

Formation of Hydrocarbon Films in the Plasma Generator PSI-2

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Introduction

The question of whether graphite can be used as target or wall material in a fusion reactor – or at first in ITER – is essentially related to two adverse properties of graphite: its high erosion probability in general and the high inventory of tritium that is possibly stored in hydrocarbon layers being formed either on the target plates proper or on other surfaces remote from the plasma. With respect to the inventory of hydrogen and its isotopes stored in the cooler parts of the target surfaces carbon-co-deposition (instead of diffusion) has been identified as the dominating process [1]. Whereas the ions play an essential role in this plasma-target interaction the formation of hydrocarbon layers (C:H films) on the remote surfaces can be caused only by fluxes of neutral particles. It is this second problem which is investigated in this paper.

Hot liner experiments

In order to address the problem of hydrocarbon film formation under conditions similar to that of ITER a so called “hot liner” arrangement has been implemented to the PSI-2 plasma generator run at IPP Berlin (see ref. [2] for further details). The hydrocarbons are generated either by chemical erosion of a graphite target or by injecting CH₄ or C₂H₄ into the plasma through a nozzle. The liner duct (1 m length, 0.15 m diameter) is mounted opposite to the target. It is equipped with a heated inlet structure and a number of silicon wafers and cavities are distributed along its length for measuring deposition or erosion of hydrocarbon films. The cavities are closed boxes furnished with a small hole in one of the surfaces. They allow in particular to assess the sticking properties of the species by comparing the thickness of films on the surfaces opposite to hole and adjacent to it.

At the entrance of the duct two different liner heads were tested. The first head was equipped by a number of heated Mo-cylinders. They were arranged in such a way that all hydrocarbons had to perform at least one collision with the hot surface (≤ 1300 K). Because of too low throughput it was replaced by another one consisting of a thin tantalum ribbon meander (220 cm²) which could also be heated to about 1000 K. This second liner had a very high throughput but neither did work as expected. Indeed, the effect of the hot surface at the duct entrance was to enhance the formation of hydrocarbon layers in the duct region. One possible interpretation for this rather strange result is a transformation of hydrocarbons with low sticking properties into more reactive species at the hot tantalum surface (pyrolysis).

The importance of film formation could be impressively documented by blowing methane into a hydrogen discharge. While methane is pumped off by 100% without plasma, only about 10% of the injected fluxes could be found in the exhaust when the plasma was present. This demonstrates that the plasma is actually an effective converter, changing molecules of low reactivity into those with very high sticking probabilities.

Formation and erosion of amorphous H:C films

In what follows we concentrate on experiments where no liner head was in place. The objective behind these studies is to identify the basic mechanisms involved in the formation hydrocarbon films. They were inspired by the observations made by Keudell and Jacob [3] when investigating ECR-produced methane plasmas that there is an interplay between deposition and erosion, the latter caused by atomic hydrogen.

In our studies we measured the growth rate of the H:C films – using an in situ optical method [2] – on silicon collectors positioned in various distances to the plasma. The hydrocarbon fluxes were this time produced by a large graphite target, covering the whole plasma cross-section ($\phi \sim 8$ cm), whose normal being inclined by 45° with respect to the plasma axis, which in turn is defined by the magnetic field ($B \sim 0.1$ T). In order to enhance the H:C fluxes in the direction pointing towards the duct the target was enclosed in a floating molybdenum box provided with two orifices: one opening allowing the plasma beam to enter, the second allowing the formed hydrocarbons to reach the hot liner entrance. It should be noted that the floating potential of the target was not changed by the presence of the Mo-box. During the experiments the box was heated by the plasma and by the radiation emitted from the target; its temperature was 500 K at least. In fact, no coatings were found at the inner walls of the box in post-experiment inspections. Thus, the hydrocarbons may be perfectly reflected at the molybdenum surfaces, but also deposition followed by efficient re-erosion could explain the clean walls.

In Fig.1 results are shown for two collectors, the first one is placed at the edge of the plasma column just opposite to the graphite target, the second one is positioned about 0.4 m away from the plasma center in the duct.

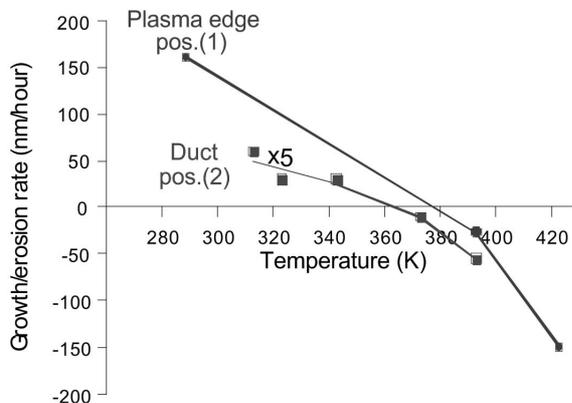


Fig.1: Growth rates of H:C films as a function of surface temperature for two collectors.

In Fig.1 the growth rate of the hydrocarbon layers on both collectors is plotted as a function of the surface temperature. As is to be seen from the figure this rate can be positive (deposition dominated) or negative (erosion dominated). In case of zero growth

rate there is a balance between erosion and deposition. This equilibrium is found for

temperatures of about 100° C, i.e. $T_0 \sim 370$ K, almost independent of collector position. For $T < T_0$ deposition prevails, but erosion rises with temperature and dominates for $T > T_0$. In tendency this result is in agreement with the observations made by Jacobs et al. [3] but they found a significantly higher transition temperature T_0 .

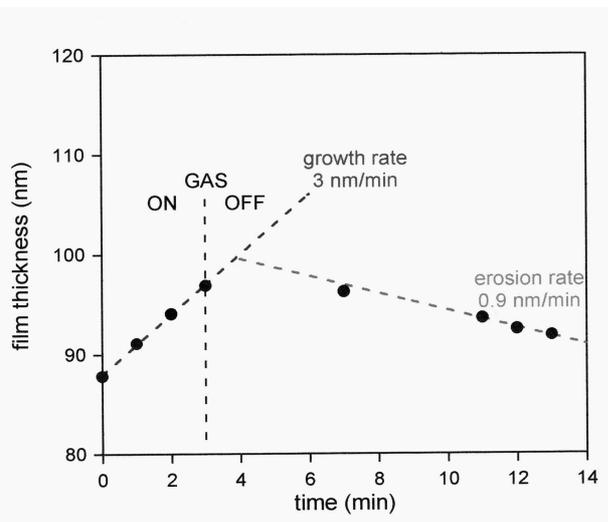
The two fluxes contributing to the net growth rate of the hydrocarbon layers lead us to the following expression for the temporal change of the deposited particles

$$\frac{dN_{CH}}{dt} = \sigma \Gamma_{CH} - \varepsilon(T) \Gamma_H, \quad (1)$$

where N_{CH} is the surface density, Γ_{CH} and σ are the flux densities and the sticking coefficients of the hydrocarbons involved. Actually here only those molecules with high sticking probability ($\sigma \sim 1$) are to be taken into account. Similarly Γ_H is the atomic hydrogen flux density and ε is the corresponding erosion coefficient, which, for the considered temperature range, is a monotonically rising function of T . Equation (1) facilitates the discussion under which conditions a net erosion can be expected. Putting $dN_{CH}/dt = 0$ and $\sigma = 1$ the critical transition temperature is obtained from $\varepsilon(T_0) = \Gamma_{CH}/\Gamma_H$. There is no reliable information with regard to the function $\varepsilon(T)$ available but from analogy with chemical erosion of graphite the following assumptions appear reasonable: $\varepsilon(T)$ reaches a maximum around $T_{max} \sim 800$ K and $\varepsilon_{max} \leq 1$. The latter statement tells us that $\Gamma_H/\Gamma_{CH} > 1$ is a necessary condition for achieving net erosion. Furthermore, the smaller the ratio Γ_H/Γ_{CH} the higher the transition temperature T_0 , provided $T_0 < T_{max}$. This shift to higher transition temperatures is also to be seen in Fig.1 by comparing the zeros of the two collector curves. In fact, we have indications that the hydrogen flux decays more rapidly than flux of the hydrocarbons with distance; at the end of duct the beneficial influence of the hydrogen atoms seems almost negligible.

Experiments with intermittent gas pulses

The previous concept of explaining the growth of H:C films by the competition of growing



and erosion, with the further assumption that only the erosion coefficient ε is a function of temperature, is strongly supported by the results obtained by injecting pulses of CH_4 into hydrogen and argon discharges.

Fig.2: Film thickness vs time applying an initial methane pulse to a H- plasma.

In Fig. 2 the layer thickness is plotted as a function of time for a collector at a constant temperature ($T = 310$ K), positioned 2 cm

behind the duct entrance. For the first 3 minutes the methane is injected at a constant rate of 0.5 sccm and switched off thereafter. The slope of the straight lines connecting the data is clearly seen to change from positive to negative when the source of the hydrocarbons has been turned off. At that later phase only the erosion term in Eq. (1) is present.

That in fact hydrogen is needed to provide erosion is demonstrated in Fig.3. Here methane is injected into an argon discharge in an intermittent manner.

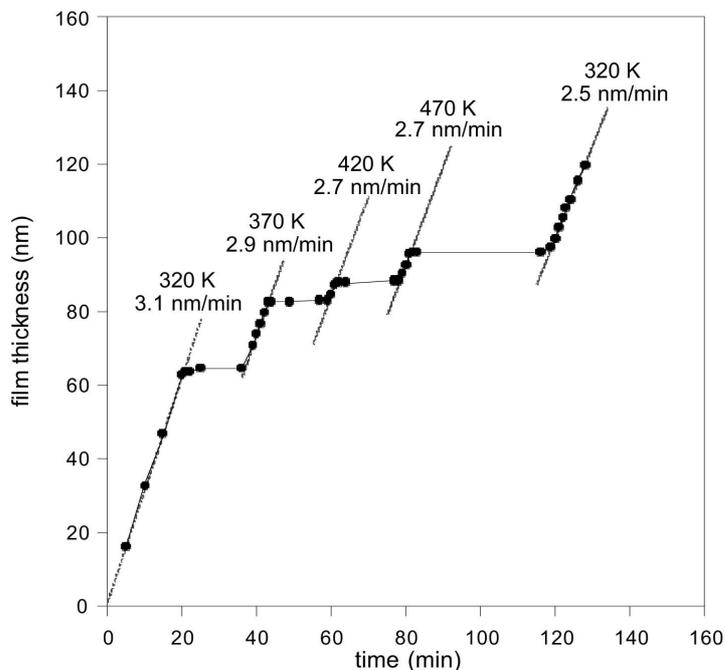


Fig.3: CH_4 injections into an argon plasma.

Intervals of constant blow rates (0.5 sccm) are followed by phases without injection. We notice that, in contrast to Fig. 2, there is this time no decay during the break intervals. Moreover, the surface temperature has been changed and rises during the four intermissions ($T = 320$ to 470 K). Nevertheless, – as indicated in the figure – the slopes do not change, i.e. the

growth rate stays constant at about 3 nm/min.

Summary

- All films are produced by very active particles with sticking coefficient near to 1. The sticking of hydrocarbon radicals is independent of temperature for $T = 320$ - 470 K.
- In hydrogen discharges the growth is determined by a balance between T-independent carbon deposition and T-dependent erosion by atomic hydrogen.
- Heating the duct to 370 K is sufficient to suppress the carbon deposition inside. Placing a hot (1000K) component at the entrance of the duct may lead to additional production of active hydrocarbons via pyrolysis.

References

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