

Temperature dependence of the erosion behaviour of deuterium beam exposed tungsten-doped carbon films (a-C:W)

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Abstract

Tungsten-doped amorphous carbon films (a-C:W) are used as model system to study the deuterium (D) retention behavior and the erosion behavior of metal containing co-deposited layers. In our recent studies, the implantation temperature was increased towards an ITER relevant range. Pure and 7.5 at% tungsten-doped carbon films have been exposed to a deuterium beam of 200 eV/D at different implantation temperatures up to 1300 K and at a fluence of 10^{24} D/m². Rutherford backscattering spectrometry with a 1.5 MeV ⁺H beam was performed to obtain the total erosion yield. For each implantation condition and for each structure of doped films, the total erosion yield is reduced significant, compared to pure films. The temperature dependence of the total erosion yield is less pronounced for doped films.

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1 Introduction

The use of carbon materials together with metallic plasma-facing materials (PFMs) in fusion devices - as planned for ITER with beryllium (Be), tungsten (W), and carbon (C) - will lead to deposition of mixed carbon-metal layers during sputtering/erosion cycles of PFMs by hydrogen [1]. Therefore, the chemical erosion behavior of such mixed layers needs to be investigated as well as their hydrogen retention behavior (tritium inventory)[2].

Our main attempt is to gain basic knowledge about the interaction between the impacting deuterium (D) beam and the mixed layers, e.g. the D retention processes and the erosion behavior. Therefore, a well characterized model system for mixed materials is currently investigated using certain experimental conditions of D beam exposure (energy, flux of incident D beam, specimen temperature) [3]. In general, our studies are restricted to tungsten-carbon mixtures with W concentration below 20%, which allows a quite limited extrapolation to ITER.

Investigation of D retention and C erosion behavior was studied at the different parameters. On one side, parameters of the D beam exposure were varied, e.g. the implantation temperature between 300 K and 1300 K. On the other side, the structure of the films was varied, producing a-C:W with different dopant concentrations below 15% W and applying a different pre-heating temperature (700 K, 1300 K); specimen were pre-heated before D beam exposure. The implantation temperature was different. W concentration and pre-heating temperature lead to changes in the structure of the carbon matrix [4] as well as to a slight formation of carbide phases around 1300 K [5]. Varying the implantation temperature, the structure was held fixed (dopant concentration: 7.5% W, pre-heating temperature: 1300 K). Varying the structure, the parameters of the D beam exposure were held fixed (300 K, 200 eV/D).

In our previous studies [6], measurements of the deuterium retention were performed varying the structure of a-C:W films. Now we focus on the erosion behavior of a-C:W films varying the exposure parameters and the structure of the films. Therefore, results already published [3,7,6]

were extended.

2 Experimental

2.1 Specimens

Investigating the influence of the film structure on the erosion behavior, the same specimens that had been exposed to the D beam investigating the D retention [6], were used in this study. They were produced by magnetron sputter deposition on Si (100) wafers. Details of this production process are described in [6].

Film thickness and tungsten concentrations were determined by Rutherford backscattering (RBS) with 4 MeV ^4He . These values were averaged over the whole specimen area with an accuracy of 10% and 5%, respectively. The film thickness varied between 0.6 and 1.2 μm , among different batches. Tungsten concentrations are given in atomic percent throughout the paper.

At higher implantation temperatures, specimens were produced and analysed by the same method as described above. W concentrations were held fixed at 7.5%. Each sample was pre-heated to 1300 K.

2.2 Deuterium beam exposure and analysis of the film thickness of eroded specimens

Specimens with deposited a-C:W films were exposed to a D beam in the Garching high current ion source facility at temperatures between 300 K and 1300 K, at a fluence of 10^{24} D/m². A mono energetic (3600 eV per D_3^+) and mass-separated D_3^+ -ion beam with a flux of 10^{19} D/m²s was used. In the incident D beam two particle species simultaneously exist, D_3^+ -ions (about 90%) and neutrals (about 10%). The ions are decelerated to 600 eV per D_3^+ by a positive bias of 3000 V applied to the target. The neutrals, however, are not decelerated by the bias voltage.

This leads to an implantation energy of 200 eV/D for the ions and of 1200 eV/D for the neutrals. The beam spot profile of the 200 eV particles was analysed in [6]. The area of the erosion spot is limited to a size of about 70 mm². A reference zone was defined, resulting in a calibration factor of about $3.3 \cdot 10^{23}$ D/m²C for the accumulated charge in each experiment. In contrast, the beam spot profile of the 1200 eV particles is extended over the whole specimen area (see section 2.4). The specimen temperature was controlled by electron beam bombardment from the rear side and optical pyrometry with an accuracy of ± 50 K.

The total amount of C and W atoms per unit area in the eroded specimen was analysed by Rutherford backscattering spectrometry (RBS) with 1.5 MeV protons under a scattering angle of 165° at the tandem accelerator facility. At this analyzing energy, the information depth is larger than 1 μ m. The analysing spot size was 1x1 mm².

In the erosion experiment and in the analysing facility the positioning of the specimens on the specimen holder and the positioning of the manipulator was performed in an analogous manner as described in [6]. Therefore, it was possible to hit the reference zone with the RBS beam while scanning over the specimen across the erosion spot. Data points generated on a part of the scan overlapping with the reference zone were averaged, resulting in an averaged amount of atoms per unit area. The accuracy of this average value is about 16%.

2.3 Data evaluation: The total erosion yield

For doped carbon materials, the erosion yield strongly depends on the fluence of incident D [8]. Several attempts evaluating an in-situ fluence dependence can be found in literature, e.g. mass spectroscopy, optical emission spectroscopy and weight loss measurements [9,10,8]. In this study the erosion behavior was analysed ex-situ, after D beam exposure at a fluence of 10^{24} D/m². The total erosion yield was obtained from the ratio between the total amount of eroded C and W atoms per unit area and the total amount of incident D particles per unit area. In this context the total erosion yield is defined as an average value over a fluence of 10^{24} D/m², evaluated over the reference zone. Results already published [3,7] are extended.

The total amount of eroded C and W atoms per unit area was obtained from the difference between the total amounts of C and W atoms per unit area in the a-C:W film, before and after D beam exposure. Both values were evaluated by integration of the C and the W peak in the energy spectrum of the backscattered particles (e.g. ^4He for the initial specimen and ^1H for the eroded specimen).

The total amount of incident D particles per unit area is defined as fluence of incident D atoms. Therefore, the 200 eV beam spot profile was analysed in each campaign as described in [6] (one D beam exposure of an a-C:H film in each campaign, assuming a reproducible beam profile during one campaign). This leads to a calibration factor for the accumulated charge in each experiment as described in [6]. The fluence was obtained with an accuracy of about 16% then. With these estimations the accuracy of the total erosion yield is about 20%.

2.4 Estimation of the concentration of neutrals in the D beam

In order to estimate the concentration of neutrals in the D beam, a thin W layer of 30 nm thickness, deposited on silicon substrate from W, evaporated by electron beam, was exposed to the D beam with an accumulated charge of 5.2 C. D_3^+ -ions were accelerated with a bias of 3600 V. The target was biased with 3000 V. As the sputtering threshold energy of W for D bombardment is 211.4 eV [11], the W layer is sputtered exclusively by the 1200 eV neutrals.

RBS with ^4He at 0.5 MeV was performed to obtain the amount of W atoms per unit area, both before and after D beam exposure. The total amount of eroded W atoms was obtained from the difference between these two values.

Before D beam exposure, the RBS analysing beam was scanned along both diagonals of the specimen. With the obtained data, initial thickness values were calculated over the whole specimen area. After the D beam exposure, thickness values were measured over the whole specimen area, applying a two dimensional scan with a rectangular grid of 1 mm step width.

Fig. 1 shows the 1200 eV erosion spot. The spot is extended over the whole specimen area and shows a flat slope: Eroded thickness values at zones near the edge (0.5 nm) increase slightly

towards the center of the specimen (4 nm).

The fluence of neutrals at an unit area is the product of the sputter yield of W (about $6.4 \cdot 10^{-3}$ at 1 keV/D and normal incidence [12]) and the averaged amount of sputtered W atoms per unit area. The concentration of neutrals at an unit area is obtained out of the ratio between the fluence of neutrals and the total fluence at this unit area. The concentration of neutrals is about 10% at the area of the 200 eV spot and about 5% at the reference zone. However, at zones near the edge of the specimen that are not affected by the D_3^+ -ion beam [6], the concentration of neutrals is 100%.

Similar investigations were performed in [13], measuring the weight loss of a gold target at 60 eV deuterium impact energy (180 eV D_3^+ beam). The concentration of neutrals at the area of the 60 eV spot was about 3%. The corresponding concentration of neutrals in this study (3600 eV D_3^+ beam) is about 10%. Comparing these two results shows that concentration of neutrals in the D beam depends on the energy of the D_3^+ -ions. Further on, it needs to be examined whether the concentration of neutrals in the D beam changes significantly with time.

3 Results and discussion

3.1 Influence of the film structure on the total erosion yield: D beam exposure at 300 K

The structure of the carbon matrix as well as the formation of W-C phases can be controlled by doping and pre-heating of the films [4]. D beam exposure experiments at 300 K intended to investigate the influence of doping and the influence of variations of dopant concentrations as well as pre-heating temperatures on the total erosion yield.

Figure 2a) shows the total erosion yield Y_{tot} of pure and W-doped films for different pre-heating temperatures and for different W concentrations up to 15% after D beam exposure at a fluence

of $10^{24}\text{D}/\text{m}^2$.

The total erosion yield is insensitive on pre-annealing, for pure, 7.5% W-doped and 15% W-doped films, i.e. insensitive on changes in chemical bondings inside the carbon matrix.

For the doped films the total erosion yield is reduced between a half and two thirds, compared to the pure films. Varying the W concentration an unexpected maximum occurs at 6% of W. In literature, the shielding effect is the most widely used explanation for a reduction of the total erosion yield after doping [14,15,9]. In this context the total erosion yield is assumed to decrease monotonically with increasing dopant concentration [10]. This, however, does not explain the observed maximum at 6% in this study.

In the Küppers cycle [16] the erosion yield of pure carbons decreases for temperatures above 600 K. The formation of volatile hydrocarbons is suppressed as the activation energy for the hydrogen release is exceeded. Changed chemical bondings in the near vicinity of W clusters and in W-C-crystallites, might result in an altered activation energy for the hydrogen release in the Küppers cycle. This effect might depend on the W concentration inside the film. This interpretation needs to be verified in future studies.

3.2 Temperature dependence of the total erosion yield: D beam exposure at temperatures up to 1300 K

In order to observe the temperature dependence of the erosion mechanisms in W-doped films, the structure of the films was held fixed. Each film was pre-heated at a temperature of 1300 K. The W concentration was fixed at 7.5%.

Figure 2b) shows the total erosion yield Y_{tot} after D beam exposure at a fluence of $10^{24}\text{D}/\text{m}^2$ and at different temperatures between 300 K and 1300 K.

At an implantation temperature of 700 K, doping with 7.5% W significantly reduces the total erosion yield compared to pure C (about a factor 20). The temperature dependence of the total erosion yield of 7.5% W-doped films shows no significant change between 300 K and 700 K. At

900 K the total erosion yield is increased by a factor of two. The total erosion yield is roughly constant above 900 K.

In general, the temperature dependence of the total erosion yield is less pronounced for the doped films, compared to the pure films.

In literature it was shown that metal-doping leads to a significant reduction of the total erosion yield [3,7], at fluences up to 10^{24} D/m² and at implantation temperatures of 300 K and of 650 - 750 K. This is in agreement with the results presented in this study. At fluences below $5 \cdot 10^{22}$ D/m² it was observed that metal-doped films show a strong temperature dependence of the CD₄ production yield [3]. In this study, however, the total erosion yield was analysed at a fluence of 10^{24} D/m². The evaluation of the temperature dependence of the total erosion yield Y_{tot} at fluences up to $5 \cdot 10^{22}$ D/m² will be an interesting future aspect.

The W enrichment at the surface, which is building up with increasing fluence, influences the amount of D getting implanted, trapped and released (as pure D or as erosion species). Therefore, it affects the fluence dependence of the erosion yield and of the D retention. Further studies are intended to examine the fluence dependence of the W enrichment.

4 Summary

Investigating the C erosion behavior parameter studies were performed. On the one side, the implantation temperature was varied between 300 K and 1300 K. On the other side, the structure of the films was varied, producing a-C:W films with different dopant concentrations ($\leq 15\%$ W) and applying different pre-heating temperatures (700 K, 1300 K). Varying the implantation temperature, the structure was held fixed (dopant concentration: 7.5% W, pre-heating temperature: 1300 K). Varying the structure, the parameters of the D beam exposure were held fixed (300 K, 200 eV/D). For each implantation condition and for each structure of the doped film, the total erosion yield is reduced significantly, compared to the pure film. Varying the W concentration an unexpected maximum occurs at 6%. Additionally, the temperature dependence of

the total erosion yield is less pronounced for the doped films.

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Figure captions

Fig. 1:

Two dimensional plot of the amount of sputtered W atoms in nm, sputtered by D neutrals in D ion beam with an accumulated charge of 5.2 C, obtained from RBS analysis with ^4He at 0.5 MeV. The spot is extended over the whole analysed area of the specimen of $14 \times 9 \text{ mm}^2$. Eroded thickness values show a flat slope from 0.5 nm to 4 nm.

Fig. 2):

Total erosion yield Y_{tot} analysed by RBS with ^1H at 1.5 MeV on pure and W-doped C-films implanted with a D beam of 200 eV/D up to a fluence of 10^{24} D/m^2 . The accuracy of the data is about 20%. a) Implantation temperature: 300 K. b) Implantation temperatures: 300 K - 1300 K. The W concentration and the pre-heating temperature were fixed at 7.5% and 1300 K, respectively.

figures

Fig. 1

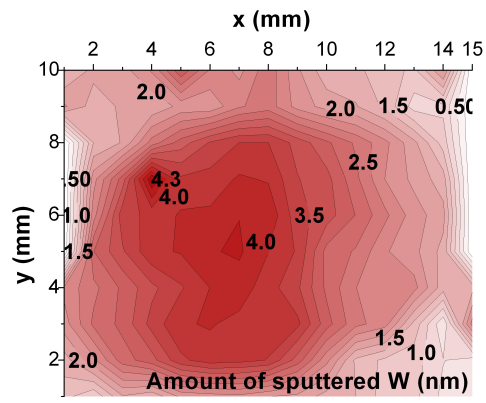


Fig. 2a)

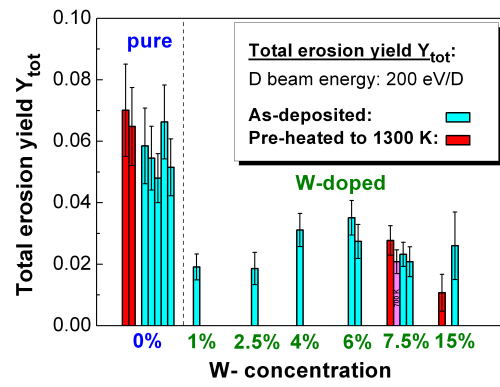


Fig. 2b)

