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S.E.T.U.P. "Experimental and Theoretical Simulations Useful for Planetology" related studies in the frame of a program of Titan's atmosphere laboratory simulations

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Abstract

One of the final goals of the SETUP program is to be able to determine the physico-chemical processes involved in the complex organic chemistry of Titan's atmosphere. With this aim, representative simulation experiments (in term of temperature, pressure and energy deposition) will be performed using a unique device (figure 1). The latter consists of a reactor where the initial gas mixture (N_2/CH_4) will be exposed, for the first time, to both major energy sources that are responsible for the chemical evolution of Titan's neutral atmosphere (i.e. electrons and photons). Indeed, cold microwave plasma discharge will be settled in order to dissociate a N_2 flow. In Titan's atmosphere, photolysis of methane mainly involves Lyman-α photons (121.6 nm ⇔ 10.2 eV). Classical H_2 discharge lamps can deliver such UV photons. Unfortunately, these continuous sources are not suitable for high-speed kinetic studies. Therefore, in our simulation experiments, the use of a pulsed laser (KrF Excimer delivering photons at 248 nm ⇔ 5 eV) has also been considered to dissociate methane. The chemistry between N atoms and CH_3, CH_2, CH photofragments will be initiated in the interaction zone simulating Titan's chemistry. Using Cavity Ring Down Spectroscopy technique, we will be able to analyze in situ, qualitatively and quantitatively, the stable species as well as the short life intermediates. Then, the chemistry will be determined precisely, and consequently, its description will be refined in theoretical models.

This paper presents two kinds of studies that have been performed as preliminary steps to check the representativeness of the planned simulations of Titan atmosphere. First, the dissociation of molecular nitrogen through microwave discharge has been investigated; secondly, methane photolysis at 121.6 and 248 nm has been examined.

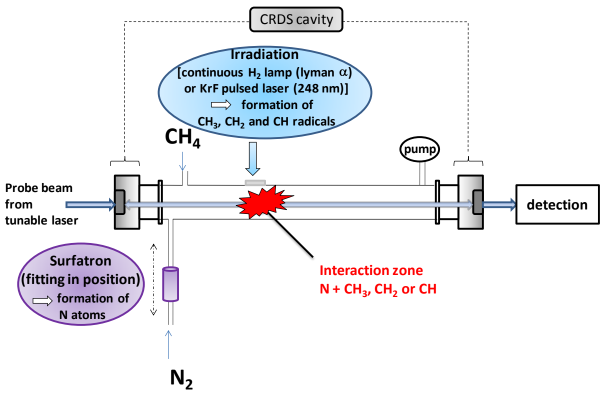

Figure 1: Experimental device for new program of representative laboratory simulations of Titan's atmosphere

Plasma studies

In our Titan's simulation experiments device, thanks to a surfatron, a microwave flowing post-discharge is created in a tube directly connected to the reactor. Nitrogen atoms in the ground state, $N(^4S)$, is generated and flow into the interaction zone. One of our first concerns about those experiments was to quantify and control the nitrogen atoms produced. TALIF (Two photons Absorption Laser Induced Fluorescence) is a powerful technique to quantify the N atoms generated in the simulation experiments [1]. We have used this technique in order to study the late afterglow of a N_2 plasma discharge within several experimental conditions: pressure range of 1-45

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Torr, flow rate between 20 and 500 sccm and microwave discharge power from 30 to 160 W (figure 2). We have shown that the control of these parameters allows adjusting the N_2 density in the reactive zone [1]. Thus, from now on, we are able to determine the position of the surfatron in order to get the proper amount of nitrogen atoms involved in the reaction with methane photofragments in the interaction zone of the reactor. Results of these plasma studies will be detailed.

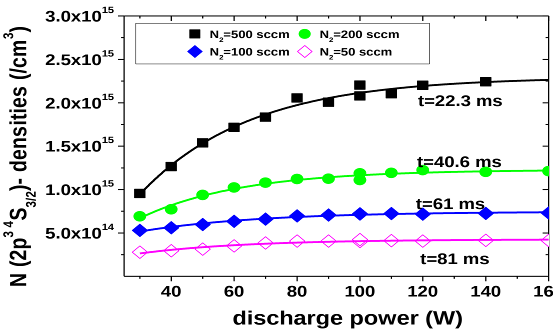

Figure 2: Absolute N (^{4}S)-atom densities in pure N_2 measured by TALIF spectroscopy diagnostic in the flowing post-discharge as a function of the discharge power for different values of residence time and N_2 flow rate.

Photochemical studies

A comparative methane photolysis study at 121.6 and 248 nm - involving one or two photons respectively - have been performed. Preliminary results based on IR analysis of the stable products formed after irradiation of CH_4 at both wavelengths have shown that photolysis at 121.6 and 248 nm could be considered as energetically equivalent (two-photon process at 248 nm) [2]. However, the nature and the abundance of the resulting hydrocarbons suggested that the photodissociation channels might be different (figure 3). To explore this hypothesis, an approach combining irradiation experiments and photochemical modelling has been developed. This methodology is indeed well-suited to test for the accurate description of the different mechanisms involved as already shown by our group in several studies [3], [4]. From the present study, some information about chemical mechanisms following both types of photolysis have been retrieved showing that primary products obtained at these two wavelengths are different. These results and their implication on the development of the experimental program will be presented.

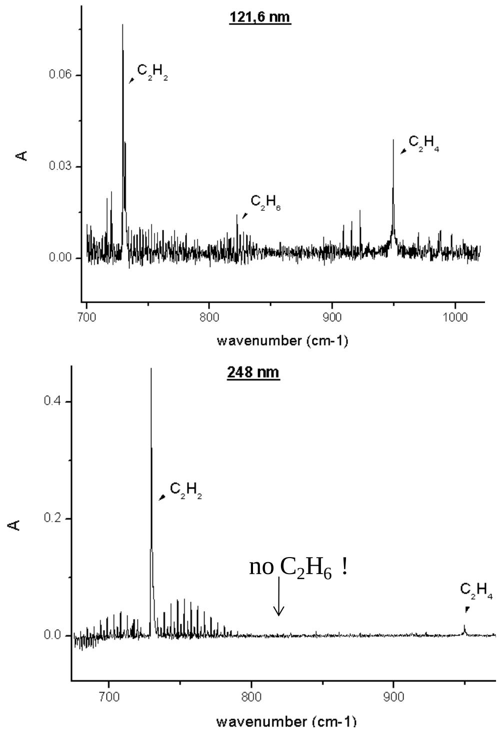

Figure 3: IR spectrum of the resulting gas mixture after methane irradiation at 121.6 nm (top) and 248 nm (bottom).

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