

Near-infrared spectra of liquid/solid acetylene under Titan relevant conditions and implications for Cassini/VIMS detections



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ABSTRACT

Acetylene is thought to be abundant on Titan according to most photochemical models. While detected in the atmosphere, its likely presence at the surface still lacks physical evidence. It is thought that solid acetylene could be a major component of Titan's lakes shorelines and dry lakebed, detected as the 5 μm -bright deposits with the Cassini/VIMS instrument. Acetylene could also be present under its liquid form as dissolved solids in Titan's methane–ethane lakes, as emphasized by thermodynamics studies. This paper is devoted to the near-infrared spectroscopy study of acetylene under solid and liquid phases between 1 and 2.2 μm , synthesized in a Titan simulation chamber that is able to reproduce extreme temperature conditions. From experiments, we observed a $\sim 10\%$ albedo increase between liquid acetylene at 193–188 K and solid acetylene at 93 K. Using the NIR spectroscopy technique we successfully calculated the reflectivity ratio of solid/liquid acetylene as 1.13. The second difference we observed between liquid and solid acetylene is a shift in the major absorption band detected at 1.54 μm , the shift of $\sim 0.01 \mu\text{m}$ occurring toward higher wavelength. In order to assess the detectability of acetylene on Titan using the Cassini/VIMS instrument, we adapted our spectra to the VIMS spectral resolution. The spectral band at 1.55 μm and a negative slope at 2.0 μm falls in the Cassini/VIMS atmospheric windows over several VIMS infrared spectrals, thus Cassini/VIMS should be able to detect acetylene.

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1. Introduction

Saturn's major moon, Titan, possesses a heavy atmosphere mainly composed of nitrogen N_2 (95–98%), methane CH_4 (1–5%) and various minor species in trace, e.g., carbon monoxide CO, dihydrogen H_2 , argon ^{40}Ar , and ethane C_2H_6 (Fulchignoni et al., 2005; Niemann et al., 2005, 2010; Coustenis et al., 2007; Dekok et al., 2007; Waite et al., 2007; Owen and Niemann, 2009). At Titan's surface pressure and temperature conditions (1.46 bar, 93.7 K (Fulchignoni et al., 2005)), methane is close to its triple point, such as water is on Earth. Methane therefore probably composes the numerous polar lakes discovered by the Cassini mission (Stofan et al., 2007; Hayes et al., 2008). According to most photochemical models (Lavvas et al., 2008), methane photodissociation in the atmosphere primarily produces ethane and propane, which would exist as more stable liquids due to their lower saturation vapor

pressure (Fig. 1). Thermodynamical models predict their presence in Titan's lakes as a major component (Cordier et al., 2009, 2013; Glein and Shock, 2013; Tan et al., 2013) and has already been confirmed thanks to its detection by the Cassini/VIMS instrument in Ontario Lacus (Brown et al., 2008).

Photochemical models also predict the production of numerous other hydrocarbons and nitriles in the atmosphere, which would exist under their solid form at the surface (Lavvas et al., 2008). These compounds have been detected in Titan's atmosphere either by *in situ* measurements performed by the Huygens/GCMS instrument (Niemann et al., 2005, 2010), or the Cassini/INMS and CIRS instruments (Waite et al., 2007; Coustenis et al., 2007; Dekok et al., 2007; Coustenis and Hirtzig, 2009; Vinatier et al., 2010). Potential surface detections of some of these hydrocarbons in VIMS data have been reported by Clark et al. (2010), notably for benzene, ethane or methane.

Acetylene is also produced in the upper atmosphere in solid form via photolysis effect of methane molecule, which then settles onto the surface. It has been thought that over geological time,

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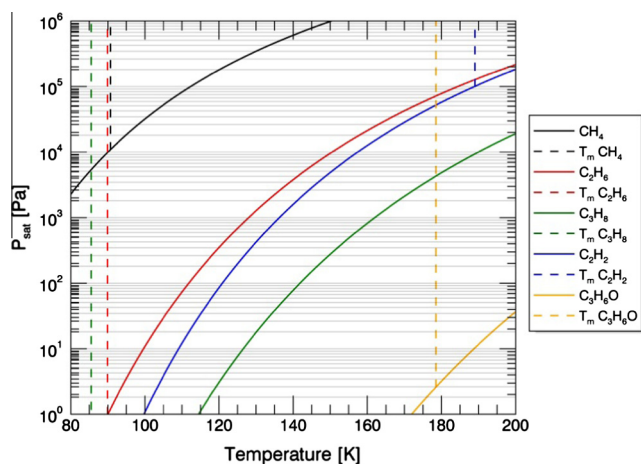


Fig. 1. Phase diagram of methane (CH_4), ethane (C_2H_6), propane (C_3H_8), acetylene (C_2H_2) and acetone ($\text{C}_3\text{H}_6\text{O}$). T_m represents the melting temperature at melting point. The vapor pressure data comes from the ChERIC database.

several hundred of meters of thick acetylene layer may have accumulated on Titan. According to most photochemical models on Titan's atmosphere and surface interaction predicts acetylene to be most abundant in the atmosphere. It is predicted that acetylene is ~ 125 – 2000 times larger than any other compound in the atmosphere such as benzene (second largest compound produced in the atmosphere as predicted by same photochemical models, Clark et al., 2010). Although predicted to be among the most produced hydrocarbons in the atmosphere (Lavvas et al., 2008) and therefore probably one of the most abundant at the surface, acetylene (C_2H_2) has only been detected as a trace compound at the Huygens Landing Site by the Huygens/GCMS instrument (Niemann et al., 2010) and Clark et al. (2010) reported the absence of acetylene detection in VIMS data on Titan's surface, based on the VIMS $5 \mu\text{m}$ and 2.7 – $2.8 \mu\text{m}$ atmospheric window observations. Acetylene, which is evaluated by various thermodynamical models to be soluble in liquid hydrocarbons under Titan surface conditions (Cordier et al., 2009, 2013; Glein and Shock, 2013; Tan et al., 2013), could also exist as a dissolved solid in Titan's lakes and seas, i.e., implying a phase change to a "liquid-like phase". Acetylene is thought to be highly soluble in both methane and ethane and if acetylene is abundant on the surface (as predicted by various photochemical models) it should dissolve in methane and ethane and can be present in liquid form in the lakes and oceans. Now, if in future we detect acetylene in oceans it may exist in its liquid form.

Given this context in which acetylene could be present on Titan's surface under possibly different forms, we present the infrared spectra of liquid and solid acetylene acquired between 1 and $2.2 \mu\text{m}$ under Titan's surface relevant conditions (90 K , 1.5 bar of N_2), thus completing the portion of the acetylene spectrum shown for the $5 \mu\text{m}$ window in Clark et al. (2010). The measures are realized using the Titan Module (Wasiak et al., 2013), primarily designed to study the stability of liquid methane and ethane under Titan simulated conditions (Luspay-Kuti et al., 2012). Since the spectrum acquired covers 4 of the VIMS atmospheric windows in which Titan's surface can be seen (Sotin et al., 2005) and that has not been yet investigated (Clark et al., 2010), we also aim at constraining the detectability of acetylene on Titan at the Cassini/VIMS instrument spectral resolution.

2. Experimental setup

To perform these measurements, we use the Titan Module developed at the University of Arkansas (Wasiak et al., 2013; Luspay-Kuti et al., 2012; Singh et al., 2014). The detailed descrip-

tion of the chamber setup is given in Wasiak et al. (2013). The Titan module is composed of three main parts: a condenser, a Temperature Control Box (TCB) and a data acquisition unit. The TCB and the condenser are surrounded by cooling coils in which liquid nitrogen is flowing to reach Titan temperatures. The condenser is located inside the module and is cooled to reach condensation temperatures of the gas samples introduced in the system. The samples are then collected into a Petri dish of 15 cm in diameter through a solenoid valve controlled manually, located right beneath the condenser. Thermocouples are scattered in several parts of the chamber in order to record and control the temperature in different locations inside of the TCB and of the condenser.

The Titan Module is equipped with several measurement systems. The primary agenda for the chamber is to perform various thermodynamic measurements on Titan hydrocarbons, including evaporation measurements (Luspay-Kuti et al., 2012). It is thus equipped with a balance, located right above the Titan Module, continuously monitoring the mass of the samples during the experiment. It is also equipped with a fiber optics operating from 1.0 to $2.4 \mu\text{m}$, connected to a Nicolet FTIR 6700 Smart Diffuse spectrometer (with a spectral resolution of 4 cm^{-1}), which allows to measure infrared diffuse reflectance spectra of the samples under Titan simulated conditions. The fiber optics is located vertically right above the Petri dish, which contains a piece of Spectralon® material inside for reference. The light ray sent to the sample pan is directional and vertical as the fiber probe is right above the sample pan. We measure the light returned to the FTIR with the same fiber probe that implies bidirectional, with the very special specular configuration where $i = e = g = 0$. A camera is located inside of the TCB to provide real-time observation into the chamber. Before each experiment, the chamber is purged by N_2 gas during several minutes in order to get Titan's atmospheric main composition and build the pressure up to 1.5 bar . The N_2 flow is then regulated during the experiment to keep a constant pressure inside the chamber.

3. Infrared spectrum of acetylene

3.1. Background in acetylene infrared spectroscopy

The first acetylene infrared spectroscopy has been performed by Bell and Nielsen (1950) and Allen et al. (1956), who noticed the presence of several absorption bands in the spectra at 3 and $7.5 \mu\text{m}$. The main absorption band regions in the infrared spectrum of acetylene, located at 3 , 5 , 7.5 and $13.6 \mu\text{m}$ (Hirabayashi and Hirahara, 2002; Khanna, 2005; Wang et al., 2009), have been reported in the work of Jacquemart et al. (2003) in order to be incorporated in the HITRAN (High resolution Transmission) database. In order to address the acetylene behavior in the infrared, Curchin et al. (2008) showed a continuous infrared spectrum in reflectance of a cryogenic acetylene powder acquired between 0.35 and $15.5 \mu\text{m}$ at 80 K . This spectrum seems to display numerous absorption features in the VIMS wavelengths range between 0.9 and $5.1 \mu\text{m}$. Clark et al. (2010) also showed portions of the infrared reflectance spectrum of acetylene (mixed with carbon black), acquired at 90 K , at wavelengths covering the VIMS $5 \mu\text{m}$ window. These portions display absorption features located at $4.854 \mu\text{m}$ (at the very beginning of the window, still affected by the decreasing atmospheric absorptions of CO and N_2 toward long wavelengths (Clark et al., 2010)) and at $4.936 \mu\text{m}$. Reporting acetylene absorption features at shorter wavelengths, such as those investigated in the present paper, could provide additional options to perform a future detection in spectral windows with a higher S/N ratio than at $5 \mu\text{m}$ (Sotin et al., 2012), but unfortunately with a stronger atmospheric influence (Rodriguez et al., 2006) to be accurately accounted for.

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