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Simulation studies on sputtering and reflection from compound materials at elevated temperatures

T. Kenmotsu [a,*], T. Kawamura [b], Zhijie Li [b,1], T. Ono [c], Y. Yamamura [c]

[a] *The Graduate University for Advanced Studies, 322-6 Oroshi-cho, Toki-shi, Gifu 509-5292, Japan*
[b] *National Institute for Fusion Science, 322-6 Oroshi-cho, Toki-shi, Gifu 509-5292, Japan*
[c] *Okayama University of Science, Ridai-cho, Okayama, 700-0005, Japan*

Abstract

Using the ACAT code, we have calculated the energy spectra of boronized graphite under D^+ ion bombardment. In the case of light ion sputtering, high energy tail of the energy spectra drop sharply compared with the Thompson formula because most sputtered atoms are not due to collision cascade. In this work, we derived a new fitting formula based on the Falcon–Sigmund model instead of the Thompson formula. This fitting formula is in good agreement with the energy spectra in the high energy part. Furthermore, we have simulated surface compositional change in the Hirooka experiment under D^+ ion bombardment at high temperature. We have applied the ACAT-DIFFUSE code to calculate the compositional change of a boronized graphite. The ACAT-DIFFUSE is a simulation code based on a Monte Carlo method with a binary collision approximation and solves diffusion equations. The ACAT-DIFFUSE was developed to estimate chemical reaction such as methane production. In the present work, we obtained the result which has about 20% in surface composition change.

Keywords: Chemical erosion; Diffusion; High temperature; Fitting formula

1. Introduction

Graphite and carbon compounds are extensively used as plasma-facing materials in present day fusion devices. Plasma-facing components such as divertors are eroded by the high-energy plasma particles. Sputtering is a major contribution of material erosion. Secondary particles sputtered from the first wall penetrate into core plasma as impurities. In this case the energy spectra is an important parameter. The energy spectra of sputtered atoms and the reflection of incident ions are calculated using the ACAT code [1]. Sputtered atoms that are ejected after sufficient collisional processes usually obey the Thompson formula [2]. However, the energy spectra of sputtered atoms under light ion bombardment differ from the Thompson formula. Thus, it is necessary to have a different fitting formula for light ion sputtering. We tried to derive a new fitting formula based on the Falcon–Sigmund model [2].

It is known that methane, acetylene and hydrogen molecules are produced when graphite and carbon compounds are bombarded with hydrogen or deuterium ions. Hirooka et al. proposed bulk-boronized graphite to suppress chemical sputtering [3]. Several experiments are suggested to reduce chemical erosion lower than that of graphite [4]. Theoretical treatments of chemical reaction on graphite surface are studied by Balooch et al. [5] and Roth for graphite [6]. But, it is not sufficient to discuss chemical sputtering theoretically and experimentally for graphite and carbon compounds. To use these materials for plasma-facing components, it is necessary to obtain more data about chemical sputtering of candidate materials. We tried to follow the Hirooka experiments on compositional change near the surface of bulk-boronized graphite at high temperature (550°C).

* Corresponding author. Tel.: +81 572 58 2222; fax: +81 572 58 2628; e-mail: kenmotsu@nifs.ac.jp

[1] Present address: Inner Mongolia Teacher's College for Nationalities, Huolinhe street 22, Tongliao, 028243, Peoples Republic of China

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For this purpose, we have applied the ACAT-DIFFUSE code [7,8] to study chemical sputtering from bulk-boronized graphite.

2. Simulation model

A detailed description of the ACAT-DIFFUSE code is given elsewhere. Only a very brief outline is presented here. The code assumes an amorphous target material and it is based on the binary collision approximation. In the calculation, the total dose Φ is divided into small dose increments $\Delta\Phi$ during which the bombarding ions do not change the target composition appreciably. The ions corresponding to $\Delta\Phi$ are assumed to hit the target material simultaneously and are slowed down instantaneously. Their slowing down together with the associated vacancy and range distributions is simulated by the ACAT routine. These collided atoms diffuse during the time interval of $\Delta\Phi/J$ (J being the incident ion current density). The diffusion process is estimated by solving the diffusion equations numerically in the DIFFUSE routine. In the code, these procedures are repeated in times, where $n = \Phi/\Delta\Phi$. The logical representation of the code is ACAT-DIFFUSE = ((ACAT)(DIFFUSE))^n.

3. Chemical reaction model

When graphite and compounds that contain carbon atoms are bombarded with hydrogen or deuterium ions, methane, acetylene and hydrogen molecules are produced. These chemical reaction models on the graphite surface were proposed by Balooch and Olander [5]. In these models, the Arrhenius behavior is assumed to these chemical reactions. In that model, the rate equation at the topmost layer is as follows:

$$\frac{\mathrm{d}n_{\mathrm{D}}}{\mathrm{d}t} = J_{\mathrm{D}} - 2(K_2^e + K_1^2 k_2)n_{\mathrm{D}}^2 - 4K_1K_2k_3n_{\mathrm{D}}^3 + \mathrm{D}\left(\frac{\partial c_{\mathrm{D}}}{\partial x}\right)_{x=0}. \quad (1)$$

Here n_{D} is the topmost layer concentration cm^{-2} of deuterium due to D ion bombardment. c_{D} is the density of deuterium (cm^{-3}). The definitions of coefficients (K_1, K_2, K_2^e, k_2, k_3) in Eq. (1) refer to Ref. [5]. The first term on the right-hand side of Eq. (1) represents the rate of adsorption of D$^+$ ions per unit surface area from the primary beam. The second term represent the loss of deuterium by the combined effects of recombination and acetylene formation. The third term represents methane production. The last term represents diffusion into and out of the bulk solid. Each chemical reaction rate is characterized by a pre-exponential factor and activation energy assuming Arrhenius behavior. Then, methane reaction rates are estimated by the following equation:

$$K_1K_2k_3 = A_1 \exp(-E_{\mathrm{CH_4}}/k_{\mathrm{B}}T), \quad (2)$$

where A_1 is constant, $E_{\mathrm{CH_4}}$ is activation energy of methane production and k_{B} is the Boltzmann constant. These values for graphite are given by Balooch and Olander [5].

At 550°C, methane production is considered to be the dominant reaction, then, acetylene and molecule production are not taken into account at this temperature. Then, we estimate the carbon erosion yield from the surface due to methane production by the following equation

$$Y = \frac{1}{J_{\mathrm{D}}}K_1K_2k_3n_{\mathrm{D}}^3, \quad (3)$$

where J_{D} is the incident deuterium flux into the target. In the ACAT-DIFFUSE, it is assumed that one carbon atom is sputtered for every methane production.

4. Results and discussion

We used 2.5×10^{17} ions/cm^2/s as the D$^+$ current density and the target temperature was set to be 550°C. Ion bombarding energy is 250 eV. Target of bulk-boronized graphite contains 20% boron atoms. The simulations were performed under the same condition as the Hirooka experiment. The respective activation energies of C, B and D are assumed to be 0.88, 0.87 and 1.2 eV for the interstitial diffusion. The vacancy diffusion is considered indirectly by solving the rate equation of trapped atoms concentration, where the detrapping energies of C, B and D are set to be 2.51, 2.50 and 2.50 eV, respectively. In addition, we assumed that vacancy and range distributions do not change during one layer removal. In the case of high incident energy (250 eV), this assumption seems to be valid, because boronized graphite does not represent apparent composition change due to deuterium ion beam bombardment.

Fig. 1 shows the energy spectra of sputtered B and C atoms from boronized graphite with 250 eV D$^+$ ion bombardment at normal incidence. Light ions do not give rise to sufficient collision cascade. Almost all sputtered atoms by light ions are primary knocked-off recoil atoms. The high-energy tails of the energy spectra by light ion drop sharply compared with the Thompson formula. The possible maximum energy of a sputtered atom at normal incidence will be roughly estimated by $(1 - \gamma)\gamma E$, where γ is the elastic energy transfer factor [9]. In this case, the value of $(1 - \gamma)\gamma E$ of sputtered B and C atoms are 62.29 and 62.48 eV, respectively. Each possible maximum energy is nearly equal to the threshold of each energy spectra. Therefore, we derived a new fitting formula of energy spectra for light ion sputtering based on the Falcon–Sigmund model. In this formula, possible maximum energy of sputtered atom is taken

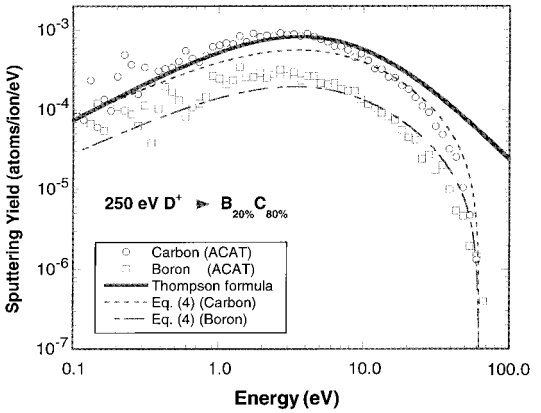

Fig. 1. Energy spectra of sputtered B and C atoms from boronized graphite with 250 eV D^+ ion bombardment at normal incidence.

into account. This formula is represented by the following expression

$$Y\,\mathrm{d}E \propto \frac{E}{(E+U_i)^2}\ln\left(\frac{T^i_{\max}}{(E+U_i)}\right)\mathrm{d}E \qquad (4)$$

where E is energy of sputtered atom, U_i is surface binding energy for element i of the compound material [10], and $T^i_{\max}$ is possible maximum energy of sputtered atom for element i of the compound material.

In Figs. 1 and 2, this formula is shown together with computer simulation data and the Thompson formula. Figs. 1 and 2 show a little difference between simulation values and a new fitting for the low-energy part (below 10 eV) of energy spectra, because this formula was fitted near the threshold energy part of the energy spectra. By this formula, threshold for energy spectra with bombarded light ion sputtering can be estimated.

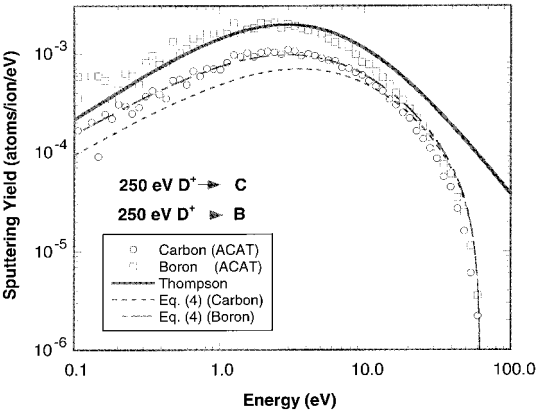

Fig. 2. Energy spectra of sputtered B and C atoms from monoatomic carbon and monoatomic boron with 250 eV D^+ ion bombardment at normal incidence.

Average energy (eV) of the reflected atom and number reflection coefficient of deuterium on the boronized graphite surface are shown in Fig. 3. The average energy of reflected atom increases with increasing incident energy. The number reflection coefficient decreases with increasing incident energy. There is no apparent difference between monoatomic target and compound material in energy and number reflection coefficient, because mass ratio between carbon and boron is not too large.

Fig. 4 shows the fluence dependence of the change of boron and carbon concentrations at the topmost layer. Two cases are calculated. One is the case under kinetic mechanism alone. The other case is under both kinetic

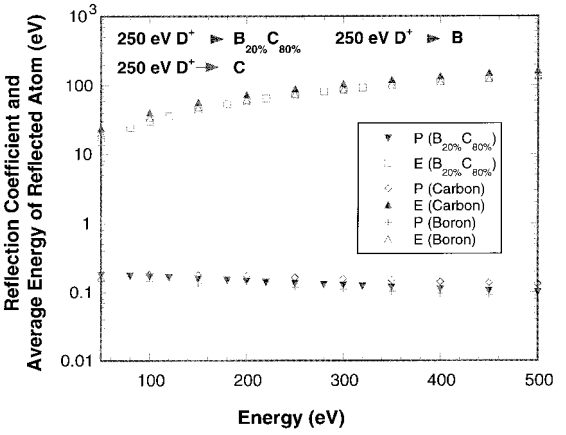

Fig. 3. Average energy of reflected atom and number reflection coefficient of deuterium on the boronized graphite surface and monoatomic target with 250 eV D^+ ion bombardment at normal incidence.

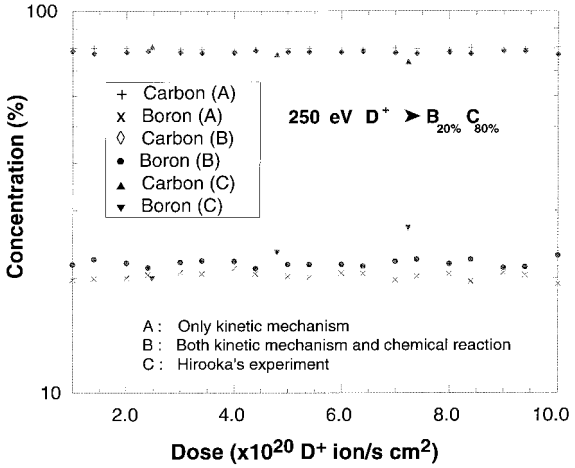

Fig. 4. The fluence dependence of the change of boron and carbon concentrations on boronized graphite surface with 250 eV D^+ ion bombardment at 550°C.

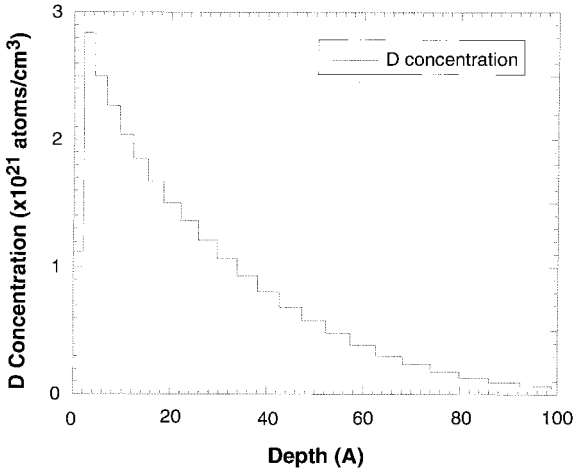

Fig. 5. The retention profile of deuterium in boronized graphite at fluence of 10^{21} D^+ ions/cm^2.

mechanism and chemical reaction. When boron carbide (or B_4C) is bombarded with ion beam, boron atom is sputtered preferentially due to the difference of surface binding energy. However, boron does not show significant preferential sputtering with light ion bombardment at higher energy [9]. Therefore, in Fig. 4 bulk-boronized graphite does not show apparent compositional change near the surface under light ion bombardment. However, Hirooka experiment showed that boron is slowly enriched to ~30% as the fluence approaches the order of 10^{21} D^+ ions/cm^2 at 550°C. It is found that this enrichment is attributed to the preferential chemical sputtering of boron. We tried to follow this chemical effect by the ACAT-DIFFUSE. In the ACAT-DIFFUSE, chemical reaction is estimate by Eq. (1). The values used for A_1 and CH_4 in Eq. (2) are 1.0 and 3.785 eV, respectively. The result including chemical reaction shows that boron is enriched to ~23%, the fluence approaches the order of 10^{21} D^+ ions/cm^2. In a recent work, we obtained the result which is about 20% difference between experimental data and the ACAT-DIFFUSE data. More detailed study is required to explain this difference.

Fig. 5 shows retention of deuterium ions in boronized graphite at fluence of 10^{21} D^+ ions/cm^2. The concentration of deuterium at the topmost layer is depleted by methane formation. This deuterium distribution is attributed to deuterium trapped in interstitial or vacancy and to diffusion from bulk to surface.

5. Conclusion

A new fitting formula of energy spectra for light ion sputtering was derived based on the Falcon–Sigmund model. This formula well reproduces the high-energy part of the energy spectra.

Using the ACAT-DIFFUSE code, we simulated surface compositional change of boronized graphite with deuterium ion bombardment at 250 eV. We tried to follow the experimental result of compositional change of boronized graphite at high temperature (550°C). We obtained the result which has about 20% difference between experimental data and the ACAT-DIFFUSE data included chemical reaction. We need to study about chemical reaction in more detail.

References

[1] Y. Yamamura, Y. Mizuno, IPPJ-AM-40, Inst. Plasma Physics, Nagoya University, 1985.
[2] G. Falcone, P. Sigmund, Appl. Phys. 25 (1981) 307.
[3] Y. Hirooka et al., Fusion Tecnol. 19 (1991) 205.
[4] Y. Ohtuka, M. Isobe, K. Nakano, Y. Ueda, S. Goto, M. Nishikawa, J. Nucl. Mater. 222 (1995) 886.
[5] M. Balooch, D.R. Olander, J. Chem. Phys. 63 (1975) 4772.
[6] J. Roth, in: R. Behrish (ed.), Sputtering by Particle Bombardment II, Springer, Berlin, 1983, p. 91.
[7] Y. Yamamura, T. Kenmotsu, Rad. Eff. Defects. Solids 142 (1997) 385.
[8] Y. Yamamura, Nucl. Instrum. Meth. B 28 (1987) 17.
[9] T. Kenmotsu, T. Kawamura, T. Ono, Y. Yamamura NIFS-DATA-46, 1998.
[10] R. Kelly, Nucl. Instrum. Meth. B 149 (1978) 553.