Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

ELSEVIER



Erosion yields of deposited beryllium layers

D. Nishijima*, R.P. Doerner, M.J. Baldwin, G. De Temmerman

Center for Energy Research, University of California at San Diego, 9500 Gilman Dr., La Jolla, CA 92093-0417, USA

ARTICLE INFO

PACS: 52.40.Hf

ABSTRACT

Erosion yields of various Be surfaces exposed to deuterium plasma, including polycrystalline Be (PC-Be), magnetron and thermionic vacuum arc deposited surfaces, and *in situ* plasma-deposited Be layers on Be (DP-Be/Be) and on graphite (DP-Be/C) targets, are measured in the linear divertor plasma simulator PISCES-B. It is observed that the enhanced erosion (~3 times higher than PC-Be) of DP-Be/Be occurs at sample temperature $T_{\rm s} \sim 310$ K, while DP-Be/Be possesses the same yield as PC-Be at a higher $T_{\rm s} \sim 570$ K. The erosion yield of DP-Be/C is found to be slightly decreased at $T_{\rm s} \sim 850$ K. This is consistent with the more effective formation of Be₂C at higher $T_{\rm s}$, which has a higher surface binding energy than Be. It has been identified from spectroscopic measurements that Be is also eroded as BeD.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

Eroded particles from plasma-facing materials in fusion devices can migrate and then (re-)deposit on surfaces of the same and/or different kinds of materials. Such a deposited layer may have a different erosion yield from its original material. Furthermore, material mixing with substrate materials can occur, and the erosion yield of the mixed material layer can be also different from both the deposited material and the substrate material. These can affect the process of material migration. A few investigations on carbon deposits have been reported; an enhanced chemical erosion yield around five times higher than graphite needs to be assumed in a simulation with the ERO Monte-Carlo code to reproduce the ¹³C deposition pattern in a carbon layer on top of an aluminum target plate [1]. In addition, it has been reported that the enhanced erosion of carbon deposits can affect tritium inventory in ITER [2].

In ITER [3] as well as during the planned ITER-like-wall phase of JET [4], the first wall is foreseen to be beryllium (Be). It is therefore expected that eroded Be will deposit on the Be first wall and the divertor carbon and tungsten materials. However, only a few investigations on the erosion properties of Be deposited layers have been made [5]. In this paper, measured erosion yields of *in situ* plasma-deposited Be layers on Be and on graphite targets exposed to deuterium plasma are presented. Furthermore, erosion yields of *ex situ* deposited Be layers fabricated by magnetron sputtering and thermionic vacuum arc techniques are also shown. The details of these layers are given in [6]. For comparison, erosion yields of polycrystalline Be are measured, which turn out to be around seven times smaller than the calculated value using the TRIM.SP code [7]. The reason for this difference is discussed in terms of the effect

* Corresponding author. E-mail address: dnishijima@ferp.ucsd.edu (D. Nishijima). of background D atom adsorbed onto the Be surface. Erosion of beryllium deuteride (BeD) from Be surfaces has been identified from its $A^2\Pi - X^2\Sigma^+$ band emission, which was also observed in a divertor plasma of JET [8]. The contribution of BeD to the total Be erosion is discussed.

2. Experimental setup

Experiments are carried out in PISCES-B [9], where the following Be surfaces are exposed to deuterium plasma:

- PC-Be: polycrystalline Be,
- MS-Be: Be layers on nickel by the magnetron sputtering technique,
- TVA-Be: Be layers on inconel by the thermionic vacuum arc technique,
- DP-Be/Be: in situ plasma-deposited Be layers on PC-Be,
- DP-Be/C: *in situ* plasma-deposited Be layers on a fine grain graphite.

In situ plasma-deposited Be layers are made by the following manner; Be atoms are injected into the plasma using a Be oven, and are ionized and subsequently transported to the target (see Fig. 1). To efficiently make Be deposits, the target is at a floating potential, corresponding to an incident ion energy, E_i , of ~10–20 eV.

The sample temperature, T_s , is measured with a thermocouple contacted to the rear side of the target. Note that T_s was kept below 1000 K by water cooling during erosion yield measurements, therefore no enhanced erosion of Be at elevated temperatures is expected [10].

Since the geometrical loss flux of eroded Be atoms is not negligible in the conditions here, i.e. the ionization flux of eroded Be atoms is smaller than the eroded Be flux, the absolute erosion yield

^{0022-3115/\$ -} see front matter @ 2009 Elsevier B.V. All rights reserved. doi:10.1016/j.jnucmat.2009.01.144



Fig. 1. Schematic of the target region in PISCES-B.

can not be obtained from the spectroscopic technique. Thus, an absolute erosion yield of PC-Be at $E_i \sim 80 \text{ eV}$ is first determined with the mass loss technique. To minimize the re-deposition fraction of eroded Be, the target was exposed for 7200 s to a low ion flux ($\Gamma_i \sim 0.9 \times 10^{22} \text{ m}^{-2} \text{ s}^{-1}$) deuterium plasma with electron density $n_e \sim 0.7 \times 10^{18} \text{ m}^{-3}$ and electron temperature $T_e \sim 9 \text{ eV}$. The plasma parameters at the center of the plasma column are measured with a reciprocating double probe system, and are almost flat over the exposed area of the sample. The ionization mean free path of eroded Be atoms is estimated to be ~300 mm, much longer than the plasma radius of ~25 mm. The resulting mass loss was 1.50 mg. As also found in Ref. [11], the obtained erosion yield of ~4 × 10⁻³ is around seven times lower than the TRIM.SP calculation [7]. The reason for this difference will be discussed in the next section.

The intensity of Be I (457.3 nm) line emission of eroded Be atoms from PC-Be at $E_i \sim 80 \text{ eV}$ is then measured through the Doppler view (see Fig. 1), and is normalized to the absolute erosion yield of 4×10^{-3} to derive the correction factor. This correction factor is multiplied with the Be I line intensities measured at different E_i of PC-Be and with other Be surfaces to obtain the E_i dependence of the absolute erosion yield. For this E_i scan by negatively biasing the target, the plasma parameters are set to $n_e \sim 2 \times 10^{18} \text{ m}^{-3}$, $T_e \sim 8 \text{ eV}$, and $\Gamma_i \sim 2 \times 10^{22} \text{ m}^{-2} \text{ s}^{-1}$ to facilitate the detection of the Be I line.

The $A^2\Pi - X^2\Sigma^+$ band emission of BeD is observed in front of a PC-Be sample through the Axial spectroscopic view (see Fig. 1). The intensity of the Be I (457.3 nm) line is also monitored.

3. Experimental results and discussion

3.1. Erosion yields of PC-Be and ex situ deposited Be layers

Fig. 2(a) shows the erosion yields of PC-Be, MS-Be and TVA-Be as well as theoretical curves. The theoretical values are normalized to 4×10^{-3} at $E_i = 80$ eV. The solid curve depicts D⁺ bombardment data, $Y_{D^+}(E_i)$ [7], while the dashed curve is obtained by considering an ion species fraction of $(D^+, D_2^+, D_3^+) = (0.25, 0.47, 0.28)$, predicted with a 0-D model [12] at $n_e = 2 \times 10^{18}$ m⁻³, $T_e = 8$ eV, and deuterium gas pressure $P_{D_2} = 6$ mTorr. Thus, the dashed curve is calculated as $0.25 \times Y_{D^+}(E_i) + 0.47 \times 2 \times Y_{D^+}(E_i/2) + 0.28 \times 3 \times Y_{D^+}(E_i/3)$ [11]. It is seen that MS-Be and TVA-Be have the same erosion yields as PC-Be. In addition, the energy dependence of the three surfaces agrees well with the theoretical curves.



Fig. 2. Erosion yields of (a) PC-Be (circles), MS-Be (squares), and TVA-Be (diamonds), (b) DP-Be/Be at $T_s \sim 310$ K (squares) and ~ 570 K (diamonds) as well as PC-Be (circles) for comparison. The theoretical yields are also shown; the solid curve presents the D⁺ bombardment case [7], while the dashed curve is obtained by considering an ion species fraction of (D⁺, D⁺_2, D⁺_3) = (0.25, 0.47, 0.28), predicted using a 0-D model [12] at $n_e = 2 \times 10^{18}$ m⁻³, $T_e = 8$ eV, $P_{D_2} = 6$ mTorr.

As mentioned above, the measured erosion yields of PC-Be are around seven times smaller than the TRIM.SP calculation [7]. This discrepancy has been already pointed out in Ref. [11], where the effect of oxide layer is discussed, and is expected to be negligible under the plasma exposure condition ($\Gamma_i \sim 10^{22} \text{ m}^{-2} \text{ s}^{-1}$). Here, we discuss effects of background D atom adsorption onto the Be surface, leading to the reduction of Be surface concentration. Note that it has been reported that D₂ does not adsorb on Be surfaces [13]. According to Ref. [14], the steady-state surface concentration of adsorbed D on Be can be written as,

$$c_{\rm D} = \frac{\Gamma_{\rm D} \gamma_{\rm D}}{\Gamma_{\rm D} \gamma_{\rm D} + \Gamma_i Y_{\rm D}},\tag{1}$$

where Γ_D , γ_D , and Y_D are the background D atom flux, the sticking probability of D on Be, and the erosion yield of D by deuterium ions bombardment, respectively. A condition, where the effects of background gas on the erosion yield of the substrate are negligible,

$$c_{\rm D} < 0.1 \quad \text{or} \quad \frac{\Gamma_{\rm D} \gamma_{\rm D}}{\Gamma_i Y_{\rm D}} \leqslant 0.1,$$
 (2)

is proposed in Ref. [14]. In the condition used here, $\Gamma_{\rm D} = (1/4)n_{\rm D}v_{\rm D}$ is estimated to $\sim 2 \times 10^{22}$ m⁻² s⁻¹ from $n_{\rm D} \sim n_{\rm D_2}/20$ [15] and assumed average Franck–Condon energy of 2.2 eV for calculating the D atom velocity $v_{\rm D}$. Here, $n_{\rm D}$ and $n_{\rm D2}$ are the densities of D and D₂, respectively. Since this D flux is comparable to $\Gamma_{\rm i} \sim 2 \times 10^{22}$ m⁻² s⁻¹, the condition Eq. (2) may be violated, although $Y_{\rm D}$ and $\gamma_{\rm D}$ are unknown. Namely, the concentration of adsorbed D on the Be surfaces increases, resulting in a decrease in the concentration of Be on the surfaces. Therefore, the erosion yield of Be may be lowered.

Unfortunately, it is difficult to scan the ratio Γ_D/Γ_i in PISCES-B to further investigate the background D atom effect. In JET, the erosion yields of the Be divertor target have been measured and compared to simulations [16]. The measured yields are also lower than the simulations, and the difference between the experiment and simulation becomes larger in the high recycling phase with a higher neutral flux than in the low recycling phase. On the other hand, it has been reported that ion beam data, performed in high vacuum, agree well with theoretical values [17]. These observations in different devices can demonstrate the effect of background D flux on Be erosion, lowering the Be erosion yield in plasma environments.

3.2. Erosion yields of DP-Be/Be and DP-Be/C

The erosion yields of DP-Be/Be are presented in Fig. 2b, where the erosion yields of PC-Be are also shown for comparison. It is clearly seen that the erosion of DP-Be/Be at lower $T_{\rm s} \sim 310$ K is enhanced by a factor of ~3 compared to PC-Be. On the other hand, the enhanced erosion is not observed at higher $T_s \sim 570$ K. This T_s dependence of the erosion yields of the deposited Be layer may be attributed to the difference in deuterium retention in the layer. At lower T_s , the amount of deuterium in the deposited layer is expected to be higher than that at higher T_s , possibly lowering the surface binding energy of Be. Therefore, the erosion of DP-Be/Be can be enhanced at lower T_s. Similarly, it is observed in PISCES-B that deposited Be layers on witness plates [18], not directly exposed to plasma but to energetic D atom flux reflected at the target, are systematically thinner at lower witness plate temperature \sim 370 K than that at \sim 570 K. This may be also due to the enhanced erosion at lower temperature.

In the case of DP-Be/C, Be₂C can be formed on the graphite surface, depending strongly on T_s [19]. In addition, a pure Be layer on the Be₂C layer can be also made [19], since Be is deposited at low $E_i \sim 10-20$ eV. The Be erosion yields of DP-Be/C at $T_s \sim 850$ K are shown in Fig. 3. The yields are corrected by Be surface concentrations, Be/(Be + C), obtained with *in situ* Auger electron spectros-



Fig. 3. Erosion yields of DP-Be/C (triangles) at $T_{\rm s} \sim 850$ K with Be deposition temperature of ~1000 K. The data indicated by (#) may represent Be₂C erosion yields. For comparison, the measured and scaled theoretical yields of PC-Be are also shown, the symbols and lines of which are the same as Fig. 2.

copy. Two data points indicated by (#) show somewhat lower yields than the others. These are measured at the end of the E_i scan, i.e. after the pure Be over-layer is removed. These may, therefore, represent erosion yields of Be₂C, having a higher surface binding energy than Be. The other data, which presumably come from the pure Be layer, show the same erosion yields as PC-Be, which is consistent with the observation of DP-Be/Be at high T_s mentioned above.

3.3. Contribution of BeD erosion

Due to the interaction between deuterium plasma and solid Be, non-volatile BeD₂ can be formed on the surface. In fact, the $A^2\Pi$ - $X^2\Sigma^+$ band emission of BeD molecules has been observed in front of Be targets. Fig. 4 presents examples of the spectrum observed in front of a PC-Be target exposed to deuterium plasma at different $T_{\rm s}$. The A–X band intensity is seen to decrease by a factor of more than 2, when T_s is increased from 355 to 638 K. Since the other plasma parameters are identical in the two cases, it is thought that the observed BeD molecules are released from the surface. This decrease in the intensity is consistent with the fact that BeD₂ starts to be decomposed at \sim 450 K [20]. On the other hand, the total Be erosion yield of PC-Be measured from mass loss, which includes erosion of both Be atoms and BeD molecules, showed no clear T_s dependence in this temperature range [11]. Therefore, the fraction of BeD to the total Be erosion is expected to be small for PC-Be. Note that the Be I (457.3 nm) line intensity from eroded Be atoms does not change (within 10% only) by this T_s increase. This is consistent with the mass loss measurements. Detailed investigations of BeD erosion for deposited Be layers will be performed in relation to the enhanced erosion at low T_s .

4. Conclusion

The erosion yields of various Be surfaces bombarded by deuterium plasma have been measured in PISCES-B. For the *ex situ* deposited Be layers (MS-Be and TVA-Be) and PC-Be (i.e. no codeposited D), the D atom flux may be important in determining the Be erosion yield. Under certain conditions ($\Gamma_i \sim \Gamma_D$ as in PISCES-B), the adsorbed D on a Be surface may lower the Be surface



Fig. 4. $A^2\Pi - X^2\Sigma^*$ ($\Delta v = 0$ sequence) band emission spectra of BeD molecules eroded from a PC-Be target due to deuterium plasma exposure at $T_s \sim 355$ and 638 K. These spectra are taken in front of the target (z = 1.4 mm). The plasma parameters ($n_e \sim 2.6 \times 10^{18}$ m⁻³, $T_e \sim 8$ eV, and $E_i \sim 34$ eV) are identical in the two cases.

concentration. This results in a lower Be erosion yield than predicted by TRIM.SP. For the *in situ* plasma-deposited Be layers on Be, the presence of co-deposited D affects the Be erosion yield. This is demonstrated by change in the yield at different temperatures. Namely, the enhanced erosion of DP-Be/Be by a factor of ~3 is observed at lower $T_s ~ 310$ K, while at higher $T_s ~ 570$ K the erosion yields are the same as PC-Be. The Be erosion yield from Be₂C surface is reduced as a result of a higher surface binding energy. There is evidence for the formation of BeD₂ on surface of PC-Be exposed to deuterium plasma from the presence of BeD band emission during erosion. The correlation of decreasing yield of BeD with increasing temperature is further evidence of the BeD₂ formation, and suggests that this is a possible mechanism for Be erosion. Ongoing work will evaluate this mechanism further.

Acknowledgements

The authors express their sincere thanks to PISCES technical staff and Be personnel for their professional skill and dedication. We appreciate Dr C.P. Lungu for providing us with the Be coating samples, and Dr E.M. Hollman for his 0-D code. We also appreciate the referees for their valuable comments. This work is conducted under the US DOE Contract: DE-FG02-07ER54912.

References

- [1] A. Kirschner et al., J. Nucl. Mater. 328 (2004) 62.
- [2] J. Roth et al., J. Nucl. Mater. 337-339 (2005) 970.
- [3] A. Loarte et al., Nucl. Fus. 47 (2007) S203.
- [4] J. Pamela et al., J. Nucl. Mater. 363-365 (2007) 1.
- [5] K. Krieger et al., J. Nucl. Mater. 390–391 (2009) 110.
- [6] A. Widdowson et al., J. Nucl. Mater. 390–391 (2009) 988. [7] W. Eckstein, Calculated Sputtering, Reflection and Range V
- W. Eckstein, Calculated Sputtering, Reflection and Range Values, Report of the Max-Planck-Institute f
 ür Plasmaphysik, IPP-Report 9/132, Garching, Germany, 2002.
- [8] G. Duxbury et al., Plasma Phys. Control. Fus. 40 (1998) 361.
- [9] R.P. Doerner et al., Phys. Scr. T111 (2004) 75.
- [10] R.P. Doerner et al., J. Appl. Phys. 95 (2004) 4471.
- [11] R.P. Doerner et al., J. Nucl. Mater. 257 (1998) 51.
- [12] E.M. Hollmann, A.Yu. Pigarov, Phys. Plasmas 9 (2002) 4330.
- [13] S. Zalkind et al., Appl. Surf. Sci. 115 (1997) 273.
- [14] H.H. Andersen, H.L. Bay, Sputtering yield measurements, in: R. Behrisch (Ed.), Sputtering by Particle Bombardment I, Springer-Verlag, Berlin, 1981, p. 145.
- [15] D.G. Whyte et al., J. Vac. Sci. Technol. A 17 (1999) 2713.
- [16] H.Y. Guo et al., J. Nucl. Mater. 241-243 (1997) 385.
- [17] J. Roth et al., Fus. Eng. Des. 37 (1997) 465.
- [18] G. De Temmerman et al., Nucl. Fus. 48 (2008) 075008.
- [19] M.J. Baldwin et al., J. Nucl. Mater. 358 (2006) 96.
- [20] R.P. Doerner et al., J. Nucl. Mater. 390-391 (2009) 681.