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Decomposition measurements of hydrocarbons in PSI-II

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Introduction

One of the major concerns for current and future fusion devices is the large magnitude of transient heat loads on plasma facing components and especially the divertor plates. So far, the only materials capable of sustaining this thermal stress are graphite and carbon based composite materials. However, as a result of chemical sputtering many different hydrocarbons are formed which are decomposed in the plasma edge. These stable hydrocarbon molecules and radicals result in formation of a-C:H-layers possibly leading to a non-tolerable tritium inventory in the devices. Therefore the understanding of hydrocarbon formation, fragmentation, transport and sticking is an important issue in fusion research.

Experimental setups and proceedures

The experiments were conducted in the plasma generator PSI-II, a stationary high current discharge confined in an axial magnetic field [2]. While the magnetic field is lower (B = 0.1 T) the plasma parameters ($n_e = 1 \cdot 10^{17} \ldots 3 \cdot 10^{18}$ m^{-3}, Te $= 4 \ldots 8$ eV) are comparable to those encountered in the divertor region of larger fusion devices.

In this work, the decomposition of methane (CH_4) was studied using quadrupole mass spectrometry (QMS) as well as infrared tunable diode laser absorption spectroscopy (IR-TDLAS) [1]. The injection was carried out under different plasma conditions and working gases (argon and hydrogen) in order to identify the processes of decomposition and formation of hydrocarbons by plasma chemistry and plasma wall interaction as charged particle-induced, atomic hydrogen-induced, or a combination of both.

A CCD camera with a spectral filter (430 ± 3 nm) and a spectrometer equipped with 16 fiber channels were used to detect the CH molecule band emission (431 nm) with a good spatial resolution. The CCD camera and the fibers were situated perpendicular to the location of injection (nozzle) at opposite sides of the target chamber. The experiments were conducted for different working gases (helium and hydrogen) and different hydrocarbons (CH_4 and C_2H_4) which were injected through the nozzle. In order to determine the influence of charged particles on the decomposition process and the CH band emission, a bias voltage was applied to the nozzle.

QMS vs. IR spectroscopy: Results for methane injection

As a diagnostic benchmark we first investigated the methane signal without plasma for a cold and a heated cathode. The results are shown in Fig. 1 (top left). As can be seen the CH_4 density measured with both diagnostics which were calibrated independently of each other show the same results. Due to thermolysis, a lower density is observed when the cathode is at operating temperature (T=1600 ^{o}C). When plasma is present, the CH_4 density decreases rapidly with incresing plasma density (Fig. 1 (top right)). Due to the localized nature of the decomposition process and the difference in position between the QMS and the IR measurements (IR at position and QMS 30 cm upstream of the injection) we now also observe a difference in the densities obtained using both diagnostics. Fig. 1 (bottom) shows the behaviour of the particle density of the injected gas CH_4 and the decomposition products for argon and hydrogen plasmas. The results for the fragmentation and generation process of methane show a strong dependency on electron density (already for $n_e = 10^{17} m^{-3}$). A comparison between hydrogen and argon plasmas show that the decomposition of the injected gas is more pronounced in hydrogen than in argon plasmas. This could possibly indicate that charge exchange (CX-processes) are involved.

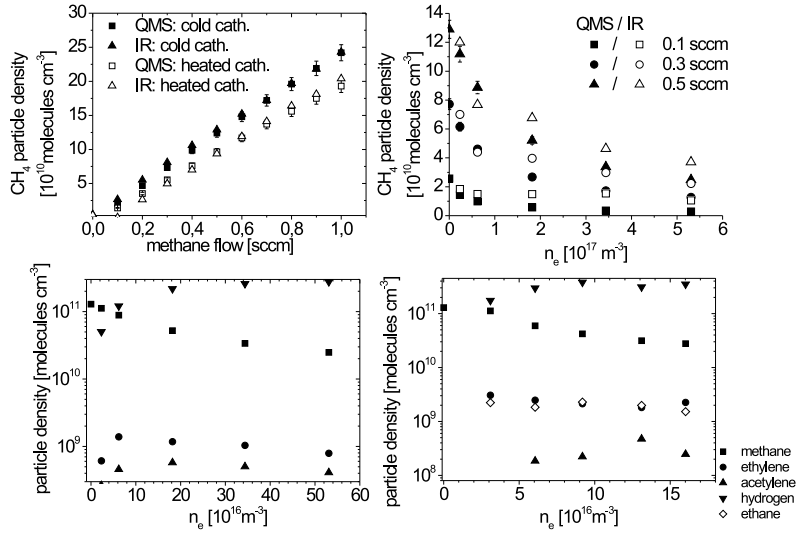

Figure 1: top: Comparison between the results for the particle density of CH_4: without plasma (left), for different amounts of CH_4 in argon plasma (right). bottom: Behaviour of the injected gas and the decomposition products: argon plasmas (left), hydrogen plasmas (right).

For both, argon and hydrogen plasma, ethylene (C_2H_4) and acetylene (C_2H_2) could be detected. Hence, these products are generated by charged particle-induced processes. In addition to this, ethane (C_2H_6) was detected in case of hydrogen plasma. This indicates that for the generation of ethane the presence of atomic hydrogen is required. While the CH_4 density decreases monotoneously with increasing plasma density, the concentration of the reaction products displays a maximum: while the generation of these species first increases with increasing density, the decomposition prevails at higher densities.

The CH_3 and CH_2 concentration was below the detection limit of the IR measurements throughout all experiments but an upper limit of the concentration can be given: $n_{CH_3} \leq 5 \cdot 10^{16}$ m^{-3} and $n_{CH_2} \leq 6 \cdot 10^{17}$ m^{-3}.

CH band emission

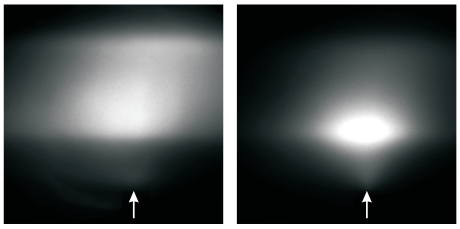

Figure 2: CH band emission patterns for the injection of CH_4 into helium plasma as taken by the CCD camera: $n_e = 5 \cdot 10^{17}$m^{-3} (left), $n_e = 3 \cdot 10^{18}$m^{-3} (right).

The determination of the spatial distribution of the CH band emission gives additional information about the decomposition process. By applying a bias voltage to the injecting nozzle it is also possible to distinguish between ion and electron induced processes in the vicinity of the nozzle. Fig. 2 shows the CH band emission as detected by the CCD camera for different electron densities in helium plasmas with injection of 0.5 sccm of CH_4. For small electron densities there is a global decomposition. However, reaching $n_e = 3 \cdot 10^{18}$m^{-3} the CH band emission and thus the decomposition is localized to the vicinity of the nozzle. This indicates a strong decomposition process from CH_4 to CH already near to the nozzle in the outer region of the plasma. The same behaviour is reflected in the spectrometer measurements depicted in Fig. 3 (left). In contrast to this the emission patterns in case of CH_4 injection into hydrogen plasmas Fig. 3 (right)) indicate a global decomposition for the different electron densities over the whole plasma cross section and in addition a local contribution near to the nozzle. The reason why we find a global process in hydrogen plasmas is on the one hand a smaller charged particle density and on the other hand an additional erosion process at the walls.

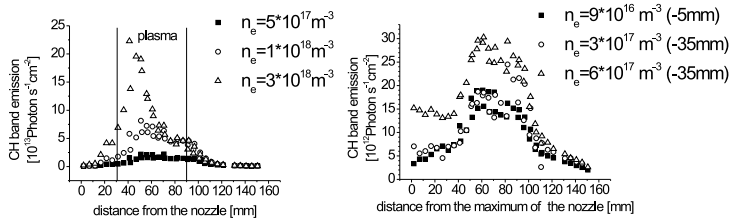

Figure 3: CH band emission measured with the fiber system: in helium plasmas (left), in hydrogen plasmas (right).

To identify whether ions or electrons are responsible for the decomposition process we apply a bias voltage to the nozzle. The results are shown in Fig. 4 for the injection of C_2H_4 into a helium plasma. As can be seen, imposing a positive bias voltage pushes the CH band emission away from the plasma. Conversely, applying a negative voltage has almost no consequence. For CH_4 injection this effect is also present, however, since the CH emission is wright at the nozzle, it is less pronounced. These observations put emphasize on the ions that obviously play an essential

rôle in the decomposition process. In order to verify that the observed shift is not simply related to a change of the local excitation conditions a reference experiment was performed by puffing argon into a helium plasma. In this case, a (slight) increase of the argon radiation is observed for positive bias, i.e. the excitation conditions are actually enhanced. We can thus conclude that the observed decrease of CH band radiation is due to a local decrease of the CH density on account of a reduced ion density.

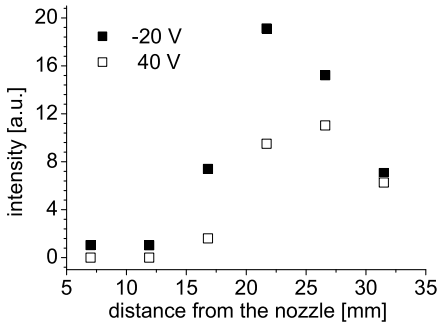

Figure 4: CH band emission integrated over the region λ = 427 - 431 nm for the injection of C_2H_4 into He plasma.

An estimation of the mean free path length λ evaluated by referring to established atomic data yields considerably larger values than those observed: $\lambda_{Ex} \approx 30$ mm. For instance in the case of the three processes

1. $He^+ + CH_4 \rightarrow CH^+ + H_2 + H + He$ [5, 3]
2. $e + CH_4 \rightarrow CH + H_2 + H + e$ [4, 3]
3. $e + C_2H_4 \rightarrow CH + CH_3 + e$ [4, 3]

we estimate $\lambda = 1.4$ m, $\lambda = 730$ m, $\lambda = 63$ m, respectively.

Summary

The results of the experiments with QMS and IR-TDLAS are in good agreement for stable molecules. The densities of the hydrocarbon radicals were below the detection limit of IR-TDLAS but an upper limit for the densities could be obtained from the measurements. It is found that the decomposition process in hydrogen plasmas is more pronounced than in argon and helium plasmas. This indicates that CX-processes are involved in the decomposition of hydrocarbons. Such interpretation is corroborated by CH band emission experiments with a biased nozzle. However, the short decay lengths inferred from the CH band emission patterns cannot be explained on the basis of present atomic data.

References

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