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Influence of diffusion on W sputtering by carbon

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Abstract

This paper concerns with temperature dependence of tungsten (W) sputtering by carbon (C). Weight loss measurements of W samples during sputtering by 2.4 keV C ions at different temperatures in the range between 300 and 1000 K showed a strong temperature dependence. At a certain fluence, which depends on the temperature, the probe's weight starts to increase again. This is due to C enrichment in the surface during ion bombardment, which eventually leads to the formation of a pure C surface. This C film then shields the underlying W from further sputtering and further growth of this film increases the probe's weight. The growth of this film depends critically on the diffusion of C into the W bulk material since this removes C from the surface and thereby increases the W concentration and W erosion. This in turn results in an increased reflection of the incoming C ions. These two coupled mechanisms hinder the growth of the C film with the diffusion accounting for the temperature dependence of the weight loss. To verify this model, C depth profiles in W were determined by means of Rutherford backscattering (RBS) with He ions. To increase sensitivity, the increased cross-section at 5715 keV was used. Analysis of depth profiles measured in the temperature range between 300 and 1000 K show that relevant diffusion processes appear at approximately 800 K. By performing Boltzmann Matano analysis on the measured depth profiles, a concentration dependant diffusion coefficient for C diffusion in W was deduced. The weight loss measurements were simulated using a combination of the Monte Carlo program TRIDYN and the diffusion code PIDAT. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Carbon; Tungsten; Erosion

1. Introduction

The first wall of fusion devices is usually made from a combination of high (W, Mo) and low (C, Be) Z materials. The high Z materials feature a high sputtering threshold and are hardly eroded by the hydrogen plasma. This is important since their maximum tolerable concentration in the plasma is low. On the other hand the low Z materials have a relatively high acceptable concentration in the plasma but are subject to physical and some also to chemical sputtering (C). Therefore, impurity ions are present in the plasma and the rest of the vessel is bombarded by energetic low Z ions. Their threshold energy for sputtering is low and they can also sputter the high Z materials.

Earlier work already dealt with the temperature dependence of C sputtering [1,2] and with the sputtering of W by C at room temperature and various angles of incidence [3]. This paper deals with the sputtering of W by C at elevated temperatures and normal angle of incidence. We will shortly present the results of weight loss simulations using the combination of TRIDYN [4] and PIDAT [5] which was already published in detail in [6]. These simulations showed that the diffusion data for C in W is difficult to apply to the diffusion of implanted C. We believe that this is because the literature data was measured using small C concentrations usually below the solid solubility (~1% in the temperature range treated here) of C in W. In the case of implanted C, we have to deal with concentration ranging from 0% to 100 %, i.e., from diffusion of C in W to self-diffusion of C in C. Therefore, a concentration dependant diffusion coefficient D(C) is proposed to describe the diffusion of C in W. The shape of D(C) is determined by applying

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Boltzmann Matano analysis to the depth profiles measured with Rutherford backscattering (RBS).

2. Experiment

The detailed experimental set-up for the weight loss measurements is described in [7]. They were performed on the Garching high current ion source. Here, CO_2 is used in the source to produce carbon ions which are then separated from other species using a sector magnet. The target fluence is measured using a current integrator. By applying a small positive voltage to the target, secondary electrons were suppressed. The beam current is typically 10^{-5} A with a beam spot of approximately 0.3 cm^2 and the pressure in the chamber during measurements is in the 10^{-7} mbar region. To clean the samples, they were heated to about 1073 K prior to every measurement.

For the weight loss experiments, polished W samples (99.95% purity) were irradiated with 2.4 keV $^{12}C^{+}$ ions under normal incidence. This energy was chosen because in fusion experiments, the multiple charged impurity ions are accelerated in the sheath potential thereby reaching energies in the lower keV region.

The weight change due to erosion and implantation of C was measured after successive fluence steps in situ using a Mettler ME21 microbalance which has a weight uncertainty of about 1 μg.

To investigate the C layer growth during implantation, we repeated the weight loss experiments removing the target from the ion source after each implantation step and measuring the C depth profiles with RBS. This was done on the 3 MV tandem accelerator using 5.7 MeV ^{4}He ions. There is an elastic resonance $C(\alpha, \alpha)$ at this energy which results in a cross-section about 120 times higher compared to the Rutherford case. This allows us to see the C peak on top of the W substrate spectra. The depth profiles were extracted from the measured RBS data using SIMNRA [8]. By iteratively changing the input depth profile for the simulation, the one that fits the experimental data best was chosen by hand. For the simulation of the RBS spectra, the measured cross-sections from [12] were used.

To determine D(C), two experiments were performed using amorphous carbon samples ($12 \times 15 \times 1$ mm^3) which were evaporated with a W layer. In the first experiment, the W layer was 100 nm thick and a single sample was heated during four successive heating steps at 773, 973, 1073 and 1173 K for 16 h each. In the second experiment, two targets were used with a 150 nm W layer. One was heated in successive steps for a total time of 16 h at 773 K and the other was heated at 973 K for a total time of 8.5 h. In both experiments, the C and W depth profiles were measured with RBS after each heating step. Using 2 MeV ^{3}He in the first case and 1.5 MeV ^{7}Li in the second experiment.

Prior to the heating experiments, the impurity concentrations in the W evaporated C samples were determined using XPS. It showed that the main impurity was oxygen with a bulk concentration of about 2% for the 100 nm samples and about 8% for the 150 nm samples. The temperature was measured using an infrared pyrometer. The emissivity was obtained by comparison to an optical pyrometer at temperatures above 973 K. Changes in the emissivity due to changes in the carbon concentration in the W layer result in a temperature uncertainty of about 20%.

3. Results and discussion

3.1. Weight loss measurements

The weight loss experiments were performed at 273, 773, 973 and 1073 K. The change in the probe's weight was measured after successive fluence steps of about 2.6×10^{17} cm^{-2} each. The results are displayed in Fig. 1. Initially the probe's weight decreases due to the erosion of W by C. At the same time, C is also implanted into the W bulk. As the erosion continues, this implanted C profile eventually reaches the surface. This results in a relatively lower relative W concentration at the surface, which reduces the reflection coefficient of the incident C ions. This together with the fact that the sputter yield for C at normal incidence is smaller than unity results in the growth of C layer on the surface. Therefore, from a fluence Q_C on the samples weight starts to increase again. The experiments performed at 273 and 773 K clearly show that behavior. For higher temperatures, the Q_C fluence increases strongly and the increase of the probes weight for fluences greater than Q_C is weaker.

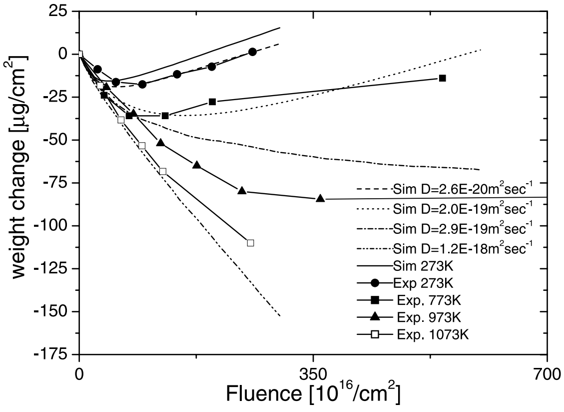

Fig. 1. Comparison between experimental weight change versus fluence and computer simulations using diffusion coefficients for five different temperatures with Q and D_o taken from literature [9].

For 1073 K, no weight gain at all was found in the observed fluence region.

One possible explanation for the temperature dependence can be the diffusion of C into the W bulk limiting the growth of the C layer during implantation.

At elevated temperatures, the implanted C diffuses into the bulk which results in lower C – and therefore higher W – concentration at the surface compared to the room temperature where we can neglect diffusion. This increased W concentration leads to a higher reflection coefficient. Both the removal of carbon from the surface by diffusion and the increased reflection hinders the formation of a C layer on the surface.

Therefore, the critical fluence Q_C for the beginning of layer growth is higher than for room temperature. The reason for the slower increase of the probes weight at high fluences is yet under discussion. A possible explanation would be the temperature dependence of the C self-sputtering yield. In [2], an increase of the sputtering yield for C on C is observed. For temperatures higher than 1200 K, it even becomes greater than unity. This could also explain the fact that no weight gain at all was observed for 1073 K.

To simulate the weight loss, the programs TRIDYN and PIDAT were combined [6]. TRIDYN is a Monte Carlo program that simulates the collision processes that occur during ion implantation thereby taking changes in the target composition into account. PIDAT simulates the diffusion of implanted species. It was developed to study the release, trapping and permeation of hydrogen implanted in materials. It simulates Fickian diffusion and additionally takes trapping and detrapping effects into account. In Fig. 1, a comparison of simulated and measured weight loss data is shown. The diffusion coefficient $D(\mathrm{C}) = D_o \exp(Q/k_\mathrm{B}T)$ was taken to be independent of the concentration and literature values [9] were used for Q and D_o. The simulations reproduce the experimental behavior but for the diffusion coefficients deviating from literature values (Fig. 2). As mentioned in Section 1, we attribute to the fact that the literature data was obtained for very small concentrations. Therefore, it cannot be applied to the our case in which C concentrations much higher than the solid solubility of C in W arise.

3.2. RBS depth profiles of implanted carbon

Diffusion effects should also be evident in depth profiles of C implanted into W. For implantation at 973 and 1073 K, the results are shown in Fig. 3. The depth profiles at 1073 K show a flatter slope compared to the 973 K profiles, indicating the diffusion of C. At both temperatures, there is no layer growth up to a fluence of 3.8×10^{18} cm^{-2} which corresponds quite well to the weight loss data in Fig. 1. At a fluence of 5.9×10^{18} cm^{-2}, there is strong layer growth on the 973 K sample in comparison to the 1073 K sample. On the latter, layer growth becomes visible not until fluences around 8.7×10^{18} cm^{-2}.

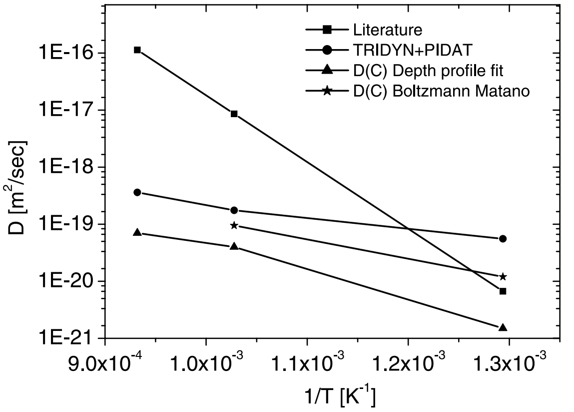

Fig. 2. Comparison of diffusion coefficients obtained from TRIDYN and PIDAT simulations and literature values. Also the D(C) values for low C concentrations obtained by the two experiments are shown.

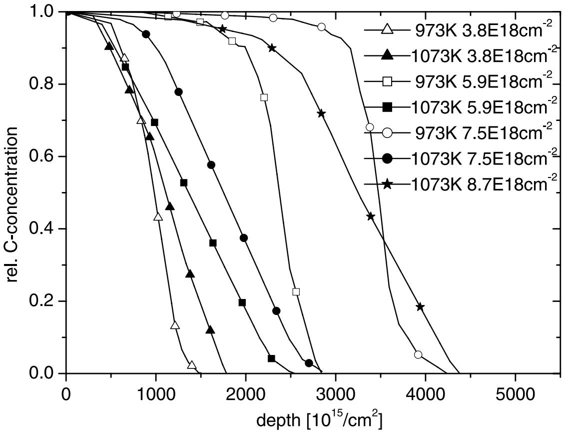

Fig. 3. RBS measurements of C layer growth during implantation. Equal symbols correspond to the same implantation fluence. Open symbols correspond to 973 K, closed ones to 1073 K.

These depth profiles support our model of diffusion limited layer growth to be the reason for the temperature dependence of the weight loss measurements. Layer growth sets in once large areas of the W substrate are covered with a C layer. Diffusion at higher temperatures enhances the fluence necessary for coverage of the surface.

3.3. Modeling the diffusion of high C concentrations in W

The diffusion of C in W is modeled by solving the general one dimensional diffusion Eq. (1) using a finite difference algorithm [10]. The diffusion coefficient is taken to be dependant on the C concentration

$$\frac{\partial \mathrm{C}(x,t)}{\partial t}=\frac{\partial D(x,t)}{\partial x}\frac{\partial \mathrm{C}(x,t)}{\partial x}+D(\mathrm{C}(x,t))\frac{\partial \mathrm{C}(x,t)^2}{\partial x^2}. \quad (1)$$

As explained above, two experiments were performed to determine the shape of $D(\mathrm{C}(x,t)) \equiv D(\mathrm{C})$. In the first experiment, the measured depth profiles from W evaporated C samples were simulated with D(C) that was based on the following assumption: for low concentrations, i.e., diffusion of C in W, D(C) is large compared to high concentration equivalent to diffusion of C in C. In Fig. 4, the results of the simulations and the dependence of the diffusion coefficient on the carbon concentration are shown. The simulations reproduce the measured profiles very well and the values of D(C) used are in the same order of magnitude (10^{-16} cm^2 s^{-1}) as literature values.

In the second experiment, D(C) was determined directly from the measured depth profiles by means of the 'Boltzmann Matano' method [11]. This allowed us to avoid the ambiguity in the choice of D(C). Boltzmann Matano method briefly works as follows: by defining a new variable $\eta = x/t^{1/2}$ and by assuming that $C(x,t) \equiv C(\eta)$, the diffusion equation can be written as

$$-\frac{\eta}{2}\frac{\mathrm{dC}}{\mathrm{d}\eta}=\frac{\mathrm{d}}{\mathrm{d}\eta}\left(D\frac{\mathrm{dC}}{\mathrm{d}\eta}\right). \quad (2)$$

Under the following boundary conditions:

$$\mathrm{C}=\mathrm{C}_0 \quad \text{for } x<0,\ t=0 \Longleftrightarrow \mathrm{C}=\mathrm{C}_0 \text{ for } \eta=-\infty,$$
$$\mathrm{C}=0 \quad \text{for } x>0,\ t=0 \Longleftrightarrow \mathrm{C}=\mathrm{C}_0 \text{ for } \eta=+\infty$$

and by integrating Eq. (2) once, one gets the following expression:

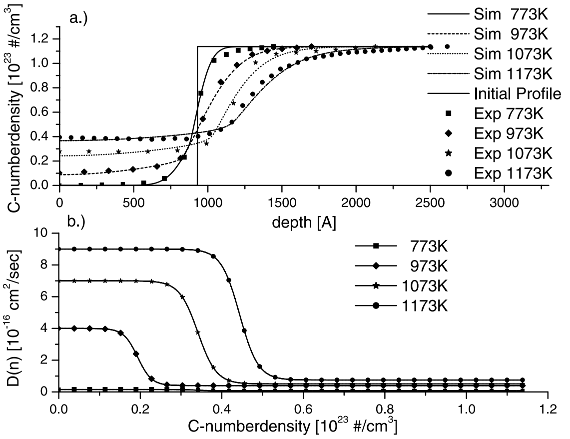

Fig. 4. Comparison between measured and experimental depth profiles (a), D(C) used to reproduce the depth profiles (b).

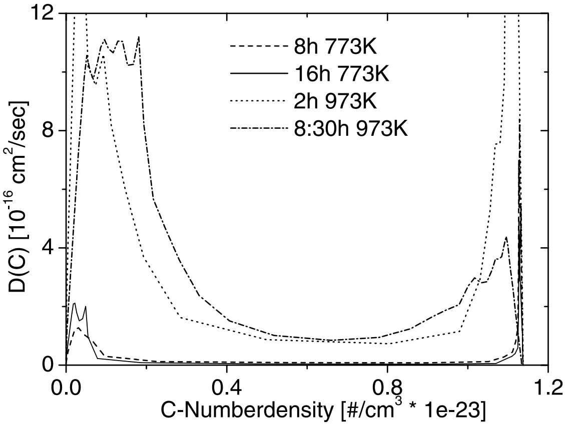

Fig. 5. D(C) values determined by applying the Boltzmann Matano method to measure depth profiles.

$$\int_{\mathrm{C}_1}^{\mathrm{C}_2} x(\mathrm{C})\,\mathrm{dC} = -2t\left\{D(\mathrm{C})\frac{\mathrm{dC}}{\mathrm{d}x}\bigg|_{\mathrm{C}_2} - D(\mathrm{C})\frac{\mathrm{dC}}{\mathrm{d}x}\bigg|_{\mathrm{C}_1}\right\}. \quad (3)$$

When $\mathrm{dC}/\mathrm{d}x|_{\mathrm{C2}} \equiv 0$, D at concentration $\overline{\mathrm{C}}$ can be written as

$$D(\overline{\mathrm{C}}) = \frac{1}{2\mathrm{t}(\mathrm{dC}/\mathrm{dx})|_{\overline{\mathrm{C}}}}\int_{\overline{\mathrm{C}}}^{\mathrm{C2}} \mathrm{x\,dC}. \quad (4)$$

Prior to evaluating Eq. (4) x(C) has to be determined such that

$$\int_{-\infty}^{\infty} x\,\mathrm{dC} \equiv 0. \quad (5)$$

The $x = 0$ position resulting from this requirement is called the Matano interface. Eq. (5) can also be understood as the requirement of conservation of mass. In deriving of D(C) from (4), the evaluation of the integral is the main error source especially for a small integration region, i.e., $\overline{\mathrm{C}} \approx \mathrm{C2}$.

The results of the heating experiments at 773 K after heating for 8 and 16 h and for 973 K after heating for 2 and 8.5 h are shown in Fig. 5.

They show similar diffusion coefficients as the ones obtained in the first heating experiment. The fact that they are higher by a factor of about 2.5 is probably due to errors in the temperatures which were measured using a pyrometer and contributes exponentially to D(C).

4. Conclusions

Our model of diffusion being the main reason for the temperature dependence in W sputtering by carbon is well supported by the computer simulations and layer-growth measurements. There is still a disagreement between computer simulation using TRIDYN and PIDAT and experimental weight loss data. We think that this is mainly due to lack of proper diffusion data for high C concentrations in W. The approach using the Boltzmann Matano analysis to directly determine *D*(C) from measured depth profiles has proven to be a promising method for determining this diffusion data.

In future experiments, we will investigate the possibility of a temperature dependence of C self-sputtering at temperatures below 1000 K which would also lead to weaker C layer growth.

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