Influence of discharge geometry on extraction of positive ion populations from atmospheric pressure plasmas

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Molecular beam mass spectrometry is a powerful tool used in the last period of time to characterize atmospheric pressure plasmas. Neutrals and ions population in helium plasmas, with air impurities or additional gases, may be sampled and studied in various electrode configurations. We found a strong influence of the discharge geometry, relative to spectrometer's sampling orifice, on both molecular mass distribution and clustering processes in the plasma volume, for a feed gas mixture of helium - hydrogen (0.1%) – hydrocarbons (1.2%) type.

1. Introduction

Laboratory experiments with relevance for molecular astrophysics offer valuable data on physical and chemical processes inside astronomical objects. Of particular interest are hot cores related processes, as these structures enveloped in giant molecular clouds, show rich chemical diversity (over 150 different organic and inorganic molecules identified to date) and relatively low temperature (100-300 K range, consistent with conventional chemical synthesis conditions). Plasma laboratory experiments, performed at atmospheric pressure, may represent a good solution to generate molecular families relevant for astrochemical processes, as it was already in pointed by some recent studies [1-2].

With regard to mass characterization of chemical compounds present in plasma volume and on surfaces, mass spectrometry is a widely used technique, applied mainly for low pressure plasmas. Nevertheless, molecular beam mass spectrometry is an alternative to study atmospheric pressure plasmas, with reliable results. As most atmospheric pressure discharges generate pulsed plasmas, with considerable spatial non-homogeneity and the mean free path is very short, allowing efficient recombination processes, the sampling process of charged species is difficult to achieve. Recent studies pointed that these difficulties may be overcome using appropriate plasma generation solutions, based on dielectric barrier discharge principle or RF discharges. Positive and negative ions studies applied for atmospheric pressure plasmas have shown a rich chemical environment based on water clusters, for helium feed discharge with air or water impurities [3-6] or even gaseous oligomers [7-8], if the discharge is feed with noble gases mixed with polymerisable molecules.

In this paper we discuss the effect of discharge geometry on extraction of positive ion populations from atmospheric pressure plasmas, feed with a gas mixture of helium - hydrogen (0.1%) – hydrocarbons (1.2%) type. This particular gas mixture was selected as potential candidate to generate complex molecular families, relevant for the molecular astrophysics of hot cores.

2. Experimental

A quadrupole-based molecular beam mass spectrometry system was used for this study (HPR-60 MBMS, from Hiden Analytical Ltd), with 2500 amu upper mass range. Time averaged data were collected, stressing the overall effect of plasma on chemical fragmentation, synthesis and clustering. Controlled gaseous atmosphere is assured by constant gas feed: helium, 2.45 L/min flow rate, hydrogen, 1 mL/min flow rate, and methane (CH_4) or butane (C_4H_{10}), 4.25 mL/min flow rate.

All studied experimental arrangements for plasma generation where hosted by a stainless steel reactor coupled directly to the mass spectrometer flange as shown schematically in Fig. 1.

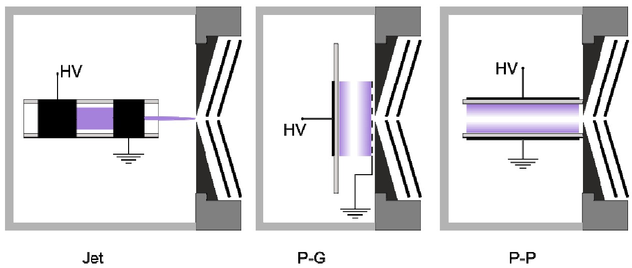

Fig. 1. Mutual arrangements of plasma generation region and mass spectrometer sampling orifice.

Three geometries, all based on dielectric barrier discharge principles, where used for plasma production in the above mentioned atmosphere: a jet like discharge (Jet, Fig. 1) and two plan parallel discharges, one having a grid as ground electrode (P-G, Fig. 1) and the other one a glass dielectric (P-P, Fig.1). High voltage sinusoidal waves are applied on power electrode, with 1 kHz frequency.

Individual current pulses (multi peak discharge), with mean amplitude of 75 mA are generated on all increasing and decreasing edges of the high voltage wave.

3. Results and discussions

Positive ions signals detected in jet and plan-plan geometries are considerably different (Fig. 2).

First of all, it was not possible to record any mass spectrum for the jet like discharge, feed with pure helium, mainly due to instabilities of the plasma jet contact with the sampling orifice. For the other two geometries, the spectrum contain fingerprints of $H^+(H_2O)_n$ clusters, with n up to 5 and signals from He^+, N_2^+, O_2^+, CO_2^+ ions.

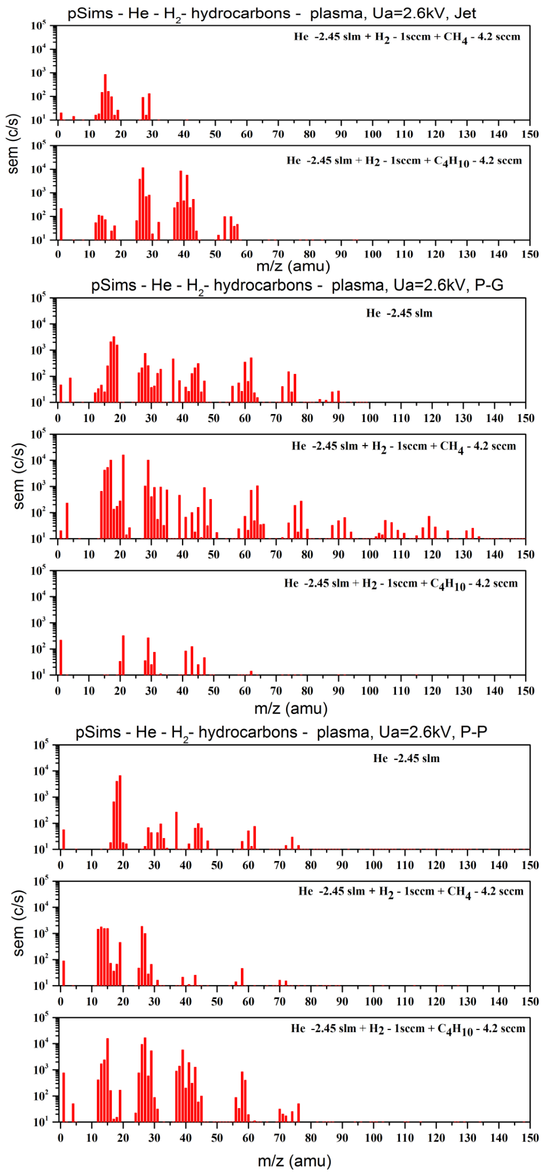

Fig. 2. Positive ion mass spectra for helium – hydrogen – methane/butane plasmas at atmospheric pressure.

The positive ion spectra of helium-hydrogen-hydrocarbons atmospheric pressure plasmas are dominated by grouped mass signals of $C_nH_{n\pm m}$ species (n up 13 and m=1-5). Also, an important feature is the possible presence of enols and keto species ($C_nH_{2n}O$, n=1-8), involved usually in astrochemical and astrobiological mechanisms.

As very well known in mass spectrometry studies of organic compounds, it is practically very difficult to identify all peaks in the mass spectra. Supplementary spectroscopic techniques should be employed to push the analysis further and to give new insights related to molecular astrophysics.

4. Conclusions

Both, molecular mass distribution and clustering processes in the plasma volume, as studied by molecular beam mass spectrometry, are influenced by the discharge generation method. Further investigations on flow regime and life time of detected species are necessary in order to tune these type of plasma sources for molecular astrophysics experiments.

5. References

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