
[8] F.C. Sze, R.P. Doerner, S. Luckhardt, J. Nucl. Mater. 264 (1999) 89.
[9] A.A. Haasz, M. Poon, J.W. Davis, J. Nucl. Mater. 266–269 (1999) 520.
[10] M. Kaminsky (Ed.), Radiation Effects on Solid Surface, American Chemical Society, 1976, p. 112.
[11] V.K. Alimov, B.M.U. Scherzer, J. Nucl. Mater. 240 (1996) 75.
[12] T. Tadokoro, S. O'hira, M. Nishi, K. Isobe, J. Nucl. Mater. 258–263 (1998) 1092.