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Gas-Phase Positive Ion Insights during Laboratory EUV Irradiation of Titan's Relevant Gas Mixtures

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Abstract

In the atmosphere of Saturn's moon Titan, the chemical growth is believed to occur through an efficient chemical coupling between radicals and reactive charged species. However, the role of positively charged particles in haze formation chemistry is far from being understood. Here, we present a mass spectrometry investigation with a relatively low pressure interface to chemically characterize the positive ions formed in the pre-haze formation phase starting from the EUV photolysis of N_2/CH_4 gas mixtures.

1. Introduction

Thanks to the discoveries of the Cassini-Huygens mission, it is assumed that the haze formation in Titan's atmosphere is initiated in the ionosphere, mainly by energetic solar photon dissociating N_2 and CH_4, the main components.[1, 2, 3, 4, 5] Heavy ions have been detected including cations up to m/z 99 by the Cassini Ion and Neutral Mass Spectrometer (INMS) at both even and odd masses suggesting positively charged C- and N-bearing species.[6, 7] However, even if they are thought as a key step in haze formation their formation is far from being understood.[2, 4]
In order to better understand Titan's haze formation, numerous laboratory experiments investigated the gas phase products in the atmosphere of Titan through irradiation of N_2/CH_4 mixtures using various energy sources (e.g., plasma discharge, electron beam, photon flux) to simulate the molecular growth occurring in Titan's upper atmosphere.[8] However, commonly used light sources allow to work only above 110nm (Far UltraViolet) which is the dominant energy in the low atmosphere and do not provide sufficient energy to ionize or dissociate N_2, essential to understand observed N-incorporation into gas-phase products. It requires EUV wavelengths (<110 nm) to address the effect of photoionisation of N_2 in the upper atmosphere of Titan. There is only two EUV studies on N_2/CH_4 gas mixtures using synchrotron radiation [9, 10] in which they showed that N_2^+ enhances the formation of unsaturated hydrocarbons at wavelength below 80nm. Thus, ion chemistry is a potential pathway for nitrogen incorporation even if there is still a lack of data characterizing ion composition in Titan's atmosphere simulation experiments.
Thus, our work aims at corroborating the major role, played by the photoionization of nitrogen in the formation of complex organic molecules by monitoring ion EUV photoproducts.

2. Experimental Method

Accordingly, here we present the chemical composition of cations produced in the APSIS (Atmospheric Photochemistry SImulated by Synchrotron) reactor during the irradiation at 73.6nm of a methane in nitrogen gas mixture using an EUV source.[11] The light source used a neon gas flow discharge in the mbar pressure range coupled windowless to the photochemical APSIS reactor. In situ mass spectra were taken with a relatively low pressure interface to determine the chemical composition of ions.

3. Results

Figure 1 illustrates mass spectra of cations measured in the APSIS reactor during irradiation at 73.6nm for different pressures, ranging from 10^{-2} to 1mbar. Main ion species already observed in previous EUV experiments are detected such as CH_5^+ and $C_2H_5^+$ along with an efficient ion production up to m/z 140 at 1mbar. Preliminary analysis allows to believe that the production comes initially from a dissociative charge-transfer reaction between ionized molecular nitrogen and methane leading to the formation of unsaturated hydrocarbons through subsequent dissociative recombination.

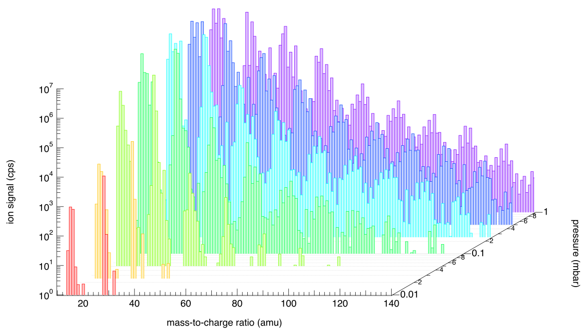

Figure 1: Positive ion mass spectra obtained from N_2/CH_4 gas mixtures irradiated 73.6nm at different pressures, ranging from 10^{-2} to 1mbar.

This work will allow to constrain the detailed chemical kinetic model by bringing informations about ionic species formed in N-dominated atmospheres. This is important to reduce the sources of uncertainty and/or bias in the model predictions of Titan's ionosphere.

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