Modeling tungsten and carbon sputtering by carbon at elevated temperatures

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Abstract. The planned use of carbon strike plates adjacent to tungsten tiles in the ITER divertor will lead to W erosion by sputtered C ions. The W sputtering yield by C decreases with increasing surface concentration of the implanted C. However with increasing temperature, the C self-sputtering yield increases, reducing the rate of C surface coverage. This study attempts to model the temperature dependent C self-sputtering process and its impact on W erosion by using modified surface binding energies (SBEs) in TRIDYN. The SBEs are benchmarked against C self-sputtering yields measured in this study and W irradiation by C is simulated and compared to previous experimental results. Good agreement between simulation and experiments is seen at 670 K, while small deviations are observed at 770 K due to surface roughening. At 870 K, significant deviation is observed which can be attributed to enhanced surface roughening as well as diffusion effects.

1. Introduction

The planned use of both carbon (C) and tungsten (W) as plasma facing materials (PFMs) in the ITER divertor [1] will lead to the presence of C impurities in the plasma due to erosion of the material, with subsequent impact on the surrounding W material. At typical plasma edge temperatures (10-100 eV), C ions occur mainly in higher ionization states. Multiply charged C ions incident at the W surface will be accelerated in the sheath potential and can therefore impact with energies in the lower keV region. The ASDEX-Upgrade tungsten divertor experiments have shown that W sputtering is dominated by such C impurities originating from erosion of graphite PFMs in the main chamber [2].

Tungsten erosion by C ions has been studied previously via weight loss measurements at RT [3] and elevated temperatures [4, 5]. The W sputter yield was seen to decrease with increasing C concentration at the surface, and subsequent formation of a closed C layer shielded the underlying W from further sputtering. Carbon diffusion resulted in changes to the implanted C depth profiles at $T > 900K$ [4] resulting in increased W sputtering by reducing the rate of C surface buildup. In addition, enhanced sputtering yield of carbon with increasing temperature occurs for all ions [5, 6, 7] due to weakly bound or sublimation of carbon interstitials formed during irradiation [6, 8]. Such enhanced C sputtering with increasing temperature plays an important role in determining the surface C concentration in tungsten irradiated by carbon.

This study presents the results of C self-sputtering by 6 keV C ions in the previously unexamined temperature range of $670 - 870K$ to benchmark the surface binding energies (SBEs) in TRIDYN [9] simulations, which critically govern sputtering yields. Although this temperature range is higher than the estimated operating temperature of $400 - 500K$ [1] at the ITER divertor (excluding strike points), the SBE required for ITER relevant temperature can be interpolated from the benchmarked SBEs at the higher temperatures. The benchmarked SBEs were also used to model the fluence dependent W sputtering and C implantation by 6 keV C ions at $670K$, $770K$, and $870K$ [10] which previously could not be fitted using the SBEs provided in Ref. [5]. Discrepancies between experimental results and TRIDYN simulations are discussed in the framework of both increased surface roughness from the annealing process and C layer growth.

2. Experiments

For the C self-sputtering experiments performed in this study, 250 nm thick amorphous C films were deposited on polished W substrates using a magnetron sputter device. X-ray diffraction analysis showed that the deposited C film structure was amorphous and annealing the C specimens at $870K$ for 4 h resulted in no significant changes in the diffractogram. Therefore the C film structure can be assumed to be thermally stable during irradiation at elevated temperatures.

The experimental details of the apparatus are described in Ref. [11] and only a short description follows. Specimens were irradiated with 12 keV C_2^- ions at an incident angle $\alpha = 15^o$ and the C_2^- ions are assumed to dissociate on target impact with resulting energy of 6 keV per C atom. The diameter of the C beam was 3-4 mm and the beam profile and fluence was measured using a beam viewing system calibrated against current measurements [10]. The specimens were heated using a ceramic heater and the temperature measured using a two wavelength pyrometer calibrated against thermocouple measurements. In-situ ion beam analysis with 2.5 MeV ^{3}He ions was performed between each irradiation step. Changes in C areal density

was measured by nuclear reaction analysis (NRA) and the amount of C implanted was determined by subtracting the initially measured C areal density (i.e. virgin C-film) from the C areal density measured following each irradiation step. The experimental details of the tungsten sputtering experiments as well as the atomic force microscope used to measure the root mean square roughness of specimens can be found in Ref. [10].

3. Results and discussion

3.1. C self-sputtering experiments

The increase in the amount of implanted C against incident C fluence for carbon films irradiated by 6 keV C ions in the temperature range of $670-870K$ is shown in figure 1. The slope in figure 1 equals $1-R_c^c-Y_c^c$, where R_c^c is the reflection yield of C on C, and Y_c^c is the C self-sputtering yield. TRIDYN calculations show R_c^c is roughly two orders of magnitude smaller than Y_c^c and can therefore be assumed zero. The slope in figure 1 is then equal to $1-Y_c^c$, and the linear fit indicates that the C self-sputtering yield is smaller than unity and constant throughout irradiation at all temperatures. The resulting values of Y_c^c determined from the linear fits in figure 1 are listed in table 1.

The measured yields are in good agreement at $T \le 670K$ when compared to the exponential fit of Ref. [5] (see dashed curve in figure 2). However, at $T > 670K$, the measured yield is larger than the exponential fit of Ref. [5] and this difference is attributed to the combined effect of varying carbon structure of the specimens (amorphous vs. pyrolytic graphite) and the incident angle used (15° vs. normal) between the different experiments. Also given in figure 2 is the best fit using the present data which requires an additional linear term. This best fit is not meant to be physically meaningful but to illustrate that the spread in the interpolated value of the C self sputtering yield using either functions is within the experimental error in determining the C self sputtering yields. Interpolation to ITER relevant temperature of $400K$ using this best fit and the fit from Ref. [5] gives a C self sputtering yield of 0.33 and 0.30, respectively.

To account for the temperature dependent C self-sputtering behavior in TRIDYN, the value of the surface binding energy of carbon ($SBE_c^{'}$) can be reduced. In order to determine the

$SBE_c^{'}$ values based on the measured experimental C self-sputtering yields, a scan varying

$SBE_c^{'}$ in TRIDYN was done to determine the dependence of Y_c^c. An exponentially decreasing

function (1) was fitted to the scan results:

$$SBE_c^{'}(Y_c^c) = 1.4 + 13.7 \exp(-Y_c^c / 0.3) \tag{1}$$

According to (1), $SBE_c^{'}$ of 4.3, 3.8, and 3.1 eV are required to correctly model the measured

values of Y_c^c at temperatures $670K$, $770K$, and $870K$. At ITER relevant temperature

$400K$, a $SBE_c^{'}$ of 6.2 ± 0.2 eV is required.

For a system involving more than one element, the effective surface binding energy of each target component can be chosen in dependence of the actual surface composition in TRIDYN using (2):

$$SBE_i = \sum_{j=1}^{NCP} SBV_{ij} \cdot c_j \tag{2}$$

where NCP is the number of components, SBV_{ij} is the surface binding energy of a surface atom i to a target atom j, and c_j is the surface atomic fraction of a target atom j $(1 \le j \le NCP; \sum c_j = 1)$. For the two component W-C system, the SBV_{ij} was obtained by

averaging the surface binding energy of pure W ($SBE_w^{'}$) and C ($SBE_c^{'}$) materials using (3):

$$SBV_{ij} = \frac{1}{2}(SBE_w^{'} + SBE_c^{'}) \tag{3}$$

For all TRIDYN calculations, the surface binding energy of W to W atom was taken as

$SBV_{ww} = SBE_w^{'} = 8.68 eV$ from the heat of sublimation value, while the surface binding energy

of W to C atom was taken as $SBV_{wc} = 8.0 eV$ from the average of $SBE_w^{'}$ and $SBE_c^{'}$ values

at RT using (3). Physically, this represents the assumption that tungsten sputtering is independent of temperature and tungsten carbide formation. The surface binding energy of C to

C atom ($SBV_{cc} = SBE_c^{'}$) at different temperatures was determined using (1), while the surface

binding energy of C to W atom was determined using (3). A summary of the SBV_{ij} values used in the TRIDYN calculations are listed in table 1.

3.2. W irradiation by C ions and comparison to TRIDYN simulations

The experimental results of W irradiation by 6 keV C ions at $RT - 870K$ [10] are now compared to corresponding TRIDYN calculations using the SBV_{ij} values in table 1 (see figure 3). The experimental trend with increasing temperature is a decrease in the C implantation rate with a corresponding increase in the amount of W sputtered. This decreasing trend of the C implantation rate with increasing temperature is well modeled by the increase in C self-sputtering yields implemented in TRIDYN simulations. Specifically, the good agreement at $670K$ between experimental data and simulation indicates that the method of reducing the carbon surface binding energies in TRIDYN can model the changes in the C implantation and W sputtering behavior from increased C self-sputtering. Higher C self-sputtering yields are therefore responsible for the shift observed in experiments to higher C fluences to form a closed C layer that shields the W from further sputtering. However, at higher temperatures, a larger incident angle in TRIDYN simulations, $\alpha_t > \alpha = 15^o$, is required to best fit the experimental data. At $770K$, the experimental data is best described using $\alpha_t = 20^o$, while at $870K$, the implanted C behavior is best described using $\alpha_t = 25^o$, although large deviations in the W sputtering behavior is still observed. The need for larger incident angles with increasing temperature can be explained by the increase in surface roughness due to the heating process and rough C layer growth. Rough surface results in a local distribution of angles altering the sputter yields due to the angular dependency of physical sputtering.

The root mean square roughness (R_q) value measured using AFM increases from $R_q = 4 \pm 1nm$ for an unannealed virgin W specimen to $R_q = 8 \pm 1nm$ for specimens annealed at $770K$ and $870K$ [10]. No increase in R_q value was observed for the annealed specimen at $670K$. Previously in Ref. [10], it was assumed that surface roughness effects could be neglected for the case of 6 keV C irradiation of tungsten where the mean C ion range (10-15nm in pure W) was comparable to R_q of the annealed specimen. The use of only a slightly larger $\alpha_t = 20^o$, to fit the experimental data at $770K$ supports this initial assumption. The agreement between TRIDYN simulations and experimental data at $T \leq 770K$ is within 10-20 % and validates the approach of using modified C-C surface binding energies in TRIDYN to model the experimentally observed dynamics of C implantation and W sputtering. However, the use of higher incident angles to fit the experimental result at $870K$ is not fully justified from the initial surface roughness measurements. Although, the growth of a much rougher C layer at

$870K$ ($R_q = 48 \pm 5nm$ [10]) compared to C layers formed at lower temperatures ($R_q = 20 \pm 1nm$) can provide the qualitative argument to increase the incident angle to $\alpha_t = 25^{\mathbf{o}}$, the possible onset of C surface diffusion and its contribution in reducing the C surface coverage by surface morphology changes cannot be completely ruled out. Unfortunately, with the available diagnostics the contribution of both surface roughness and diffusion processes at $870K$ could not be separated.

4. Conclusions

To summarize, experimental C self-sputtering yields at $670K$, $770K$, and $870K$ were measured by irradiation of C films using 6 keV C ions and used to benchmark the surface binding energies used in TRIDYN simulations. These SBEs were used to simulate previously published [10] experimental results of 6 keV C irradiation of W at the same temperatures. For $T \leq 770K$, the TRIDYN model of reducing the carbon SBEs can describe within 10-20%, the increased W sputtering and decreased C implantation rate from increased C self-sputtering with temperature. The use of a larger incident angle, $\alpha_t = 20^{\mathbf{o}}$ in TRIDYN to best model the experimental data at $770K$ can be explained by the increase in surface roughness from the annealing process. However, at $870K$, the use of an even larger incident angle, $\alpha_t = 25^{\mathbf{o}}$, indicates significant contribution from surface roughness resulting in different dynamics from a smooth 1-D plane as compared to a realistic rough surface. Also, the possible onset of C surface diffusion and its contribution in decreasing the C surface coverage by surface morphology changes could not be quantified.

In conclusion, in the temperature range $T \leq 770K$, TRIDYN can model within 10-20% the C implantation and W sputtering behavior using experimentally benchmarked C-C surface binding energies. A $SBE_c^{'} = 6.2 \pm 0.2eV$ can be used as a benchmarked input parameter in present day simulation codes utilizing TRIDYN to better model the plasma wall interactions of mixed C-W materials at the ITER relevant temperature of $400K$.

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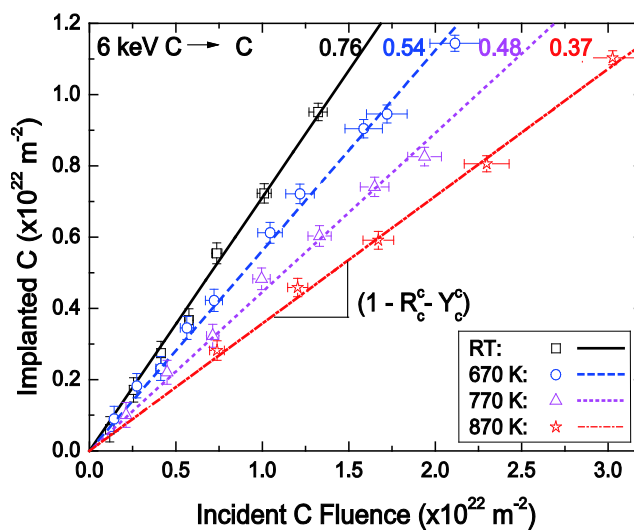[11] Bizyukov I, and Krieger K 2006 *Rev. Sci. Instrum.* **77** 043501

Figure 1. Experimentally measured increase of the implanted C amount as a function of incident C fluence at $RT - 870K$ for C films irradiated by 6 keV C ions. The implanted C amount was determined by NRA following each irradiation step and subtracting the initial C areal density of the virgin specimen. Also plotted are the linear fits with their corresponding slopes indicated by the numbers.

Table 1. Experimentally measured C self-sputtering yields and the SBV_{ij} in eV used in all TRIDYN calculations to simulate the increased C self-sputtering with temperature.

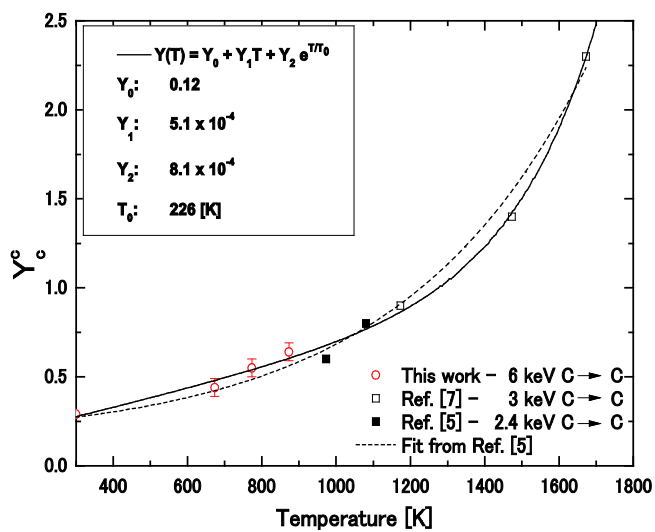| Temp	Slope	Y_c^c	SBV_{cc}	SBV_{cw}	SBV_{wc}	SBV_{ww}
RT	0.76 ± 0.01	0.24 ± 0.01	7.4	8.0	8.0	8.68
670 K	0.54 ± 0.03	0.46 ± 0.03	4.3	6.5	8.0	8.68
770 K	0.48 ± 0.03	0.52 ± 0.03	3.8	6.3	8.0	8.68
870 K	0.37 ± 0.05	0.63 ± 0.03	3.1	5.9	8.0	8.68

Figure 2. Comparison of the C self-sputtering yields plotted against published literature data in Refs. [5,7] along with the best fit to all the data.
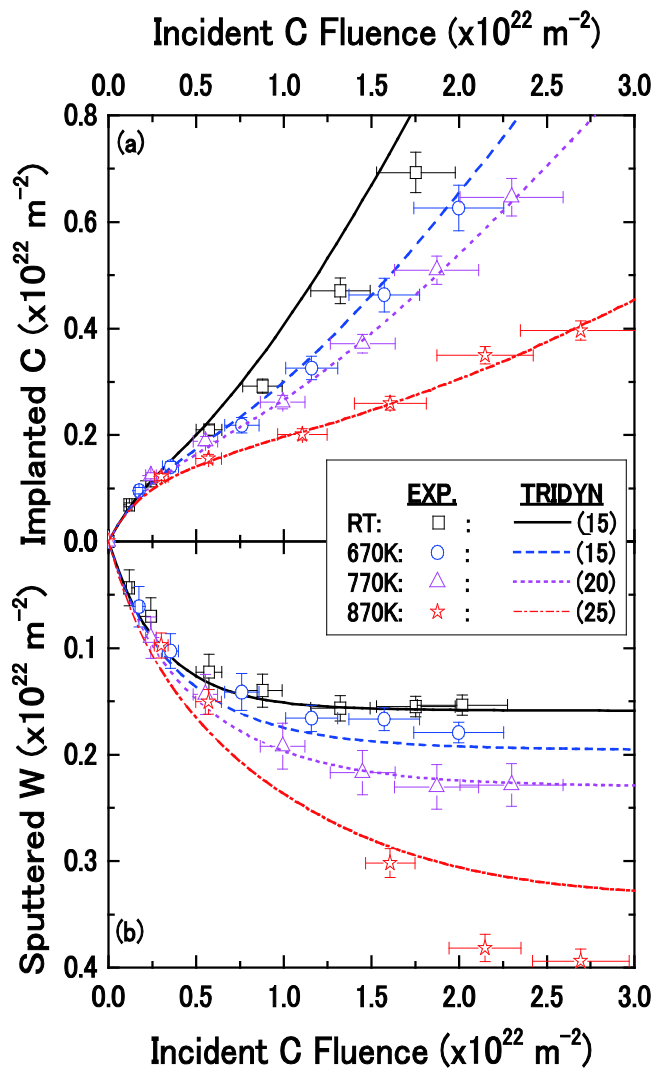
Figure 3. Comparison of the experimentally measured changes of (a) implanted C and (b) sputtered W as a function of incident C fluence of W films irradiated by 6 keV C ions at $RT - 870K$ [10] with TRIDYN simulations using the SBV_{ij} from table 1. The bracketed numbers in the legend indicate the angle of incidence, α_t in degrees, that was used in TRIDYN simulations.

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Figure 2. Comparison of the C self-sputtering yields plotted against published literature data in Refs. [5,7] along with the best fit to all the data.

Figure 3. Comparison of the experimentally measured changes of (a) implanted C and (b) sputtered W as a function of incident C fluence of W films irradiated by 6 keV C ions at $RT-870K$ [10] with TRIDYN simulations using the SBV_{ij} from table 1. The bracketed numbers in the legend indicate the angle of incidence, α_t in degrees, that was used in TRIDYN simulations.

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