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Transport and deposition of injected hydrocarbons in plasma generator PSI-2

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Abstract

The transport and deposition of hydrocarbons were studied in the stationary plasma of plasma generator PSI-2. CH_4 or C_2H_4 were injected into the plasma at different positions in the target chamber. After an interaction between the plasma and the hydrocarbons, different species are produced, some of them having high sticking probabilities and forming a:CH films on a temperature controlled collector. The film growth is studied in situ for different plasma parameters. The 3D Monte Carlo code ERO including three different sets of atomic data is used to describe the formation of hydrocarbon films.

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1. Introduction

It is well known that in all fusion experiments which use carbon as wall materials, long term hydrogen (tritium) retention occurs which would not be acceptable for ITER [1].

An extrapolation from existing facilities is necessary as well as modelling of the processes which occur. In PSI-2, a stationary high current arc discharge, experiments were performed to obtain the relevant information about transport and sticking of hydrocarbons [2]. Well defined amounts of CH_4 or C_2H_4 were injected into the plasma (hydrogen, deuterium or argon discharges) and the deposition patterns were measured in situ for different plasma parameters. The growing rates detected for the a:CH films were compared with ERO-code calculations carried out for our particular conditions.

2. Experimental set up

The experimental set up is explained in [2]. In the middle of the target chamber a temperature controlled collector is fixed 0.06 m away from the plasma (see Fig. 1). There are two possibilities for hydrocarbon injection: (1) via the nozzle which is located opposite the collector and near to the plasma edge having an inner diameter of 1 mm, and (2) via the valve which is located on the same side 0.40 m upstream and close to the vessel wall (distance

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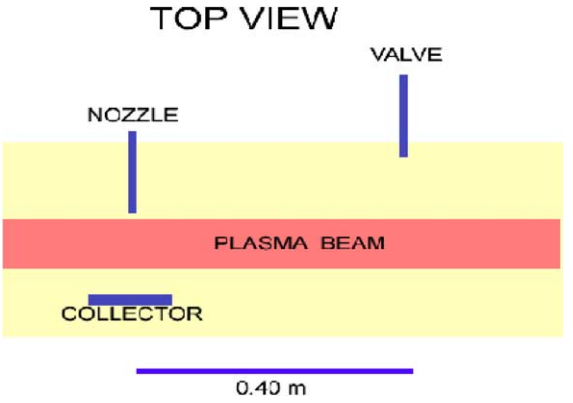

Fig. 1. Target chamber: arrangement of nozzle, valve and collector.

Table 1
Plasma parameters and discharge current for different working gases

Working gas	Electron density (m^{-3})	Electron temp. (eV)	Discharge current (A)
Argon	$(2...50)\times10^{17}$	4...8	50...200
Hydrogen	$(1.5...14)\times10^{17}$	4...8	50...300
Deuterium	90×10^{17}	1...2	400

to the plasma 0.15 m). The radial profiles of the electron density and temperature were determined using a scanning Langmuir probe (see Table 1).

The thickness of the a:CH films was measured in situ by reflection spectroscopy (see [2]).

3. Experimental results and discussion

The same amount of hydrocarbons is injected through the nozzle or the valve. For low density plasma, the majority of injected molecules passes the plasma beam without suffering disintegrating collisions. After reflection at the chamber walls, they enter the plasma beam repeatedly until being converted to a species with high sticking probability which then contributes to the formation of a:CH films on the collector and the inner surface of the target chamber. In this case we expect a more or less uniform distribution of a:CH films along the target chamber and therefore only a small change in the deposition rate at the collector when switching the gas inlet from nozzle to valve. When C_2H_4 is injected into a low density plasma ($n_e = 0.2\times10^{18}\,m^{-3}$), the nozzle and valve rates are found to be nearly equal (0.57 nm/min and 0.48 nm/min) (see Fig. 2).

With increasing plasma density, a larger amount of the injected molecules is converted to high sticking radicals during the first passage through the plasma. This

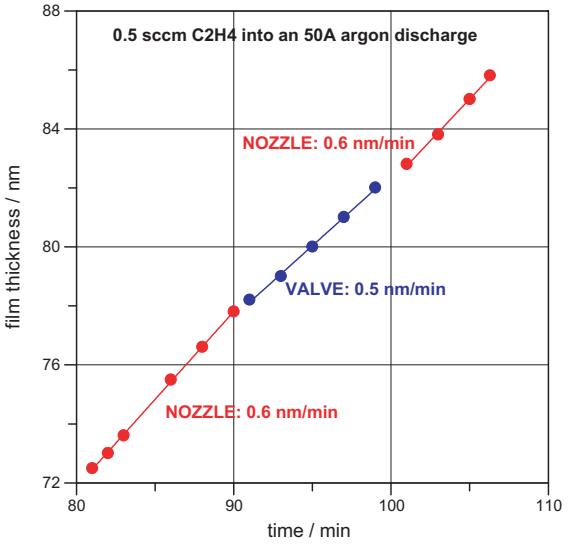

Fig. 2. Film thickness versus time for an argon discharge ($n_e = 2\times10^{17}\,m^{-3}$, $T_e = 2.5$ eV). Injection of 0.5 sccm ethylene.

behaviour is demonstrated in Fig. 3. The same amount of C_2H_4 is injected into an Ar discharge. The ratio of the growth rates (nozzle/valve) rises with increasing density. In contrast to Fig. 2, for an electron density of $5\times10^{18}\,m^{-3}$ there is a clear difference between the two positions of injection. For nozzle injection the growth rate is about 20 times higher than for valve injection. The decomposition of hydrocarbons for such conditions is a local process, that means the mean free path (mfp) for decomposition is smaller than the plasma diameter.

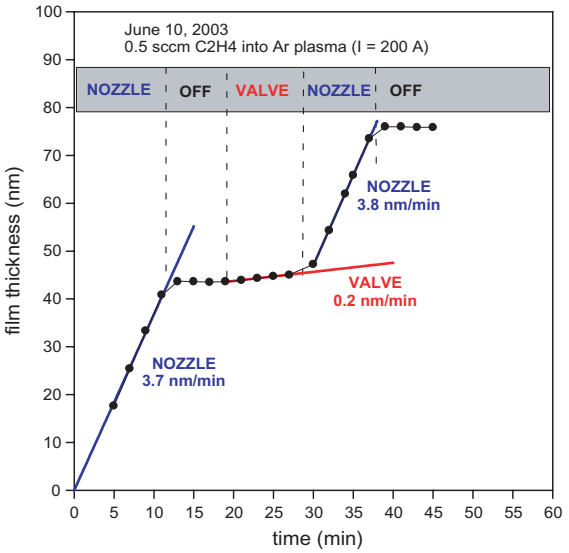

Fig. 3. Film thickness versus time for an argon discharge ($n_e = 5\times10^{18}\,m^{-3}$, $T_e = 5$ eV). Injection of 0.5 sccm ethylene.

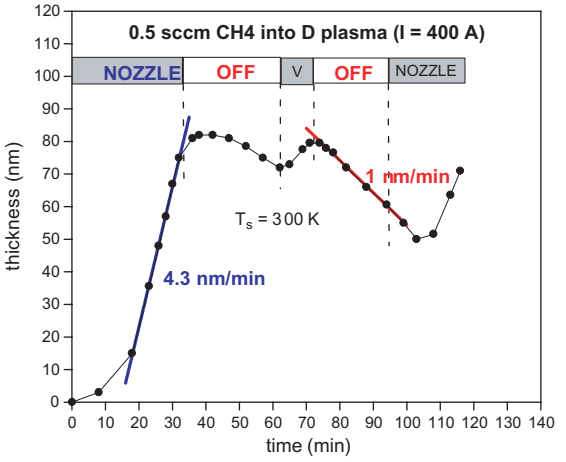

Fig. 4. Film thickness versus time for a high density deuterium discharge ($n_e = 9 \times 10^{18}\,m^{-3}$, $T_e = 2\,eV$). Injection of 0.5 sccm methane.

More complex behaviour was found for hydrogen and deuterium discharges. In contrast to the Ar plasma experiments, no clear local decomposition of hydrocarbons was found. Some experiments were performed with methane injection into a recombining D plasma. In Fig. 4 the behaviour of the a:CD film for methane injection is depicted. The growth rate for injection through the nozzle is only five times higher than for valve injection (for a lower electron density in argon we found a factor of 20). Without injection erosion of the film could be detected independent of the collector position, consequently it cannot be caused by ions. Only atomic hydrogen can explain such a behaviour.

3.1. Modelling

The transport of the injected gas molecules (CH_4 or C_2H_4) has been analysed with the 3D Monte Carlo code ERO [3] which has been adopted to the geometry of plasma generator PSI-2 and the set up used in these experiments.

The profiles measured for the plasma parameters T_e and n_e corresponding to discharge current and the working gas (Ar, H and D) are included in the program. Three different compilations of reaction rate coefficients as provided by Ehrhardt/Langer [4], Alman/Ruzic/Brooks [5,6] and Janev/Reiter [7] are implemented.

The motion of the molecules inside the nozzle is included in the modelling. Almost all neutral particles (with a temperature of about 470 K and a flux of 2.2×10^{17} molecules per second) are directed toward the collector plate. Crossing the plasma column they are ionised and dissociated, producing a broad spectrum of C_xH_y and $C_xH_y^+$ hydrocarbon species, as well as H, H_2 and C (Table 2). The processes considered include electron-impact ionisation and dissociation of C_xH_y, dissociative excitation, ionisation and recombination of $C_xH_y^+$ with electrons, as well as charge and atom exchange collisions of C_xH_y with protons. In the simulations one million particles are started for a good statistics.

Table 2
Hydrocarbons and their sticking coefficients used in modelling

C_xH_y	Sticking coefficient
CH_4	0
CH_4^+	1
CH_3	0.001
CH_3^+	1
CH_2	0.026
CH_2^+	1
CH	0.05
CH^+	1
C	1
C^+	1
C_2H_6	0
$C_2H_6^+$	1
C_2H_5	0.01
$C_2H_5^+$	1
C_2H_4	0
$C_2H_4^+$	1
C_2H_3	0.35
$C_2H_3^+$	1
C_2H_2	0
$C_2H_2^+$	1
C_2H	0.8
C_2H^+	1

The film growth is determined by the number of generated hydrocarbons impinging onto the collector multiplied with their sticking probability. Table 2 lists the hydrocarbons.

With the Ehrhardt/Langer and the Alman/ Ruzic/ Brooks compilations similar results are obtained. In contrast the Janev/Reiter compilation predicts much higher deposition rates since in this case more carbon atoms with high sticking probability are produced during the methane break-up inside the plasma. For further calculations only the Janev/Reiter compilation has been chosen (more results in [2]). It is evident that almost all particles impinging onto the collector plate are neutrals. The (high sticking) ions remain well confined by the magnetic field inside the plasma column. In addition, the collector is protected from ion bombardment by an additional limiter.

The deposition efficiency increases with increasing plasma density due to the simple fact that the number of reactions in a certain volume is proportional to n_e. Thus, in high density plasmas the injected methane molecules are efficiently decomposed to carbon. For example, taking an atomic density of 9.2×10^{28} C-atoms m^{-3} in the (hard) a-C:H film [8], one estimates an average deposition rate of about 0.02 nm/min (see

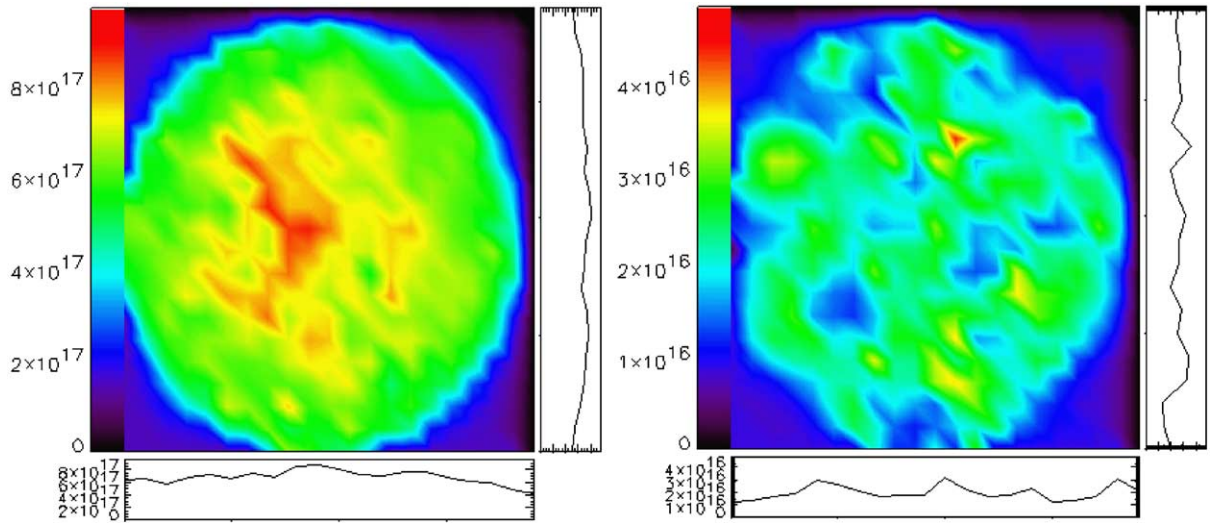

Fig. 5. C_2H_4 and CH_4 injection into a hydrogen discharge ($n_e = 3 \times 10^{17}\,m^{-3}$): Modelled C-deposition rate per m^2s onto the collector and the experimentally determined growth rates for CH_4 (right modelled: 0.02 nm/min, expt.: 1.8 nm/min) and C_2H_4 (on the left modelled: 0.46 nm/min, expt. 2.8 nm/min).

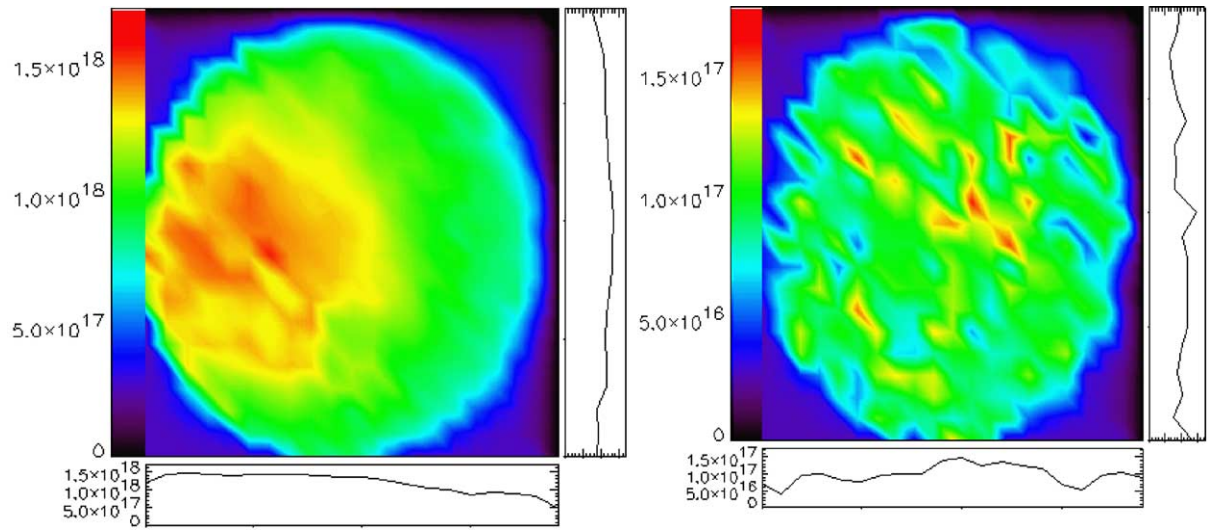

Fig. 6. C_2H_4/CH_4 injection into an argon discharge. Modelled C-deposition rate per m^2s onto the collector for C_2H_4 (on the left: $n_e = 4 \times 10^{18}\,m^{-3}$ modelled: 1 nm/min expt.: 3.8 nm/min) and CH_4 (on the right: $n_e = 1.4 \times 10^{18}\,m^{-3}$ modelled: 0.08 nm/min, expt.: 0.4 nm/min).

Fig. 5) which is considerably smaller than the rate of 1.8 nm/min measured in the experiment for CH_4 injection into a 100 A discharge in hydrogen. Using an atomic density of $5 \times 10^{28}\,C\,m^{-3}$ (about $1\,g/cm^3$) – typical for polymer like films – reduces the discrepancy only partially. The main reason for this discrepancy is the fact that for such conditions the mfp for decomposition is comparable to or larger than the dimensions of the modelling volume. For a 200 A discharge in argon the experimentally determined growth rate is also larger than the rate determined by modelling. Modification of the C-density reduces the difference to a factor of two (see Fig. 6, left hand side).

4. Summary

In situ measurements of erosion/deposition in PSI-2 have been performed under various plasma conditions using gas injection as a hydrocarbon source and Si-wafers as collectors. Such experiments provide a unique

opportunity to check atomic data and ERO modelling by studying the transport and deposition phenomena. Modelling by means of the Monte Carlo code ERO predicts lower deposition rates (by factors of 4–5) than observed in the experiments. In particular for low density conditions, the measured deposition rates turned out to be unexpectedly high. This work has been carried out within the research programme of the European Task Force on Plasma Wall Interaction and was partially supported by TWR-TVM-CFC2.

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