Diagnostics of Hydrocarbons and Their Influence on Hydrogen Plasmas

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Abstract. Spectroscopic diagnostics of hydrocarbons is applied to low-pressure methane plasmas and hydrogen plasmas which are in contact with graphite. The CH radiation (at 431 nm) is correlated with the CH density and with the methane flux. This correlation is generally applied in fusion experiments to obtain methane fluxes. The present investigations show that the C_2 radiation (Swan band at 516 nm) corresponds to the C_2 density which refers to the C_2H_2 flux and provide a spectroscopic diagnostic tool for these higher hydrocarbons. By introducing various surfaces into hydrogen plasmas, atomic hydrogen fluxes are lowered and reflection coefficients are derived from the Balmer radiation. Furthermore, the vibrational population of hydrogen molecules is influenced, in particular in case of graphite. Measurements in methane plasmas show that the enhancement of population corresponds to the formation of hydrocarbons which dissociate in the plasma and can separate vibrationally excited hydrogen molecules.

1. Introduction

Hydrocarbons are present in fusion experiments due to chemical erosion of carbon tiles. The resulting methane fluxes are generally determined by measuring the radiation of the CH or CD molecule applying the so-called D/XB ratio [1, 2]. However, higher hydrocarbons, i.e. C_2H_y can be produced as well and the C_2 radiation may be a candidate for determining these fluxes. The paper presents systematic investigations of spectroscopic diagnostics in methane laboratory plasmas with special emphasis on the contribution of radicals to CH and C_2 radiation. Analysis of these plasmas will then be applied to hydrogen plasmas being in contact with graphite. A comparison of carbon fluxes, which are determined by weight loss measurements and by emission spectroscopy is carried out. Furthermore, measurements of reflection coefficients of hydrogen atoms at various surfaces are presented.

Additionally, hydrocarbons as well as vibrationally excited hydrogen molecules may play a role in divertor recycling, i.e. affect detachment by molecular assisted recombination (MAR) [3, 4]. The processes which produce these molecules have to be understood in detail, assisted by measurements. The interpretation of results for vibrational populations of H_2 in H_2 plasmas on graphite surfaces [5] is supported by a comparison of H_2 with CH_4 plasmas. A discussion of methane dissociation channels supplements the presented results.

2. Diagnostics of Hydrocarbons

Diagnostics of hydrocarbons was carried out in low-pressure laboratory plasmas. These plasmas allow systematic parameter studies with well known plasma parameters (n_e, T_e, n_n, T_n).

In methane plasmas, particle densities of radicals were determined by spectroscopic diagnostics and mass spectrometry. The residual gas analyser was calibrated such that particle densities of CH_4, H_2 and C_2H_y (C_2H_2, C_2H_4 and C_2H_6) were detectable. Spectroscopy of an emission line of He (728 nm) allows a precise determination of T_e. Helium was added to the

discharges as diagnostic gas and for active variation of T_e. The radiation of the Balmer lines was used to obtain the atomic hydrogen density. The analysis of T_e and n_H is described in [6]. The emission band of CH (431 nm) was used to determine the CH density. The C_2 density was derived from the emission of the Swan band at 516 nm. Details of the spectroscopic method and the corresponding rate coefficients are given in [7].

Figure 1 shows T_e and particle densities measured in 5%CH_4/He plasmas in a MW discharge with electron cyclotron heating. As expected, methane dissociates in the plasma and is a factor of 5-10 below the density which is fed into the chamber. The dominant species is H_2. Higher hydrocarbons (C_2H_y, with 60% of C_2H_2) are produced and their density is 10-30% of the methane density in the plasma. As a consequence, for the analysis of particle densities of CH and C_2 from emission bands, dissociative excitation from methane and higher hydrocarbons was taken into account in addition to direct excitation from the radicals themselves. A dissociative excitation path from methane to C_2 does not exist. From the measured CH_4 and C_2H_2 densities the contribution of dissociative excitation to the radiation of CH and C_2 bands was derived (Fig. 2) and is determined to be in the range of 10%. Just like the CH radiation is correlated with the CH density, the C_2 radiation yields the C_2 density. Similar relations are found in erosion plasmas, i.e. hydrogen plasmas (10%H_2/He) on graphite surfaces (EK98). Measured particle densities and T_e are also shown in Fig.1. The CH_4 concentration in the plasmas reaches 0.1%, the C_2H_y formation is a factor of four lower. Calculating the dissociation of methane and that of higher hydrocarbons, the CH and C_2 radiation can be used for measurements of these densities, respectively. The method is already established for CH band emission, which yields the methane flux in fusion experiments. Since C_2 particles are formed by dissocia-

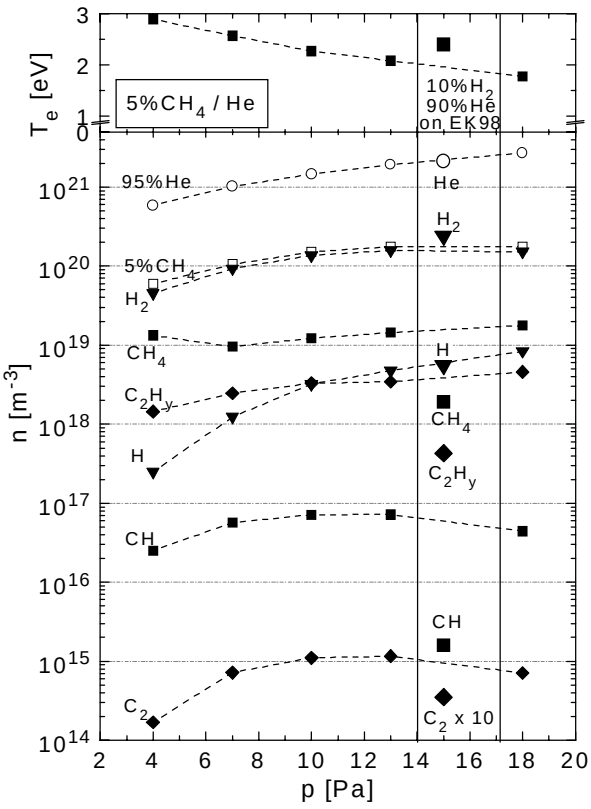

Fig. 1: *Measured electron temperature and particle densities in CH_4/He plasmas as well as in a H_2/He plasma being in contact with graphite (EK98).*

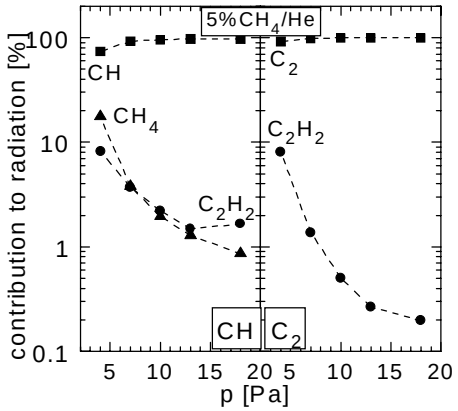

Fig. 2: *Contributions to CH and C_2 radiation originating from direct and dissociative excitation.*

tion of C_2H_y, the presented investigations at laboratory plasmas suggest that C_2H_2 fluxes can be derived from the radiation of the C_2 Swan band if the corresponding D/XB ratio will be determined. First calculations of dissociation channels have shown that the dominant reaction chain, which produces C_2, is $C_2H_2 + e \rightarrow C_2H + H + e$ followed by $C_2H + C_2H \rightarrow C_2H_2 + C_2$. The C_2 particles are then excited by electron impact and decay by spontaneous emission of, e.g., the Swan system.

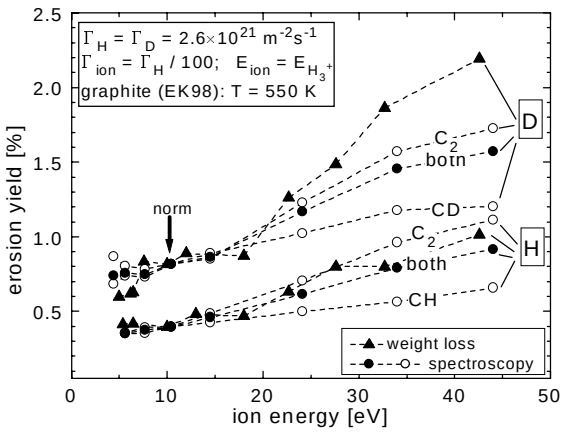

Fig. 3: *Erosion yields of graphite in hydrogen and deuterium plasmas.*

The spectroscopic diagnostic method was applied to measure erosion yields of graphite in hydrogen and deuterium plasmas [8]. Absolute values of carbon fluxes were determined from weight loss measurements. Figure 3 compares results from these measurements with results from emission spectroscopy considering CH (CD) or C_2 radiation and a combination of both. The measurements were carried out in inductively coupled plasmas, which provide homogeneous plasma parameters above the graphite surface. Atomic hydrogen and deuterium fluxes are 2.6×10^{21} $m^{-2}s^{-1}$, ion fluxes are a factor of 100 lower. The dominant ion species is H_3^+ (D_3^+). The graphite temperature was 550 K. In order to compare dependencies, spectroscopic results were normalized to the absolute value of the weight loss measurements at E_{ion} = 10 eV. In general, C_2 radiation increases stronger with ion energy than CH (CD) radiation, which indicates a stronger formation of C_2H_y. Dependencies on ion energy are reproduced only by spectroscopic results if the radiation of CH and C_2 is taken into account.

3. Hydrocarbons in Hydrogen Plasmas

Since hydrocarbons can affect hydrogen plasmas, the radiation of Balmer lines and molecular bands (Fulcher band) was analysed. Atomic hydrogen fluxes were determined from Balmer lines with and without additional surfaces in the plasmas [8]. The motion of the neutral particles is dominated by diffusion and the density gradient refers to the atomic flux. A material dependence (at 650 K) as well as a clear isotope effect was obtained. The following reflection coefficients were derived from the atomic fluxes: high-grade steel: R = 5%(H), 40%(D); graphite: R = 40%(H), 65%(D); copper: R = 5%(H), aluminium: R ≅ 100%(H).

Concerning the vibrational population of hydrogen or deuterium molecules, a remarkable enhancement occurs by introducing graphite surfaces into hydrogen plasmas [5]. In order to check if this is caused by surface effects or by hydrocarbons which are formed at the surface and penetrate into the plasma, investigations were carried out in hydrogen and methane plasmas. Figure 4 shows vibrational temperatures in the ground state of molecular hydrogen (T_{vib}) in hydrogen plasmas (open symbols) and methane plasmas (filled symbols) depending on the

admixture to helium. The corresponding T_e is labelled in the figure. In hydrogen plasmas, T_{vib} decreases with increasing molecular content in the plasma, which is correlated with a decrease in T_e. In methane plasmas, the dominant particle species is molecular hydrogen (Fig. 1). An enhancement of T_{vib} is observed at low methane admixtures in comparison with hydrogen admixtures. T_e is lower and the decrease of T_{vib} with molecular contents becomes stronger. This characteristics indicates a threshold temperature for dissociation processes of hydrocarbons which can separate vibrationally excited hydrogen molecules $H_2(v)$. The following reaction channels may separate $H_2(v)$: electron impact dissociation of methane, which depends on T_e and n_e; heavy particle collisions of CH_y radicals (dependence on T_n and n_n), which produce also higher hydrocarbons (e.g. $CH_4 + CH_4 \rightarrow C_2H_6 + H_2(v)$) and electron impact dissociation of higher hydrocarbons. Thus, interpretation of measurements of T_{vib} has to be correlated with individual particle species in the plasmas, which is in progress.

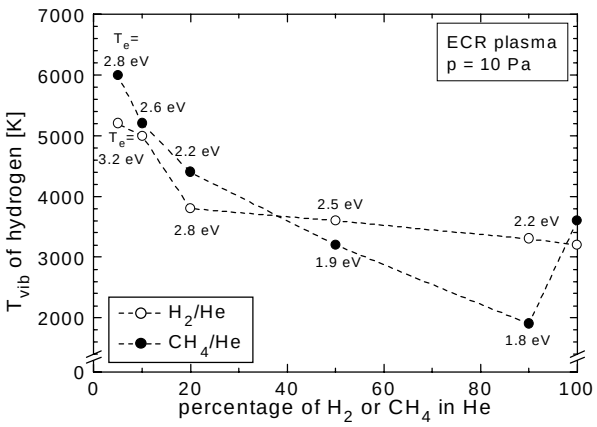

Fig. 4: *Vibrational temperatures of molecular hydrogen in H_2/He and CH_4/He plasmas.*

4. Conclusions

Investigations of methane dissociation and erosion yields in low-pressure plasmas (T_e = 2 – 4 eV, $n_e = 10^{17}$ m^{-3}) were carried out by using emission spectroscopy as diagnostic tool for hydrocarbons. In particular, the radiation of the C_2 Swan band was analysed to correspond to the C_2 density. A dissociation path was suggested to be modelled to receive the density of C_2H_2 from the Swan band. Thus, measurements of erosion yields can be supplemented by C_2H_2 fluxes, which are easily accessible by emission spectroscopy of C_2 radiation at 516 nm. Furthermore, it was shown that the dissociation of hydrocarbons enhances the vibrational population of hydrogen molecules. Measurements of C_2H_2 fluxes and their influence on $H_2(v)$ are necessary for an improved understanding of their influence on divertor recycling, which is the aim of further investigations.

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