ELSEVIER

Journal of Nuclear Materials 258–263 (1998) 628–633

journal of nuclear materials

Erosion and surface morphology of graphite materials under high flux beam irradiation

Y. Ueda *, T. Sugai, K. Shiota, Y. Ohtsuka, Y. Isobe, M. Nishikawa

Department of Electronic, Information Systems and Energy Engineering, Graduate School of Engineering, Osaka University, 2-1 Yamada-Oka, Suita, Osaka 565, Japan

Abstract

Surface morphology and its relation to the erosion mechanism of graphite materials (pyrolytic graphite (PG) and isotropic graphite) irradiated by high flux beams were studied. The irradiation was made by a 5 keV Ar beam with a maximum flux of 1.3×10^{21} Ar/m^2 s with irradiation angles and fluences, and sample temperatures as experimental parameters. In the case of PG, the surface morphology was different between irradiation at RT (physical sputtering) and at elevated temperatures (1980 K, radiation enhanced sublimation (RES)). In addition, irradiation at oblique incidence (60°) tended to flatten the surface at RT, while at 1980 K the surface roughness developed to some extent. No clear flux dependence of the surface morphology was observed at 1980 K as long as the mean eroded depth was similar, though the RES yield was flux-dependent (decrease with flux). In the case of isotropic graphite, the distinctive surface morphology observed for PG was not found.

1. Introduction

Ion induced particle emission is characterized by yields of sputtering and electron emission, which are generally related to types of materials, species of incident particle and its energy and an angle of incidence. In actual plasma facing materials (PFMs), however, the effective yields are much more complicated functions due to the effects of not only multiple ion species in plasma, and energy and angular distribution of incident particles but also the surface conditions of materials.

One of the important surface characteristics affecting the yields of particles is surface morphology. In actual edge plasmas, the angle of incidence of impinging ions onto PFMs is oblique and it is important to know the angular dependence of the yield. However, the surface roughness tends to smear out the angular dependence of the effective sputtering yield [1,2]. In addition, surface roughness also affects the effective yield of secondary electron emission [3–5]. Therefore, it is important to study the evolution of surface morphology under actual edge plasma conditions.

The evolution of surface morphology of ion irradiated surfaces was studied in the field of surface analysis, for instance, depth profiling of thin films by ion beam etching [6]. In this case, surface roughness caused by ion irradiation degraded a depth resolution. The surface morphology modification caused by ion irradiation appears as ripples, cones, whiskers, and so on. It is known that the effects of physical sputtering alone is not sufficient to form the morphology [7]. The importance of surface diffusion has been also stressed for the modification.

For the last several years, we have paid special attention to high flux effects on erosion of graphite and graphite based materials in order to simulate high flux conditions at edge plasmas by using high flux ion beam generator [8–12]. One of our important results is the reduction of the yield of radiation enhanced sublimation (RES) [13] at elevated temperatures irradiated at high fluxes close to edge plasma conditions [9]. RES is known to be initiated by interstitial production followed by its diffusion to the surface and sublimation [14,15]. This model predicts significant reduction of the RES yield at high fluxes, which is caused by the increase in recom-

* Corresponding author. Tel.: +81 6 879 7236; fax: +81 6 879 7867; e-mail: yueda@ppl.eng.osako-u.ac.jp.

0022-3115/98/$19.00 © 1998 Elsevier Science B.V. All rights reserved.
PII: S 0 0 2 2 - 3 1 1 5 (9 8) 0 0 1 1 1 - 1

bination rate of interstitials and vacancies with flux due to the increase in the density of vacancies. Since the diffusion of atoms can play an important role in the modification of surface morphology as was mentioned in the previous paragraph, the surface morphology of graphite irradiated by high flux beams could show some flux dependence and give some new information on the surface modification mechanism. So far, however, no report was found from this viewpoint.

In this paper, we will show the observation results of surface morphology of graphite materials (PG and isotropic graphite) irradiated by high flux beams for physical sputtering and RES with the discussion on the relation between the morphology and erosion characteristics.

2. Experiment

The high flux beam irradiation device consists of a conventional bucket type ion source equipped with a spherical multiaperture extractor and an irradiation chamber with a movable target stage. The schematic drawing of this device was shown elsewhere [16]. On this movable stage, a rotatable sample holder and a two-dimensional calorimeter array for beam power density measurements are installed. The effective diameter and the radius of curvature of the ion beam extractor are 14 and 50 cm, respectively, and the convergence angle is about 8°. This system operates in a pulsed mode with a maximum pulse length of 2.0 s (a duty cycle of about 2 min) due to accumulation of discharge gas in the irradiation chamber. The irradiation was made by a 5 keV Ar beam with the maximum flux of about 1×10^{21} m^{-2} s^{-1}. Irradiation fluxes shown in this paper are always with respect to sample surfaces.

Irradiation samples were isotropic graphite (IG-430, Toyo Tanso) and pyrolytic graphite (PG, Union Carbide). The sample sizes were $40 \times 10 \times 0.2$ (Isotropic), or $40 \times 10 \times 0.1$ (Pyrolytic) mm^3 and heated directly by passing a current. During irradiation the current was reduced to compensate for the increase in heating power from the beam, which made the surface temperature almost constant (within ±25 K). The surface temperature was measured by a two-color infrared pyrometer. Surface morphology was observed by a scanning electron microscope (SEM), in which the observation angle can be varied from 0° to 90°.

3. Surface morphology of pyrolytic graphite

Fig. 1 shows the surface morphology of PG irradiated at RT (room temperature) for different irradiation angles. During irradiation (0.5 s duration in this case) the sample temperature increased from RT to about 1000 K due to the high beam power (0.6 MW/m^2). Even at this maximum temperature (1000 K), physical sputtering still dominates the erosion of graphite. The mean eroded depths for normal incidence (Fig. 1(a)) and oblique incidence (60°, Fig. 1(b)) are 1.5 and 1.7 μm, respectively.

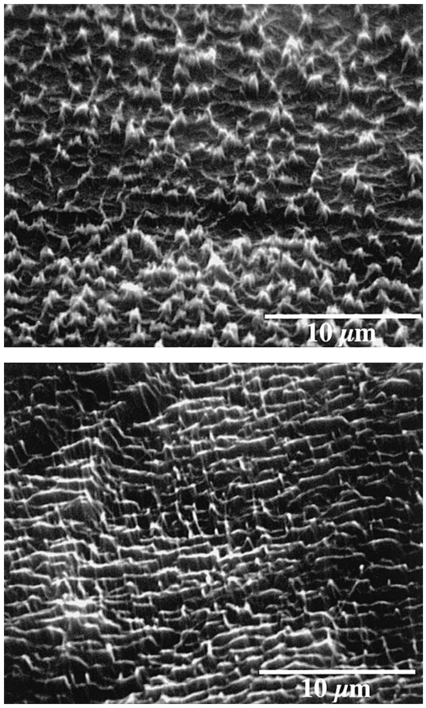

Fig. 1. SEM micrographs of PG irradiated at RT at irradiation angles of (a) 0° and (b) 60°. Irradiation flux is about 8×10^{21} m^{-2} s^{-1}. The viewing angle of SEM is 45°.

For normal incidence (Fig. 1(a)), surface microstructures consisting of a dense array of uniformly sized cone-like structure were observed. This structure was quite similar to that obtained by other ion beam experiments (0.75 keV Ar beam with similar fluence) [3], in which this structure grew to larger sized cones afterwards. Therefore, our experimental results at the temperature in which physical sputtering dominates the erosion are basically the same as that of the previous work even though irradiation flux is much higher in our experiments (8×10^{21} m^{-2} s^{-1}, roughly an order of magnitude higher). For oblique (60°) angle of incidence (Fig. 1(b)), the surface morphology became much flatter than that of the normal incidence because of shadowing effect of physical sputtering [7]. Some experiments done by other groups showed whiskers, which were thin and long columns pointing in the beam direction observed on graphite [17–19]. In our experiments, such whiskers were not found in any of the cases probably due to very high heat load and/or the distribution of angle of incidence (±8°) due to geometrical focussing.

Fig. 2 shows the surface morphology at 1980 K, in which RES process dominates the erosion. The irradiation flux was 2.0×10^{20} Ar/m^2 s. The fluences were 4.4×10^{18} Ar/m^2 (Fig. 2(a)) and 8.4×10^{18} Ar/m^2 (Fig. 2(b)), leading to a mean eroded depths of about 1.7 μm and 3.1 μm, respectively, estimated from weight losses. By comparing the surface morphology between the RES case (Fig. 2(a)) and the physical sputtering case (Fig. 1(a)), some different features were observed even though the mean eroded depth was similar. In the physical sputtering case the small cone-like protuberances are scattered almost uniformly, while in the RES case some of the protuberances coalesce to form wrinkle-like structures. It is noteworthy that this structure grew self-similarly both in width and height roughly in proportion to the fluence as the fluence was increased from 4.4×10^{22} (Fig. 2(a)) to 8.4×10^{22} Ar/m^2 (Fig. 2(b)). Since crystallization of graphite by annealing process was clearly observed around 1300 K [20,21], the diffusion of carbon atoms could be significant in the RES condition at 1980 K compared with the physical sputtering condition at temperatures less than 1000 K. Therefore, the difference of surface morphology could be attributed to the surface diffusion effect of carbon atoms.

In the case of oblique incidence (60°) at 1980 K (Fig. 3), the surface of graphite irradiated with the fluence of 8.0×10^{22} Ar/m^2 also shows the similar surface protuberances to that of the normal incidence case but they are aligned along beam direction, the height is lower, and density is higher. In Fig. 1, shadowing effect of physical sputtering seems to work to flatten the surface, while in the RES case the effect is not sufficient for flattening and still small protuberances of the order of μm remain.

Fig. 4 shows the SEM micrograph in the case of RES at higher fluxes (1.3×10^{21} Ar/m^2 s^1) than that shown in Fig. 2. Although the irradiation fluences were close between the cases of Figs. 2(a) and 4(a), and that of Figs. 2(b) and 4(b), the evolution of the structure was different. In the high flux cases (Fig. 4), the height of the protuberances was much lower than that in the low flux cases (Fig. 2). This result can be related to the decrease of RES yield with flux. Fig. 5 shows the RES yield and flux of interstitial to the surface of graphite calculated by the simple model including interstitial production and diffusion [14]. The RES yield curve is a good approximation of experimental data [10]. According to this model, the RES yield over 10^{20} m^{-2} s^{-1} changes as $\phi^{-0.25}$

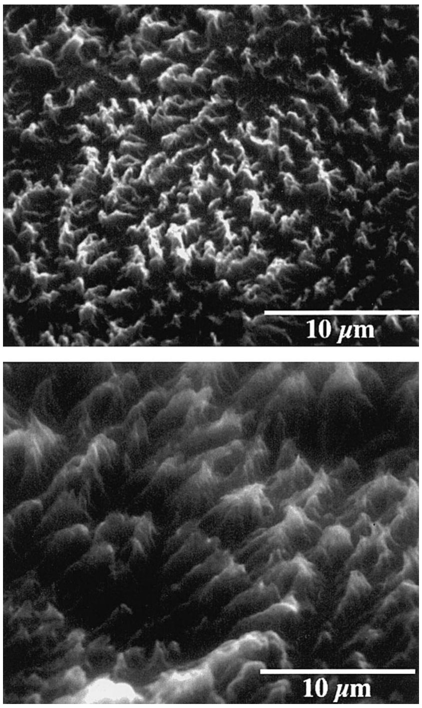

Fig. 2. SEM micrographs of PG irradiated at 1980 K at an angle of 0° with a flux of 2.0×10^{20} Ar/m^2 s^1. The irradiation fluences are (a) 4.4×10^{22} Ar/m^2 and (b) 8.4×10^{22} Ar/m^2. The viewing angle of SEM is 45°.

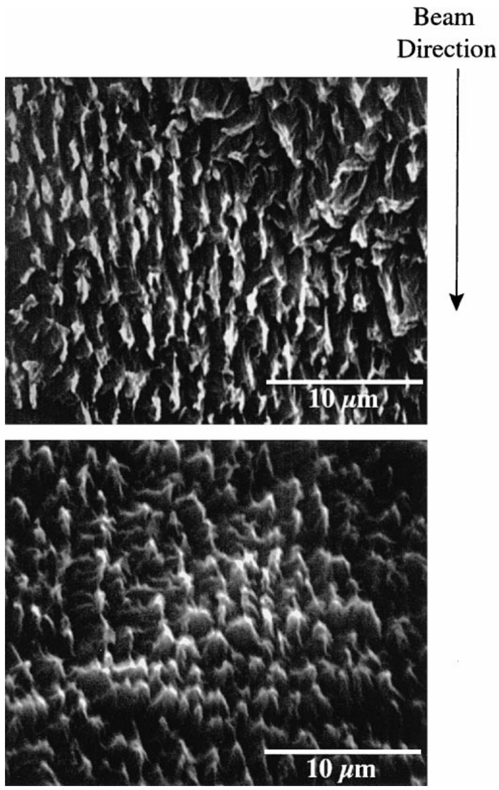

Fig. 3. SEM micrographs of PG irradiated at 1980 K at an angle of 60° with a flux of 2.0×10^{20} Ar/m^2 s^1. The viewing angles of SEM are (a) 0° and (b) 45°.

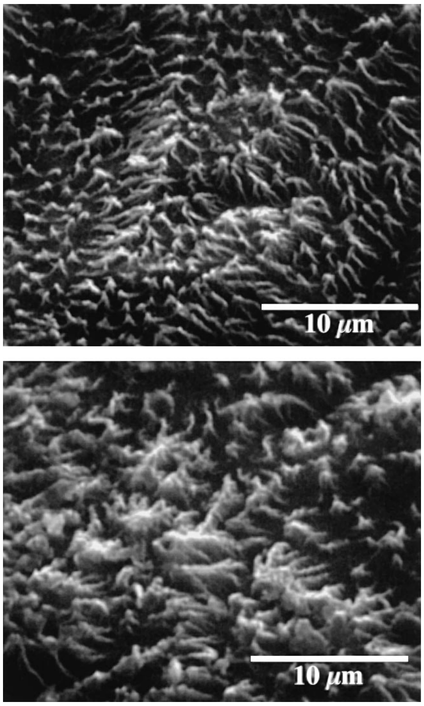

Fig. 4. SEM micrographs of PG irradiated at 1980 K at an angle of 0° with a flux of 1.3 × 10^{21} Ar/m^2 s^1. The irradiation fluences are (a) 3.4 × 10^{22} Ar/m^2 and (b) 7.8 × 10^{22} Ar/m^2. The viewing angle of SEM is 45°.

(ϕ: flux), and the interstitial flux to the surface scales $\phi^{0.75}$. The mean eroded depth in the high flux cases were 1.1 and 2.2 μm for the cases of Fig. 4(a) and (b), respectively. By comparing Fig. 2 (low flux cases) and Fig. 4 (high flux cases), it is considered that the dimensions of the protuberances roughly scale with the mean eroded depth regardless of the flux.

According to the work made by Rossnagel [17], flux (ion current density) as well as temperature could relate to the surface diffusion. The surface diffusion is enhanced with the increase of flux, though the mechanism for this enhancement is still unclear (probably related to dynamics of irradiation-induced defects). In our experiments, however, there is no clear difference of surface morphology by changing the flux (relating to interstitial flux to the surface shown in Fig. 5) by an order of magnitude.

4. Surface morphology of isotropic graphite

In real PFMs, isotropic graphite or C/C composite is used in general and it is important to study the evolution of surface morphology of these types of graphite. Fig. 6 shows the SEM micrographs of isotropic graphite (IG-430) irradiated at RT (Fig. 6(a), flux: 8.5 × 10^{20} m^{-2} s^{-1}) and at 1980 K (Fig. 6(b), flux: 1.3 × 10^{22} m^{-2} s^{-1}). Isotropic graphite IG-430 has relatively large grains and the magnification of SEM was reduced compared with the previous micrographs in order to observe global structures. In irradiation at RT, the mean eroded depths for normal incidence (Fig. 6(a)-1) and oblique incidence (60°) (Fig. 6(a)-2) were 1.1 and 2.6 μm, respectively. For normal incidence, the surface topology does not change by ion irradiation in contrast to the PG (Fig. 1(a)). This result suggests that the production of the ion bombardment-induced topography has close relation to the graphite structure. For oblique angle of incidence (60°), it was found that the surface smoothing took place except for pores.

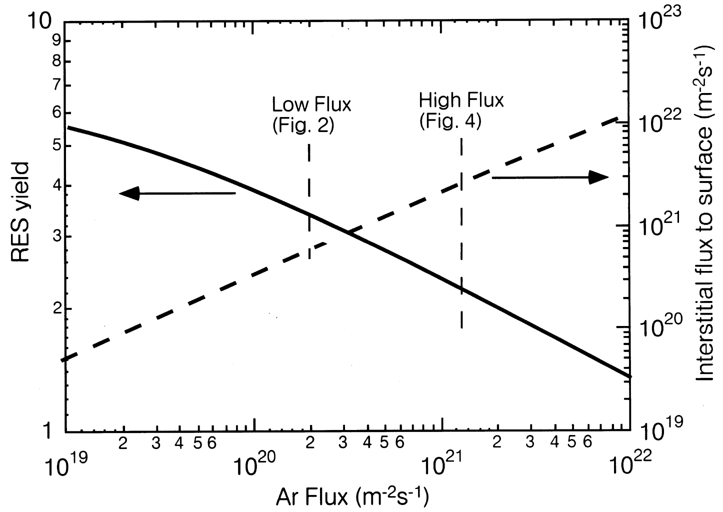

Fig. 5. Flux dependence of RES yield and interstitial flux to the surface of graphite irradiated by 5 keV Ar.

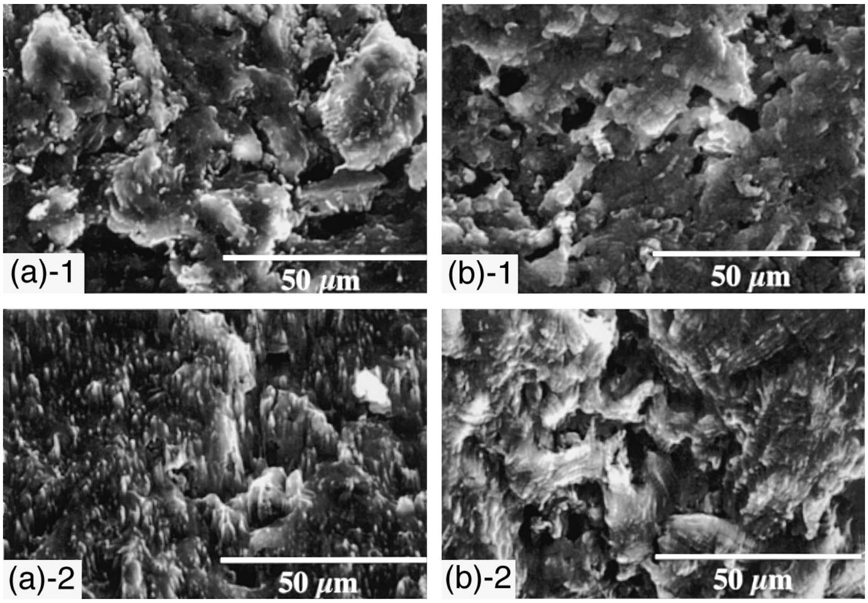

Fig. 6. SEM micrographs of IG-430 irradiated at RT for (a) and at 1980 K for (b). The irradiation angles are 0° for (a)-1 and (b)-1, and 60° for (a)-2 and (b)-2. The viewing angle of SEM is 0°.

In irradiation at 1980 K, the mean eroded depths were about 4.2 μm both for normal incidence (Fig. 6(b)-1) and oblique incidence (60°) (Fig. 6(b)-2). For normal incidence, round edge structure was observed in contrast to that irradiated at RT. The wrinkle-like structure observed in PG was not observed in this case. For oblique incidence, the surface flattening did not clearly occur compared with that irradiated at RT (Fig. 6(a)-2). This result is qualitatively similar to the case of PG irradiation. In addition, the edge of graphite crystal structure of grains appeared.

5. Discussions and conclusions

The angular dependence of the yields of physical sputtering and RES of graphite irradiated by our high flux beam was shown in Ref. [12]. The physical sputtering yields increase with an angle of incidence (less than 75°) in the same manner for PG and isotropic graphite (IG-430), though their surface morphology of unirradiated surface was quite different. These results would have a connection with the surface morphology after irradiation. For normal incidence, irradiation-induced roughness developed for PG, while IG-430 had intrinsic rough surface structures due to its porosity, which remained after irradiation. For oblique incidence (60°), the surfaces of both materials showed flattening due to shadowing effects. As a result, the surface structure was modified from the original one for both materials and the angular dependence might become similar.

In the case of RES, the experimental results on angular dependence were weaker than that of physical sputtering. In contrast to physical sputtering, the angular dependence of the RES yield from an ideal flat surface is not known very well. According to the interstitial diffusion model, however, the angular dependence of the RES yield could be weaker than that of physical sputtering [12]. In addition, significant enhancement of surface roughness was observed in any cases even at oblique incidence, which could also affect the weak yield dependence on the angle of incidence.

When studying the mechanism of RES at elevated temperature, it is usually assumed that the total yield is the sum of physical sputtering and RES. Therefore, RES yield is usually calculated as the total yield subtracted by physical sputtering yield at RT under the assumption that the physical sputtering yield is constant regardless of temperature. It is noteworthy that the surface morphology for samples irradiated at RT (e.g. in Fig. 1) is not the same as for those irradiated at elevated temperature (e.g. Fig. 3). For oblique incidence, surface roughness tends to be enhanced at elevated temperature and not at RT (flattening). Since surface roughness would reduce the yield at oblique incidence, the use of physical sputtering yield at RT for the estimation of RES yield could be inaccurate and a more comprehensive analysis would be needed.

In conclusion, for pyrolytic graphite physical sputtering at RT caused cone-like structure at normal incidence (not observed for IG-430) and flattening at oblique incidence (observed for IG-430 except for pores), while RES at 1980 K enhanced wrinkle-like rough structure at normal incidence (not observed for IG-430) and similar structure also appeared at oblique incidence (smaller than that at normal incidence). No flattening was observed for IG-430 irradiated at oblique incidence (60°) at 1980 K in contrast to the result at RT. These surface morphology could relate to the angular dependence of the effective yield in our experiments.

The change of irradiation flux (1.3×10^{21} and 2.0×10^{20} Ar/m^2 s) for RES at 1980 K did not clearly affect the characteristics of surface morphology. The dimension of surface structure roughly scales with the mean eroded depth (not with the irradiation fluence because of the decrease of the RES yield with the flux).

Acknowledgements

This work was supported partly by a Grant-in-Aid from the Ministry of Education, Culture and Science, Japan and partly by the Program of Collaborative Study, National Institute for Fusion Science.

References

[1] W. Eckstein, J. Bohdansky, J. Roth, Nucl. Fusion (Suppl. 1) (1991) 51.
[2] T.K. Chini, S.R. Bhattacharyya, D. Ghose, D. Basu, Nucl. Instr. and Meth. B 72 (1992) 355.
[3] E.G. Wintucky, A.N. Curren, J.S. Sovey, Thin Solid Films 84 (1981) 161.
[4] J. Kawata, K. Ohya, J. Plasma Fusion Res. 70 (1994) 84.
[5] J. Kawata, K. Ohya, J. Plasma Fusion Res. 70 (1994) 1298.
[6] M.P. Seah, C. Lea, Thin Solid Films 81 (1981) 257.
[7] G.S. Bales, R. Bruinsma, E.A. Eklund, R.P.U. Karunasiri, J. Rudnick, A. Zangwill, Science 249 (1990) 264.
[8] Y. Ohtsuka, M. Isobe, K. Nakano, Y. Ueda, S. Goto, M. Nishikawa, J. Nucl. Mater. 220–222 (1995) 886.
[9] Y. Ueda, K. Nakano, Y. Ohtsuka, M. Isobe, S. Goto, M. Nishikawa, J. Nucl. Mater. 227 (1996) 251.
[10] Y. Ueda, Y. Ohtsuka, M. Isobe, M. Nishikawa, in: Proceedings of the 16th International Conference on Fusion Energy, Montreal, Vol. 3 (1997) p. 525.
[11] Y. Ohtsuka, J. Ohashi, Y. Ueda, M. Isobe, M. Nishikawa, J. Nucl. Sci. Technol. 34 (1997) 792.
[12] Y. Ueda, K. Shiota, Y. Kitamura, Y. Ohtsuka, M. Isobe, M. Nishikawa, Fusion Eng. and Design (to be published).
[13] W. Eckstein, V. Philipps, Physical sputtering and radiation-enhanced sublimation, in: W. Hofer, J. Roth (Eds.), Physical Processes of the Interaction of Fusion Plasmas with Solids, Academic Press, San Diego, 1996, p. 93.
[14] J. Roth, W. Möller, Nucl. Instr. and Meth. B 7/8 (1985) 788.
[15] V. Philipps, E. Vietzke, H. Trinkaus, J. Nucl. Mater. 179–181 (1991) 25.
[16] M. Nishikawa, Y. Ueda, S. Goto, Fusion Eng. Des. 16 (1991) 351.
[17] S.M. Rossnagel, Effects of surface impurities and diffusion on ion bombardment-induced topography formation, in: G. Kiriakidis, G. Carter, J.L. Whitton (Eds.), Erosion and Growth of Solids Stimulated by Atom and Ion Beams, Nijhoff, Dordrecht, 1986, p. 181.
[18] Y. Hoshi, A. Suzuki, J.H. Judy, Jpn. J. Appl. Phys. 33 (1994) 4991.
[19] B.A. Kalin, V.L. Yakushin, V.I. Polsky, Y.S. Virgilev, J. Nucl. Mater. 212–215 (1994) 1206.
[20] R.B. Wright, R. Varma, D.M. Gruen, J. Nucl. Mater. 63 (1976) 415.
[21] N. Kangai, T. Tanabe, J. Nucl. Mater. 220–222 (1995) 776.