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Wall conditioning by microwave generated plasmas in a toroidal magnetic field

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Abstract

The suitability of microwave generated plasmas for the purpose of wall coating in pure toroidal magnetic fields is investigated in a special test bench. We report on the results of layer deposition using methane plasmas as a systematic case study for future boron or silicon deposition by other gases (B_2H_6, SiH_4). The produced coatings can be characterized as polymer-like soft a-C:D-films with a high D/C-ratio due to the low energy of particles hitting the wall. Neutral hydrocarbon radicals could be identified to play the major role for film deposition. On the other hand, strong re-erosion induced by deuterium ions is observed in regions with plasma-wall contact. The spatial homogeneity and the characteristics of produced coatings are presented and observations are correlated with measured plasma parameters. The use of pulsed plasmas for wall conditioning is compared with steady-state discharges.

Keywords: a-C:D; Wall coating; ECR; Future fusion devices

1. Introduction

The application of superconducting field coils in future fusion devices challenges the development of new wall conditioning methods because the techniques used nowadays [1] like DC-discharges are incompatible with the semi-permanent magnetic fields. Using microwaves for creating process plasmas, e.g. with the electron-cyclotron-resonance (ECR), is a promising route.

The feasibility of ECR-discharges for wall conditioning has been shown in different devices (e.g. TEXTOR [2], ALCATOR C-Mod [3]). The low availability of fusion machines for such systematic studies causes the need for a special test device. For this purpose, a toroidal magnetic field system (TOMAS) is operated at the FZ-Jülich in collaboration with the Ruhr-Universität-Bochum.

The vessel geometry ($R = 0.78$ m, $a = 0.26$ m) is chosen half the size of the TEXTOR vacuum chamber while the variable toroidal B-field can reach a maximum value of 120 mT on the torus axis. A magnetron with fixed frequency ($f = 2.45$ GHz, 6 kW max) is used as microwave source. By variation of the coil current the resonant B-field of 87.6 mT for 2.45 GHz is achievable at any position of the minor diameter. The device can be operated steady state and is described in detail in [4].

The microwave generated discharge can be characterized as low temperature plasma ($T_e = 2$–12 eV, $T_i < 0.5$ eV, $T_{gas} \approx$ RT) with densities from typically 2×10^{10} up to 1.5×10^{11} cm^{-3}. As the wave energy is not only absorbed by ECR but also by upper-hybrid-resonance (UHR) [5,6], the shape of the radial plasma profiles shows increasing densities directed radially outward from the ECR-position (Fig. 1). This is superimposed on an outward oriented $E \times B$-drift where E is in vertical direction caused by charge separation in the simple toroidal magnetic field. The density decreases

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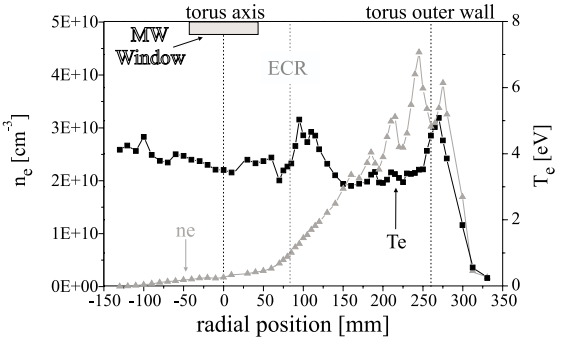

Fig. 1. Radial profiles of electron density (grey) and temperature (black) in an argon standard discharge (parameter given in the text). The plasma is not only produced by ECR but also by UHR mechanism in a wide region.

rapidly from the ECR layer towards the high field side of the vessel. The T_e-profile is smooth around 4 eV except for temperature peaks at the ECR position and at the plasma edge. The potential difference between plasma and wall is in the range of 5–15 V which leads to ion impact energies of the same range as no external electrical field is applied. This is different from wall conditioning by DC-discharges where sheath potentials of several hundred volts occur. We thus expect the formation of so-called 'soft' films [7].

2. Experimental

The following experiments on film deposition have been made by using purely deuterated methane (CD_4). As standard discharge parameters a neutral gas pressure of 2.5×10^{-2} Pa (flow 3.5 sccm), magnetic field of 97 mT at the torus axis and an input of 1200 W microwave power have been chosen. For the determination of the layer characteristics partially covered Si-samples were installed on four poloidal stainless steel strips each equipped with eight poloidally distributed samples. The steel strips were arranged clockwise at 80°, 170°, 260° and 350° toroidal position where the location of microwave input is defined as 0°. Samples were analyzed ex situ by ellipsometry, thermal desorption spectroscopy (TDS), ion beam analysis (IBA) and electron probe X-ray microanalysis (EPMA). For additional information about the thickness distribution the wall itself has been analyzed by colorimetric observations [8]. The change of the exhaust gas composition was investigated by a quadrupol mass analyzer (QMA).

To achieve reproducible initial conditions, the machine was cleaned before every deposition by ECR- or DC-glow-discharges in oxygen or hydrogen.

3. Results

3.1. Characterization of the layers and formation mechanism

The deposited films are polymer-like soft amorphous hydrocarbon films (a-C:D) with a carbon concentration of $3.5 \pm 0.4 \times 10^{22}$ cm^{-3}. This leads to a C mass density of 0.7 ± 0.08 g cm^{-3} which is slightly lower than published data [9] while the quantified D/C-ratio of 1.7 ± 0.2 is higher. The results for the optical properties, refraction index ($n = 1.55 \pm 0.03$) and extinction coefficient ($k = 0.0015 \pm 0.001$), are typical for polymer-like a-C:D films [9].

These layer properties show no systematic dependence neither on different sample positions nor on variation of the discharge parameters. Contrary to that, the film thickness distribution is sensitive to discharge conditions and strong spatial variations can be found on the vessel wall. In Fig. 2(a) and (b) such a distribution is shown after 5 h discharge at standard parameters but with factor of two higher gas pressure (5×10^{-2} Pa). The film thickness (given in nm) is measured on the Si-samples. Colored areas are indicating zones with deposited layers while white regions (bare metal wall) are erosion dominated. The formation of these erosion domains can be explained when the polygon shape of the vessel is taken into account (Fig. 2(c)). In combination with the magnetic field ripple (green) regions which are protruding in the plasma (red) and quasi-shaded sections (blue, vessel corners) alternate toroidally. Even for the poor magnetic confinement the radial transport of charged particles across the magnetic field lines into the corners is reduced. In the net-erosion regions layer formation is suppressed due to the high re-erosion by D-ions. It is obvious that these ions are formed in large concentration, because the D_2 content is enhanced in the gas phase as a result of the nearly complete methane decomposition during the steady-state discharge. It has been shown [9] that for low wall temperature ($T < 350$ K) 'ion-assisted chemical erosion' can lead to significant erosion rates even with low energetic hydrogen ions in the range of 20 eV. Thus deposited hydrocarbons are promptly re-eroded partly in form of radicals which can then stick again on the wall. The regions without layer deposition are poloidally limited by the intersection of the ECR-zone with the vacuum chamber, and can only be found outward directed from that (Fig. 2(b)). This is in good agreement with the measured plasma profiles (Fig. 1) which shows that the plasma density decreases rapidly inward directed from the ECR.

In regions with nearly negligible plasma density film removal and deposition occurs mainly by neutral particles. Layer formation in shadowed areas and on the inner wall parts is thus attributed to neutral hydrocarbon radicals.

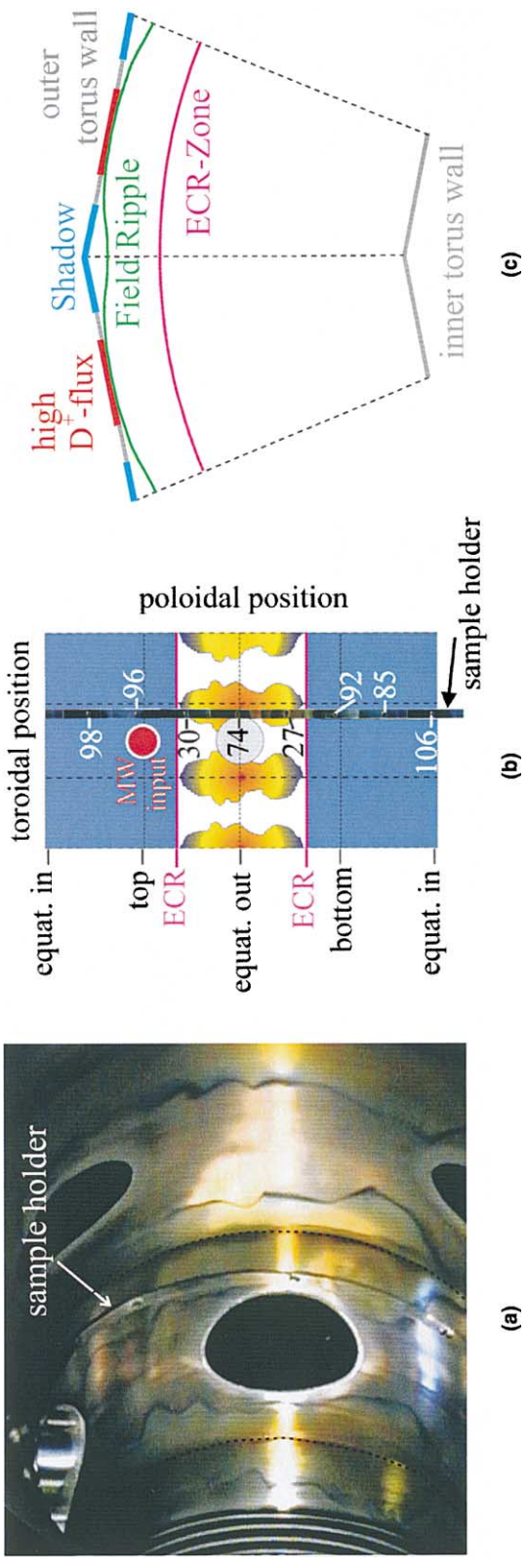

Fig. 2. (a) View on the coated torus wall. (b) Thickness distribution (nm) on the unfolded torus part. White areas: regions without layers, colored areas: net-deposition zones. (c) Influence of device geometry on film distribution. Zones of higher plasma-wall contact (red = erosion) alternate toroidally with shaded areas (blue = net-deposition) caused by polygon shape of the vessel and the magnetic field ripple.

Radicals reaching the wall may have enough reactivity to be chemisorbed without impact of energetic particles (ion induced sticking [9]). So far the concentration of the different radicals in the discharge has not been measured directly and it is not clear which of them has the highest contribution to the layer formation. The CD_3 radical, which is obviously produced by the decomposition of methane, has a very low sticking probability of 10^{-3} [10]. This could lead to its further dissociation in the plasma and, on the other hand, to the formation of larger molecules by neutral–neutral or ion–neutral reactions. Other hydrocarbons (e.g. C_2D_2) can be easily formed at the wall or in the gas phase and can be found in the residual gas during discharges in methane by QMA.

3.2. Heat treatment in vacuum

To assess the threshold for thermal layer-stability samples were exposed under vacuum of 2×10^{-6} Pa at temperature of 500 and 600 K. The released gaseous products were observed by a mass spectrometer. After exposure the films were analyzed by ellipsometry.

In the first case (500 K) the layer thickness was reduced by 5–10 nm after 12 h of heating. Only during the first 15 min a noticeable particle release could be observed which consists mainly of water and mass 44(CO_2/C_3D_4). The water obviously was absorbed by the film from atmospheric humidity during storage under air. No change of the optical layer properties could be found after heat exposure.

On the other hand, heat treatment at 600 K over 6 h removed a 200 nm layer nearly completely. Remaining coatings showed a slightly higher refraction index which might be affected by the small film thickness (15 nm).

The threshold of thermal stability between 500 and 600 K is in good agreement with thermal desorption spectra (Fig. 3) where large release of higher hydrocarbons starts at temperatures above 500 K with a maximum at 625 K, e.g. for mass 28 (C_2D_2 or CO). Correlation with other hydrocarbon masses and cracking products identify the main contribution as acetylene. The recorded mass spectra deviate slightly from published results (e.g. [11]) with a lower contribution of mass 20 (CD_4) and much higher signal of D_2. The observed deuterium is mainly the result of decomposed C_xD_y and not of bounded D_2 in the film. This leads to the conclusion that the layers consist of loosely bound

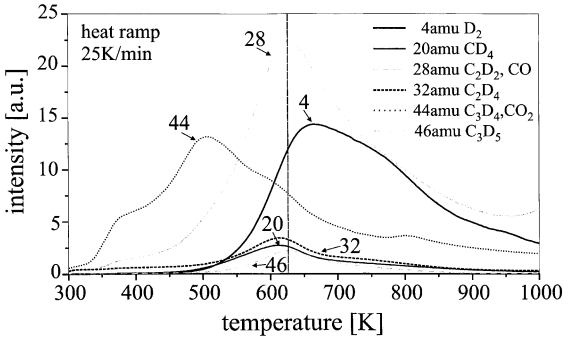

Fig. 3. Thermal desorption spectra of a polymer-like soft a-C:D-layer. Release of mass 44 at low temperature ($T <$ 500 K) is correlated with water release from the film surface. Above 500 K significant signals for higher hydrocarbons can be found with maxima around 625 K (e.g. for C_2D_2).

hydrocarbons which are released even at low temperatures (600 K).

3.3. Comparison of steady-state and pulsed wall coating

To improve the spatial homogeneity of the layer distribution one has to reduce the high re-erosion from D^+ ions which are produced as a consequence of the enrichment of D_2 in the gas phase. By using pulsed ECR-discharges with short pulses ($t = 100$ ms) and long off-periods in the range of the pump out time ($T = 9$ s) the concentration of deuterium in the gas phase is reduced close to the initial value between pulses.

The characteristic time of ion loss is in the order of the decay time of the plasma density which is in the range of 10^{-3} s confirmed by probe measurements. So re-erosion by D^+ stops nearly immediately at the end of a microwave pulse. As a result of the relatively low pumping speed, the radicals have a high chance to stick somewhere on the wall or to be converted into volatile products before being pumped out. As their sticking probability is inversely proportional to the number of wall collisions without being attached, the achieved homogeneity of spatial layer distribution should be higher for low sticking ($CD_3/10^{-3}$) than for high sticking radicals (e.g. $C_2D_2/0.8$) [10]. The achieved thickness distribution for a pulsed methane discharge (after 20 h, Fig. 4(b)) shows no erosion dominated zones as they have been found for 5 h steady-state discharges (Fig. 4(a)). For the pulsed case, the thickest layers are deposited at the equatorial outside with a toroidal maximum near the microwave source (0°). The toroidal thickness inhomogeneity is in good agreement with the measured toroidal plasma density distribution. The latter is caused by the limited propagation of the microwave and by the electron free-mean-path length. Together with the observed maximum at the equatorial outside this leads to the assumption that in pulsed plasmas a non-negligible effective sticking coefficient for the particles dominating the film growth has to be taken into account. It is likely that different mechanisms dominate the layer formation in these two types of discharges although the optical properties of the films are very similar in both cases. For the steady state case it is obvious that effective re-erosion leads to an accumulation of sticked hydrocarbons at areas without plasma contact.

Although it takes 20 h to achieve maximum layer thickness of 110 nm with the used pulsed microwave discharge, the deposition rate per single pulse is very high because the overall time plasma duration is less than 14 min (on/off = 1/90). This leads to a maximum deposition rate of nearly 8 nm per minute of plasma duration.

4. Conclusion

Using ECR-discharges in methane leads to the deposition of polymer-like soft a-C:D films with a high

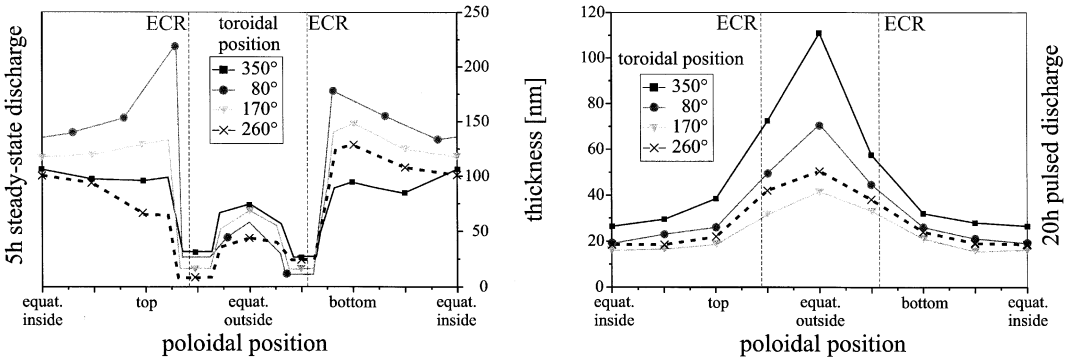

Fig. 4. Poloidal thickness distribution (Si-samples) at four toroidal positions. (a) After 5 h steady-state discharge: thickest films inward from the ECR/erosion at ECR. (b) After 20 h pulsed discharge: thickest layers directed outward from ECR/no significant re-erosion zones.

hydrogen content. The deposited layers have a low thermal stability and can easily be re-eroded by D-ions. The effects of erosion and redeposition lead to high spatial inhomogeneities in layer distribution which are strongly reduced in pulsed plasmas. To make pulsed microwave discharges a tool for wall coating more work on the optimization of the on/off-time is needed. Pulsed ECR-discharges in gases like B_2H_6 and SiH_4 are promising candidates for the production of fusion relevant surface coatings. This will also apply to pulsed ECR process plasmas in a confining stellarator field configuration.

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