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# Tokamak and laboratory modeling of hydrocarbon film deposition on metallic mirrors

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ABSTRACT

In this work, amorphous hydrocarbon (a-C:H) film deposition on metallic mirrors was studied during working shots in tokamak T-10 and at exposure in $Ar/CHD_3/D_2$ dc magnetron discharge in a special laboratory high vacuum setup. Analysis of film composition (including hydrogen content) was carried out using nuclear physical methods. Thickness and optical parameters (refractive and extinction coefficients) of the films were estimated by ellipsometry. Laboratory films can be characterized as soft a-C:H films in comparison with hard tokamak films ($\rho = 1.2$ and 1.8 g/cm$^3$, respectively). For the first one, a linear dependence of deposition rate on mirror temperature was observed in a wide temperature range. The addition of methane into initial $Ar/D_2$ magnetron gas mixture leads to an increase of deposition rate. The data obtained should be taken into account to prevent hydrocarbon film formation on the surface of first mirrors in ITER.



## 1. Introduction

Transport, deposition and redeposition of atomic carbon and hydrocarbon radicals in modern tokamaks and future ITER are the reason of amorphous hydrocarbon (a-C:H) film deposition on a surface of in-vessel elements. In particular, hydrocarbon film formation on the first mirrors of ITER optical and laser diagnostics can change drastically of optical characteristics of the mirrors [1,2]. Therefore, an investigation of the a-C:H film deposition at ITER-relevant conditions is necessary for the development of techniques for protection and cleaning of the mirrors [3,4]. In this work, the recent laboratory results on carbon film deposition on molybdenum and stainless steel (SS) mirrors exposed in deuterium magnetron discharge at ITER-relevant conditions are compared with data on growth of carbon films obtained in tokamak T-10.

## 2. Experimental

Hydrocarbon films were produced inside the tokamak T-10 (RRC 'Kurchatov Institute') during experimental campaign of 2002 year [3]. The 'limiter' poloidal section of T-10 with most intensive a-C:D film deposition was chosen for exposure of polished SS samples (SS316, $10 \times 10 \times 4$ mm$^3$). The mirrors were located in upper limiter port at distance of 22 cm from the plasma, within straight view of plasma and movable graphite limiter located at bottom of working chamber. The samples (three mirrors) were placed inside a metal box with a shutter to prevent film deposition during vacuum vessel conditioning. Thus, hydrocarbon films for investigation were produced during working discharges only. The typical parameters of the discharge were the following: working gas – $D_2$, electron temperature of core plasma – up to 1 keV, electron density – $(1–6) \times 10^{19}$ m$^{-3}$, ion temperature 450–700 eV. The exposure time was about 27 min (more than 1500 plasma discharges with 1 s duration). To our estimation the temperature of samples during plasma discharges was maintained at about 350 K. Most probably mechanism of hydrocarbon film formation was the following: during working pulses a graphite (MPG-8) limiter was subjected to intensive erosion as a result of bombardment by high flux of deuterium atoms; a fraction of eroded carbon atoms and hydrocarbon radicals was ionized in the plasma and drifted in the gradient of toroidal magnetic field in vertical direction. The ions with energy in the range of 10–100 eV could cross the border of the plasma column and reach the mirrors forming hydrocarbon films. Moreover, the mirrors were irradiated by change–exchange neutrals.

Hydrocarbon film deposition depending on mirror temperature was made in laboratory ITER-relevant conditions. A high vacuum ($3 \times 10^{-5}$ Pa) setup equipped by a direct current magnetron device with a graphite (MPG-8) cathode was used for experiments. Polished stainless steel (SS316) and polycrystalline molybdenum samples ($10 \times 10 \times 4$ mm$^3$) were inserted in a special holder combined with an ohmic heater. Mirror temperature was measured by a thermocouple in the temperature range 313–623 K. A part of sample surface was screened by a mask. After deposition the clean

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area was served as initial surface both in spectroscopic and ellipsometric measurements.

Working gas was previously prepared in a special gas balloon as a mixture of argon, methane ($CHD_3$) and deuterium ($D_2$). Note, that methane fraction in the initial gas mixture was equal to 5% and 35% with substitution of deuterium by methane. Composition of the gas mixture in the working chamber was monitored by a mass-spectrometer before and after each experiment at maximal level of pumping speed and maintained with an accuracy of 5–10%. Deposition experiments were carried out at the same operating pressure (about 3 Pa) varying gas input and pumping speed. In separate experiments at 363 K it has been shown that film thickness linearly depends on exposure time. It means that gas composition in the working chamber was not changed significantly during all time of deposition (2 h).

Basic parameters of the magnetron discharge were the following: pressure of working gas – 3 Pa, discharge current – 100 mA, cathode potential – 370 V. Electron temperature and density and ion energy of the plasma were about 10 eV and $2 \times 10^{16}\ m^{-3}$ and 260 eV, respectively. Physical sputtering of the graphic cathode by Ar ions resulted in generating (additionally to flux of methane decomposition products and hydrocarbon products of chemical erosion of carbon by deuterium) an atomic carbon flux ($\sim 1 \times 10^{18}\ m^{-2}s^{-1}$ with average energy of about 8 eV) on the mirror. The samples were placed at the distance of 5 cm against cathode center and have floating potential about −30 V. In all experiments the exposure time of mirrors in the discharge was 2 h.

After deposition in the tokamak T-10 and laboratory setup, the surface morphology of the samples was investigated by scanning electron microscopy. Rutherford backscattering coupled with resonance elastic scattering was used for analysis of deposit composition (including hydrogen content) [3]. The thickness and optical properties (refractive and extinction coefficients) of the films were measured by ellipsometry (manual ellipsometer Gaertner L119XUV, $\lambda = 632.8$ nm). Stylus profilometer (Tencor Instruments) was used to roughly estimate of deposited layer thickness. Relative reflectance of mirrors before and after exposition was measured by means of a specular reflectance accessory of Lambda 35 UV/Vis spectrophotometer (Perkin–Elmer).

## 3. Results and discussion

The dependence of film thickness on deposition temperature for three gas mixtures is presented in Fig. 1. It is seen that starting with certain temperature (e.g., 363 K for $Ar/D_2$ gas mixture) all curves can be described by linear dependence up to a critical temperature of transition from deposition to erosion. The temperature was equal to 460 K for $Ar/D_2$ gas mixture and 470 and 490 K for 5% and 35% methane in the gas mixture, respectively. Thus, the deposition rate linearly decreased with increasing temperature both for SS and Mo substrates. A partial substitution of $D_2$ by $CHD_3$ in the gas mixture leads to a marked increase of deposition rate and transition temperature. It means that neutral products of methane decomposition can give additional contribution into the film growth [5]. Supposing only one step reactions for neutral gas molecules in the magnetron plasma at our pressure, we can conclude that methyl radical ($CHD_2$) is most abundant species in the plasma. It is known that a sticking coefficient of methyl is increased from $10^{-5}$ to $10^{-3}$ at presence of atomic hydrogen [6]. Thus, additional flux of the radicals can be responsible for the enhanced deposition rate.

Note, that after reaching of minimal thickness (about 10 nm) between 450 and 500 K it did not change significantly at higher temperatures. In this connection the detailed ellipsometric analysis of the films deposited with using of 35% methane gas mixture

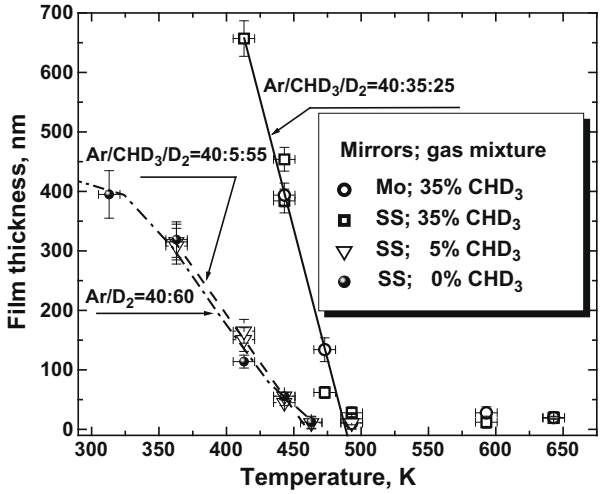


**Fig. 1.** Film thickness versus temperature of stainless steel and molybdenum mirrors for different working gas mixtures. Dash-dot, dash and solid curves correspond to gas mixtures $Ar/D_2 = 40:60$, $Ar/CHD_3/D_2 = 40:5:55$ and $Ar/CHD_3/D_2 = 40:35:25$, respectively.

was carried out (Fig. 2). It was shown that in the temperature range 363–493 K the refractive coefficient remained constant whereas extinction coefficient increased with temperature. The increase of the extinction coefficient means transformation (dehydrogenation) of $sp^3$ into $sp^2$ carbon groups [4,7]. At the temperature of transition from deposition to erosion (about 500 K) the reflectance index dropped sharply from 1.75 to 1.2 whereas the extinction coefficient was constant. To our opinion, above 500 K a specific thin hydrocarbon film on the mirror surface was formed.

The recent experiments on DIIID have shown that heating of Mo mirrors prevented a-C:H film deposition, but a decrease of total reflectivity was detected on the mirrors [8]. The latter was connected with surface oxidation of Mo in the absence of a-C:H film. The similar processes of oxide and carbide formation on the mirror surface can be responsible for degradation of optical properties also in our experimental conditions. Generally, a difference between optical parameters and deposition rate of a-C:H films on SS and Mo mirrors were not observed. However, Mo mirrors were more chemically stable at elevated temperatures (above 500 K).

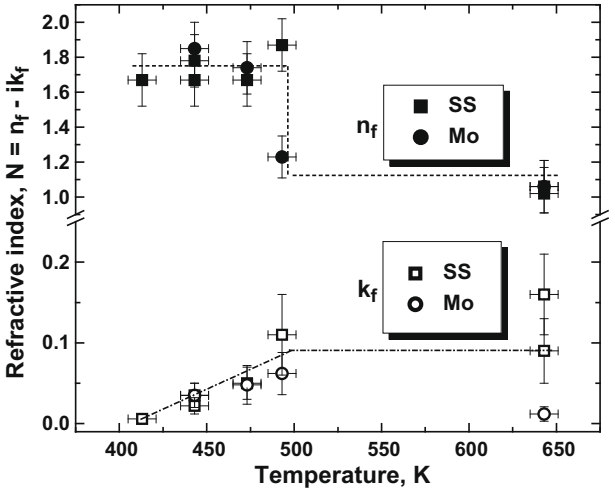


**Fig. 2.** The dependence of a-C:H film refractive ($n_f$) and extinction ($k_f$) coefficients on the deposition temperature for the gas mixture $Ar/CHD_3/D_2 = 40:35:25$. Solid and dash lines are guide for eyes only.

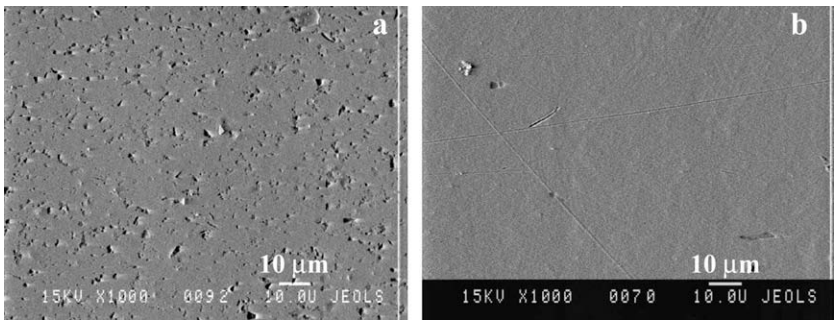


**Fig. 3.** (a, b). Surface morphology of a-C:H films on stainless steel mirrors exposed in the tokamak T-10 (a) and laboratory setup (b) at about 350 K.

Hard hydrocarbon film ($n_f = 2.1$, $k_f = 0.11$) deposited on single crystal silicon substrate and stable at elevated temperatures was used for investigation of film erosion. The film was exposed in the magnetron discharge at the gas mixture with 5% methane during 2 h at 500 K. Note that, the temperature is above the temperature of transition from deposition to erosion for the gas mixture (470 K). After the exposure color of the film was changed from gold to green. It can be a result of additional deposition instead of erosion. Indeed, ellipsometric analysis in a framework of one layer model shown that thickness of the film increased from 290 to 320 nm. However, the reflectance coefficient dropped down to 1.66 at about the same extinction coefficient ($k_f = 0.145$). It means that the film has a lower density compared with the initial film density (1.2 and 1.8 g/cm$^3$, respectively). To our opinion, the result can be explained by penetration of atomic hydrogen into the bulk of the hard film at the elevated temperatures and transformation of the film into soft hydrogen-saturated one.

Surface morphology of a-C:H films on SS mirrors exposed in the tokamak T-10 and the laboratory setup at about 350 K is presented in Fig. 3(a) and (b). In general, both films are smoothed. At the same time, the dust particles on the surface of the tokamak films were found (Fig. 3(a)). Some properties of the tokamak and laboratory films are summarized in Table 1. It is seen that the magnetron films can be characterized as more soft a-C:H films in comparison with quite hard tokamak films ($n_f = 1.7$ and 2.1, respectively).

Evolution of SS mirror reflectivity in laboratory deposition experiments for the gas mixture with 5% methane at different temperatures is depicted in Fig. 4. Note, that relative reflectance spectra of SS mirrors exposed with and without the magnetron discharge at 493 K are practically the same in the wavelength range of 400–1000 nm. It means that although heating of the mirrors is quite effective protective procedure, chemical stability of mirrors at elevated temperatures is subject for additional investigations.

**Table 1**
Some properties of a-C:H films deposited on stainless steel mirrors in the tokamak T-10 (sample 1) and laboratory setup at 350 K.

| Gas mixture composition | | $n_f$ | $k_f$ | H/H + C | H/C | $\rho$ (g/cm$^3$) |
|---|---|---|---|---|---|---|
| T-10 | $D_2$ | 2.1 | 0.1 | 0.35 | 0.55 | 1.8 |
| Lab | $Ar/D_2 = 40:60$ | 1.69 | 0.01 | 0.44 | 0.8 | 1.2 |
| | $Ar/CHD_3/D_2 = 40:5:55$ | 1.71 | 0.02 | | | |
| | $Ar/CHD_3/D_2 = 40:35:25$ | 1.67 | 0.006 | | | |

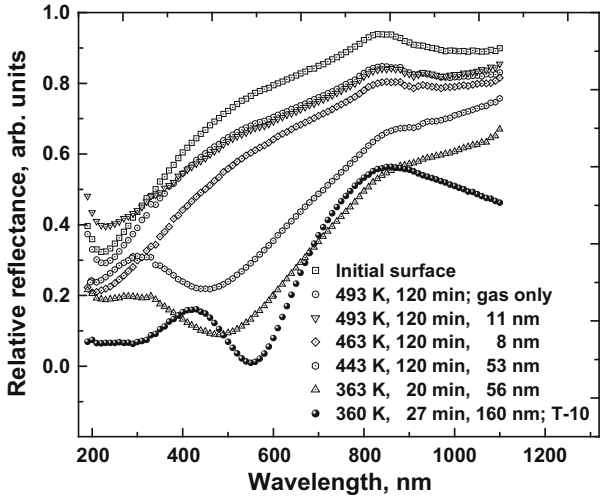


**Fig. 4.** Effect of a-C:H film deposition on reflectance of stainless steel mirrors for laboratory ($Ar/CHD_3/D_2 = 40:5:55$) and tokamak T-10 conditions. The reflectance for mirrors exposed at 493 K with and without (in working gas mixture only) magnetron discharge is practically the same in wavelength range 400–1000 nm.

## 4. Conclusion

Using laboratory ITER-relevant conditions we showed that the heating of mirrors mitigates the growth of hydrocarbon films. On the other hand, an enhanced chemical activity of a hot metallic surface can result in modification of the surface. Moreover, reflectance degradation of the hot mirrors takes place even without plasma discharge at exposure in working gas mixture only.

It could be concluded that the right choice of material is very important for preservation of mirror quality in ITER environment. The investigation of mirror reflectance stability at elevated temperatures will be subject of our future activity.

In our laboratory experiments a transition from deposition to erosion was observed in the temperature range of 450–500 K. The transition temperature can be recommended as an optimal working temperature for first mirrors in ITER. The exact value of the temperature should be searched in situ.

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