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Laboratory light-scattering measurements with Titan's aerosols analogues produced by a dusty plasma

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ABSTRACT

The chemistry leading to the formation of solid aerosols (tholins) in Titan's atmosphere is simulated by a capacitively coupled plasma in a N_2–CH_4 gas mixture. The solid grains are produced in volume directly in the gas phase and studied ex-situ by SEM imaging and by light scattering on clouds of particles. The scattered light properties depend on the physical properties of the particles (morphologies, size distribution), as well as on the phase angle and the wavelength of the light. The particles may be aggregated or agglomerated grains. The grains size distribution is studied as a function of plasma parameters such as initial methane concentration introduced into the discharge, gas flow, absorbed RF power and plasma duration. The average grain size increases when the amount of CH_4 increases, when the gas flow decreases, and when the plasma duration increases up to a limit for each production condition.

For all the samples, the absorption decreases with increasing wavelength in the visible domain. As usually found for irregular particles, the polarization phase curves have a bell-shaped positive branch and a shallow negative branch. The maximum of polarization (P_{max}) increases when the average grain size decreases (sub-μm-sized grains). To obtain P_{max} values within the range of those measured in Titan's atmosphere; the average grains diameter has to be smaller than 100 nm, in agreement with the space observations results. In the light-scattering experiment, the size of the agglomerates in the clouds is in the 40–80 μm range in equivalent diameter. As a consequence P_{max} increases with decreasing wavelength due to the increasing absorption, in agreement with observations of Titan from outside the atmosphere.

1. Introduction

The atmosphere of Titan contains a large amount of solid aerosols (tholins) made of organic materials and induced by the photochemistry of nitrogen (N_2) and methane (CH_4), its major gaseous components. Their presence in the atmosphere gives its orange–brown color. Although the aerosols have been chemically analyzed in-situ by the ACP–GCMS instrument (Israel et al., 2005; Niemann et al., 2005), their physico-chemical properties are still not well known. Light-scattering observations (with linear-polarization measurements) have been made by different space probes: (i) outside the atmosphere with Pioneer 11 and Voyager 2 (Tomasko and Smith, 1982; West et al., 1983) and (ii) inside the atmosphere by Descent Imager Spectral Radiometer (DISR) experiment aboard the Huygens probe at altitudes lower than 140 km (Tomasko et al., 2005, 2008).

In order to interpret the light-scattering measurements different numerical models were developed to describe the solid particles structure. West and Smith (1991) have proposed aggregated grains to interpret the high polarization and important forward intensity in the observations. Cabane et al. (1993) proposed a fractal model. At high altitudes (350–400 km), the grains are small and form growing aggregates when settling down in the atmosphere. A vertical distribution can be inferred down to 80 km. Plane parallel and spherical radiative transfer models have been proposed (e.g. Salinas et al., 2003), with an increasing optical depth as the altitude decreases. Larger aggregates with open structures were introduced to satisfy DISR observations (Bar-Nun et al., 2008). The fractal aggregate model allowed Tomasko et al. (2008) to infer the aerosols size, shape and optical properties.

Experimental simulations to produce tholins have been developed. Photochemistry experiments in hydrocarbons mixtures were first used (Bar-Nun and Podolak, 1979). These tholins had a fractal structure. In nitrogen–methane gas mixtures, the most efficient method is to use a plasma discharge, for instance DC discharges (Coll et al., 1999) or inductively coupled plasma

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(Khare et al., 1984; Imanaka et al., 2004). In such experimental setups, tholins are deposited on reactors walls or on transparent plates inside the reactor (for optical measurements). Tholins could then have shapes and structures different from those of Titan's aerosols. However, such materials provide measurements of the refractive indices, of interest for further modeling (Khare et al., 1984; Ramirez et al., 2002).

Another way to produce tholins consists in using of a capacitively coupled plasma (CCP) discharge, which has been considered in the present paper. This technique is currently used in the so-called 'dusty plasmas' studies (Merlino and Goree, 2004). The 'PAMPRE' (French acronym for Production of Aerosols in Microgravity with a REactive Plasma) experiment devoted to produce analogues of Titan's aerosols in laboratory (Szopa et al., 2006). The produced tholins are formed in volume directly in the reactive plasma without any interaction with solid surfaces.

The organization of this paper is as follows. After a short description of the PAMPRE experiment, we try to correlate the dust physical properties (size of the grains, morphology), studied by field emission gun scanning electron microscopy (FEG-SEM) analysis, to the operating conditions. In a second step, we correlate their optical properties (linear polarization of scattered light measured on clouds of particles by the PROGRA^2 experiment) to their physical properties. Finally, we compare our results to those obtained on Titan's aerosols by Pioneer 11, Voyager 2 and Huygens space probes.

2. Samples production: PAMPRE experiment

A complete description of the experimental setup can be found in Szopa et al. (2006). The experiment works with a CH_4/N_2 gas mixture at room temperature. In order to produce sufficient amount of solid grains for analyses, gas is continuously injected through the polarized electrode. The CH_4/N_2 ratio can be changed from (2/98)% to (10/90)%. Residual gases in the experiment are continuously pumped out with a rotary van vacuum pump. The total gas flow, calculated for normal temperature and pressure conditions, is in the range 10–55 sccm (standard $cm^{-3}\,mn^{-1}$). It is possible to change the pressure from 10 to 1000 Pa (about 100 Pa for the present series of experiments). Out of this pressure range, the plasma cannot be switched on. The absorbed RF power for the presented results is in the 10–50 W range. A cylindrical grid cage confines the plasma.

Electron collisions dissociate N_2 and CH_4 molecules in the plasma. The dissociation products react to form HCN, which is supposed to be a precursor of tholins (Pintassilgo et al., 1999) and other chemical gaseous species. The energy of electrons is in the range of a few electron volt, i.e. the same energy range as solar photons. As the electrodes in the experiment are distant of a few centimeters, a pressure of 100 Pa is necessary to obtain a sufficient amount of electron collisions in the discharge to generate an efficient chemistry. Such a pressure is representative of a few meters thickness in Titan's stratosphere. The gaseous chemistry, taking place in the plasma, produces complex gaseous species, and also solid organic particles, which are considered as analogues of Titan's aerosols or 'tholins'.

In the plasma discharge, the solid organic particles are electrically charged and experience different forces: electrostatic forces, neutral gas drag forces and gravity (Bouchoule, 1999). During their formation, they are maintained in levitation by the electrostatic forces. When they have grown and reached a critical size, the gas drag forces become predominant and expel them out of the plasma. They are ejected through the grounded grid and collected in a glass vessel to be analyzed ex-situ.

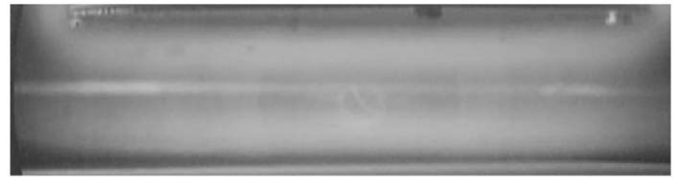

Fig. 1. PAMPRE experiment: plasma and laser beam scattered by dust particles between the electrodes.

Table 1
Operating conditions and size distribution of the grains.

Samples number	Gas mixture (%)	Total gas flux (sccm)	Absorbed power (W)	Plasma duration	Mean grain size (nm)	Largest detected grain (nm)
1	$2CH_4+98N_2$	55	30	CWC	315±185	1400
2	$\mathbf{4}CH_4+\mathbf{96}N_2$	55	30	CWC	300±60	600
3	$\mathbf{6}CH_4+\mathbf{94}N_2$	55	30	CWC	270±130	1000
4	$\mathbf{10}CH_4+\mathbf{90}N_2$	55	30	CWC	595±390	2500
5	$2CH_4+98N_2$	55	**50**	**CWC**	375±115	1250
6	$2CH_4+98N_2$	55	**10**	**CWC**	210±85	700
7	$2CH_4+98N_2$	**10**	30	CWC	1070±440	6000
8	$2CH_4+98N_2$	**27.5**	30	CWC	720±130	1000
9	$2CH_4+98N_2$	55	30	**10 s**	90±30	140
10	$2CH_4+98N_2$	55	30	**20 s**	105±85	240
11	$2CH_4+98N_2$	55	30	**30 s**	135±80	240
12	$2CH_4+98N_2$	55	30	**40 s**	200±90	340
13	$2CH_4+98N_2$	55	30	**50 s**	207±30	380
14	$2CH_4+98N_2$	55	30	**90 s**	300±80	600
15	$2CH_4+98N_2$	55	30	**110 s**	280±100	520

Sample 1 is considered in the present study as the reference. The parameters, which have changed as compared to sample '1' conditions, are in bold. Pressure about 100 Pa.

Apertures are placed all around the reactor. One is used for in-situ study of the plasma by optical emission spectroscopy (OES) in UV–visible range (Szopa et al., 2006). The levitating dust is illuminated by a laser beam (532 nm) and the scattered light is observed through a large window. For this purpose, the metallic cage and the glass vessel are removed (Fig. 1).

The plasma can work in continuous working conditions (CWC) or in pulsed mode. In CWC, the plasma is turned on during 8–24 h depending on the production conditions in order to get a sufficient amount of materials. Gas is continuously injected and tholins are continuously produced. In pulsed conditions, the plasma duration is limited in order to stop the tholins growth. The time interval between two pulses is a few seconds, allowing the gas renewal. Eight samples were prepared in CWC. Seven other samples were produced with duration in the 10–120 s range. The different parameters in the operating conditions for each sample are described in Table 1.

3. Tholins morphology

We use the word 'grain' for individual entity (called monomers in Tomasko et al., 2005, 2008) as found on SEM images, the word 'aggregate' is used for bond grains, the word 'agglomerate' is used in the light-scattering experiment when grains and aggregates stick together and are lifted at the same time in a small limited volume. The word 'particle' is more general.

Tholin grains have a quasi-spherical shape, a rough surface, and sometimes form aggregates with touching or sintered constituent grains. Fig. 2 presents SEM images of some samples with different morphologies of grains and aggregates that can