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EUROPLANET SCIENCE CONGRESS
EPSC2022

EPSC Abstracts
Vol. 16, EPSC2022-448, 2022
Europlanet Science Congress 2022

Heterogeneous chemistry on Titan : Evolution of Titan's tholins through time with gas phase chemistry

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1 - Introduction

In the atmosphere of the satellite Titan, the photochemistry of its two main components N_2 and CH_4 leads to the formation of complex organic molecules, up to the production of solid aerosols, in the form of an orange haze. Observations from the Cassini-Huygens mission [1], as well as models [2] and laboratory experiments [3], strongly suspect that once formed in the ionosphere, the haze will reside for some time in Titan's atmosphere until settling on the surface. Our aim is to investigate experimentally the interaction of the haze particles with their atmospheric chemical environment, focusing on possible reactive molecules produced by gas phase photochemistry of N_2 and CH_4 such as HCN, HC3N, C_2N_2, C_2H_2, C_2H_6. We more specifically addressed the absorption processes of the gases on the particle (uptake coefficients).

2 - Experimental method

In this experimental study, a dusty plasma reactor is used to simulate the atmospheric chemistry of Titan [3], as well as the synthesis of Titan's aerosols analogues (tholins). The gaseous precursors formed by electronic dissociation were monitored in-situ by mass spectrometry, simultaneously with the formation and growth of the haze particles. The properties of the tholins are analyzed by scanning electron microscopy (morphology and size) and high resolution mass spectrometry, LDI-FTICR (chemical composition). In this study, the injection gas flow rate was optimized in order to increase as much as possible the residence time of the gas mixture in the reactor. The chemical growth of the solid particles is thus favored, allowing to follow simultaneously the formation and the evolution of the particles, as well as the co-evolution of the composition of the gas mixture until reaching a stationary gas chemistry, which will not change any more during the whole experiment.

3- Results

3.1 - Temporal evolution of the gas phase by mass spectrometry

In a previous study [4], MID monitoring by mass spectrometry was performed for CH_4 and HCN (Figure 1). From these results, we distinguish two kinetic regimes of gas-particle interaction: a transient regime corresponding to the production and consumption of gases and correlated to the evolution of tholins solid particles, and a stationary regime where the gas mixture ratio is stabilized. In this study, the MID monitoring is carried out for gas-phase molecules suspected to participate to the tholins chemical growth (so called "precursors") : C_2H_2, C_2H_6, HC3N, C_2N_2.

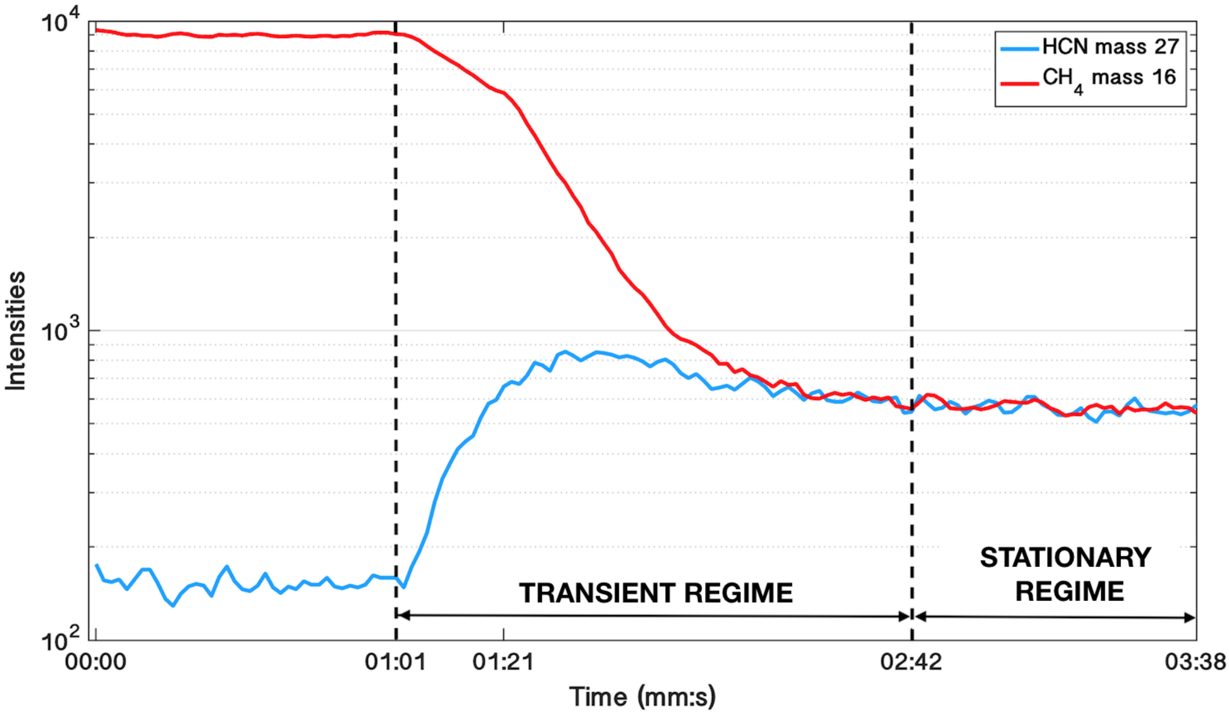

Figure 1 - Time evolution of the masses m/z 16 (CH_4), 27 (HCN), obtained with a mass spectrometer [4].

3.2 - Microphysical evolution by scanning electron microscopy

The samples were observed by scanning electron microscopy. The images show two growth phases, each corresponding to a gas-particle kinetic regime distinguished by the MID monitoring. Tholins during the transient regime exhibit nanoscale spherical monomers, not exceeding ~200 nm in diameter (Figure 2.A). Tholins formed in the stationary regime show an evolution of spherical monomers up to diameters of a few μm, and the formation of aggregates (Figure 2.B et 2.C).

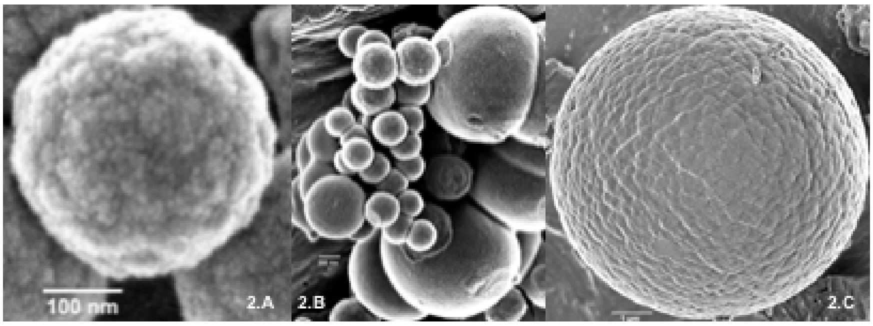

Figure 2- Morphologies of Titan's tholins obtained with SEM. Figure 2.A : Tholins formed during the transient regime have an average diameter of 200 nm. Figure 2.B : Evolution of spherical nanometric to micrometric particles. Figure 2.C : Tholins formed during the strationnary regime, have an average diameter of a few µm.

3.3 - Kinetic modeling of the gas-particle interaction

Based on a kinetic model performed by Pöschl et al. in 2007 [5], the two kinetic regimes observed in the experiment are fitted. From it, the absorption coefficient γ (uptake coefficient) of Titan tholins was deduced for each monitored precursor.. For each regime, an absorption coefficient γ is calculated taking into account the different interactions between gas-surface of the particles, as well as between surface-bulk of the particles, i.e. adsorption, desorption and diffusion effects.

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