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Optical constants of Titan's tholins produced in a RF plasma discharge in different N_2-CH_4 concentrations

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Abstract

The different numerical models that have been developed to explain the characteristics and properties of the Titan's haze require knowledge of optical constants of Titan's aerosols. Here, we present the optical constant characterization of Titan aerosol analogues or "tholins" produced with the PAMPRE experiment. Tholins have been produced in different N_2–CH_4 gaseous mixtures to discuss the effect of the initial methane concentration on the optical constants. The real (n) and imaginary (k) parts of the complex refractive index have been determined using the spectroscopic ellipsometry technique.

1. Introduction

The Cassini-Huygens mission has detected haze layers in the upper atmosphere of Titan. In Titan's atmosphere, the dissociation of N_2 and CH_4 by solar UV radiation and Saturn's magnetosphere electron bombardment induces a complex organic chemistry that results in the production of solid aerosols present from the lower stratosphere up to the upper atmosphere. This haze is one of the main drivers of the radiative transfer in the atmosphere of Titan [6] and strongly influences its thermal structure [4]. For these reasons, determining the optical properties of the haze is critical in the analysis of the observational data. As no return sample is planned from Titan's atmosphere at short term, the use of laboratory experiments is of primary importance to produce analogues of Titan's aerosols. Indeed, the study of these analogues and of their way of production enables to provide important information on the processes taking place in the Titan's atmosphere.

In a recent study [7], we have shown that the C/N ratio in tholins increases with the initial CH_4 concentration. So a change of the optical constants as a function of initial CH_4 concentration is expected. Moreover, knowing that in Titan's atmosphere, the CH_4 concentration varies with altitude and was varied during the evolution of this atmosphere [8], it is of utmost importance to determine the optical constants of tholins synthesized from N_2-CH_4 gas mixtures with different CH_4 concentrations. We present here a first study of optical properties of tholins synthesized from gas mixtures with four different CH_4 concentrations (1, 2, 5 and 10%) .

2. Tholin film production

The PAMPRE experimental setup has been described in detail in a previous publication [7]. In the PAMPRE reactor, the tholin production results from the chemistry induced by electron impact in different N_2-CH_4 gas mixtures using a low pressure radiofrequency capacitively coupled plasma (RF CCP) discharge. The RF CCP discharge is driven between two electrodes with a continuous total gas flow rate giving a constant pressure of 0.9 mbar during the tholin production. In the study presented here, tholin thin films were deposited onto two different substrates: a Pyrex substrate covered with an aluminum layer and a SiO_2 layer (Al-SiO_2) and a CaF_2 substrate. The substrates were placed inside the plasma at the center of the grounded electrode. A 2 hour-long deposition time was chosen in order to produce tholin films no more than ~1 µm thick. With this production protocol, thin, homogeneous tholin films were produced that allowed the use of spectroscopic ellipsometry measurements and analysis for the determination of the tholin film refractive index.

3. Spectroscopic ellipsometry analyses

Spectroscopic ellipsometry is a technique that measures the change in polarization of the light reflected from a material: from a linearly polarized incident beam to an elliptically polarized reflected beam. The polarization change is represented as the amplitude ratio, Ψ, between the parallel and perpendicular, respectively, components of the polarized light, and their phase difference, Δ. The ellipsometry parameters Ψ and Δ depend on the number of layers, their thicknesses, their structure and the wavelength. In order to extract useful information about a sample (thickness and optical constants of the film) the experimental ellipsometry data are compared with the data generated by a multilayer model which describes the structure of the sample and its optical response [1].

Figure 1 presents the n and k values of the tholins synthesized from a 95-5 N_2-CH_4 gas mixture and deposited onto an Al-SiO_2 substrate. The values of n decrease from n = 1.64 (at 370 nm) to n = 1.57 (at 900 nm), and imaginary part k ranges from $k = 1.67\times10^{-2}$ (at 370 nm) to $k = 1.57\times10^{-3}$ (at 640 nm). The shapes of the n and k curves are in agreement with previous studies of the optical constants of Titan aerosol analogues (see Table 1) [2,3,5]. All experiments compared here were done at room temperature and covered the 370-900 nm range (at least) except Hasenkopf et al. 2010 who obtained values of n and k at 532 nm only. Some differences in terms of n and k values are present, these differences could be due to the different experimental methods for tholins production which could change their composition.

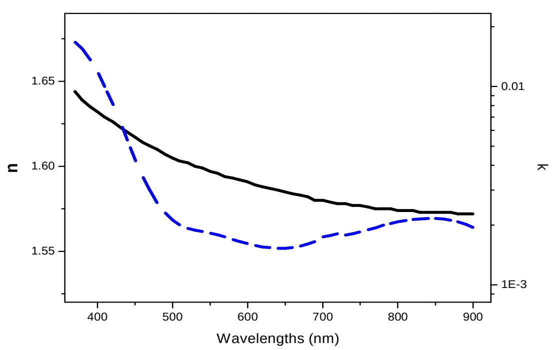

Figure 1: Real, n, (solid line) and imaginary, k, (dashed line) parts of the complex refractive index in the UV-Vis region. Tholins were produced from a 95-5 N_2-CH_4 gas mixture and deposited onto an Al-SiO_2 substrate.

Table 1: Comparison with other tholin studies. Minimum and maximum values obtain in the 370-900 nm range.

Reference	Gas mixture	Energy source	n	k ($\times10^{-2}$)
Khare 1984	90:10 N_2:CH_4	Plasma (DC)	1.72-1.66	9.0 - 0.11
Ramirez 2002	98:2 N_2:CH_4	Plasma (DC)	1.60-1.53	1.00 - 0.2
Hasenkopf 2010	99:1 N_2:CH_4	UV lamp	1.35	2.1
This work	95:5 N_2:CH_4	Plasma (RF)	1.64-1.57	1.60 - 0.15

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