Optical Study of CH_4 Decomposition in an Atmospheric-Pressure ns-Discharge

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Abstract: We report the results of an optical study related to CH_4 decomposition in a He-based ns-pulsed discharge operated at room temperature. The evolution of the absolute densities of He metastable (He^{met}) and H ground state atoms upon CH_4 admixtures is investigated using AAS and calibrated TALIF techniques, respectively. The H_β line Stark broadening analysis delivers the electron density in the plasma core (10^{15} cm^{-3} in pure He).

1. Introduction

Several plasma technologies are being intensively investigated for the reforming of hydrocarbons, aiming at the synthesis of CO_2-free H_2. Nowadays, warm plasmas with a high energy density, such as gliding arc discharges and microwave torches, are often considered as the most promising processes. This has comprehensively drawn most of the attention on the development of near- and at equilibrium discharges for reforming purposes.

Meanwhile, in non-equilibrium plasmas, the role of vibrational excitations in several molecular dissociation mechanisms may lead to a much higher energy efficiency [1]. This study examines a non-equilibrium plasma by focusing on CH_4 decomposition in a highly non-equilibrium atmospheric ns-pulsed jet-type discharge.

2. Methods

The studied ns-discharge (Fig. 1) was designed based on a single thyristor. It produces 10 ns-wide current and 130 ns-wide voltage pulses [2,3] and is operated with a pure He flow of 0.5 L min^{-1} as a basis. Small admixtures of CH_4 are then added to investigate the CH_4 decomposition mechanisms, whereas water vapor and dry air may also be used to implement rotational spectroscopy.

The H ground state atoms are detected using the two-photon absorption laser-induced fluorescence (TALIF) technique, which is calibrated with Kr admixtures [4]. The gas temperature is probed using N_2, N_2^+, CO and OH rotational emission bands [2]. Finally, the electron density is measured through Stark spectroscopy, using the broadening of He and H_β lines [3].

3. Results and Discussion

The previous diagnostics performed in this discharge already revealed that the electron density in the plasma core could approach 10^{16} cm^{-3} [3], whereas the gas temperature would remain close to 300 K [2]. Moreover, the electric field could rise as high as 25 kV cm^{-1} [3], and the density of He^{met} atoms could exceed $2 \cdot 10^{14}$ cm^{-3} (7 ppm) in the pure He plasma case [5]. These results suggest a strong non-equilibrium degree in the plasma core itself. Admixtures of either dry air or water vapor resulted in a drop in electron density to the 10^{14} cm^{-3} range in the core, which may also be expected in the case of CH_4 admixtures.

It is now stated that the well observed overexcitation of the N_2^+ rotational bands as dry air is injected in the plasma may be a consequence of a He^{met}-rich environment, and that these populations tend to collapse as CH_4 is added, resulting in the vanishing of the N_2^+ overexcitation. We investigate this phenomenon with emission spectroscopy.

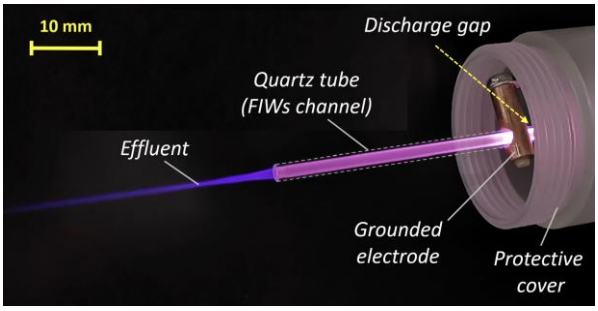

Fig. 1. A photograph of the ns-jet discharge working in a pure He gas flow. The high voltage electrode is located inside the protective cover on the right side. [5]

Furthermore, the combination of electric measurements and H ground state atom quantification through laser-based spectroscopy delivers the trends in H production energy cost as a function of the CH_4 admixture.

4. Conclusion

This study investigates the impact of CH_4 addition in a He-based ns-pulsed non-thermal discharge and suggests that a strong non-equilibrium in the plasma core may promote a rather low-energy CH_4 chemistry during the plasma pulse, compared to near-equilibrium plasmas.

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