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Formation of NH_3 and HCN in atmospheric glow discharge fed by gaseous CH_4-N_2 mixture

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Abstract

The formation of products produced in an atmospheric glow discharge fed by a N_2-CH_4 gas mixture has been studied using Fourier transform infrared (FTIR) and optical emission spectrometry (OES). The measurements were carried out in a flowing regime at ambient temperature and pressure. FTIR-measurements reveal that HCN and NH_3 are the major products in such a discharge with traces of C_2H_2. These same molecules have been detected in Titan's atmosphere and the present experiments may provide some novel insights into the chemical and physical mechanisms prevalent in Titan's atmosphere with these smaller species believed to be the precursors of heavier organic species in Titan's atmosphere and on its surface.

1. Introduction

The Cassini-Huygens space mission to Saturn and the release of its Huygens probe onto its largest moon, Titan, has led to a wealth of data on the atmospheric and surface composition of Titan, presenting us with a set of unexpected results including the observation of hydrocarbon lakes – the first liquid 'seas' on a solar system body outside the Earth; and the observation of anions in the upper atmosphere (ionosphere) [1]. In order to understand the physical and chemical processes leading to such observed phenomena additional laboratory simulations are required.

The dense atmosphere of Titan is mostly composed of N_2 with a few percent of CH_4. The most important minor compounds detected by Cassini Huygens are nitriles (HCN, HC_3N, HC_5N, C_2N_2) believed to be formed by as a result of dissociation of nitrogen and methane either by solar induced photolysis or by electron impact [2] and hydrocarbons (C_2H_2, C_2H_4, C_2H_6, C_3H_8, C_3H_4 [2]). The presence of clouds and strong convective motions are a particular feature of Titan's lower atmosphere. Charged particles, originating from the Saturnian magnetosphere, can accumulate on droplets within the clouds of the troposphere. Neutralization of these charged particles may lead to corona discharges within the clouds which can induce chemical reactions in the troposphere [2]. Therefore, electrical discharges are believed to be the best environment for the study of electron-molecule and ion-molecule reactions [3-5] within Titan's atmosphere.

2. Experiment

The apparatus used in these experiments is shown schematically in Figure 1. The reactor was connected to the long path IR gas cell equipped with KCl windows and placed in a Nicolet Nexus FTIR spectrometer. OES using Jobin Yvon TRIAX 550 spectrometer with CCD detector was used to monitor the optical emission from the N_2/CH_4 plasma. The flow rates through the reactor for both CH_4 and N_2 were regulated using MKS mass flow controllers. All measurements were carried out in flowing regime with a total flow rate of 200 sccm. The discharge electrode system had the standard configuration of a classical gliding arc, a pair of stainless steel holders positioned in parallel to the iron electrodes but in this case the plasma was not gliding due to the low flow rate and therefore stable abnormal glow plasma occurred between the electrodes at their shortest distance of 2 mm, thus forming plasma channel with diameter of 1mm. With increasing current (15-40 mA) the voltage was slightly decreasing from 400 V to 350 V. Electrical parameters have been measured by Tektronix oscilloscope using high voltage probe and

10 Ω resistor for current measurement. The reactor chamber had a volume of 0.3L. The discharge was powered by a home-made DC HV source. The present experiments were performed for different N_2:CH_4 ratios in range from 0.5 % to 2% CH_4 in N_2 at atmospheric pressure. A pure CH_4/N_2 gas mixture without plasma was measured as the background spectra for FTIR measurements.

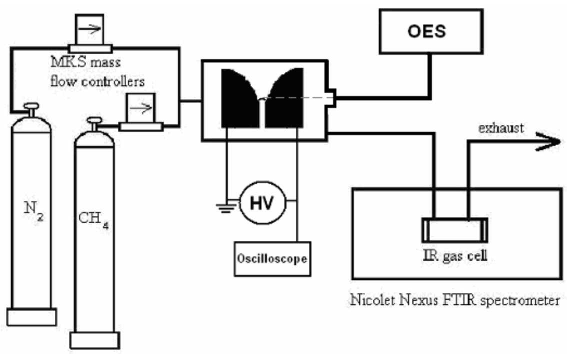

Figure 1: Schematic diagram of the experimental apparatus.

3. Results

We present the results of both a FTIR and OES study of the gaseous products and radicals formed in the glow discharge fed by four different atmospheric pressure mixtures of N_2:CH_4 (0.5, 1, 1.5 and 2 % CH_4) operated in a flowing regime at different discharge currents (from 15 up to 40 mA) and at ambient temperature. FTIR analysis of the gaseous products showed that HCN, C_2H_2, NH_3 are the main products in our CH_4/N_2 glow plasma. The yields of these compounds are such that HCN > NH_3 > C_2H_2. The discharge current has a significant effect on the product synthesis and electron density (Figure 2). The continuous glow discharge was the hottest one with gas temperatures (Tg) reaching 3200 K, since most of electron energy is expended in gas heating. The plasma was close to local thermodynamical equilibrium conditions but not close enough to assume Maxwell distribution. Using OES study we estimated vibrational and rotational temperatures T_r and T_v which - together with the electrical parameters - allowed us to calculate the current and electron number densities in the discharge with typical values of 1.9-5.1 A/cm^2 and n_e ~ 1013 cm^{-3}.

Such experiments can provide information that will aid our understanding of the physical and chemical processes in Titan's atmosphere. Within the discharge we observed the formation of the same compounds as observed in Titan's atmosphere by the Huygens Surface Package and by Cassini Observer. Furthermore discharges can provide relevant information on the formation of the anions [1] and allow some of the anions observed by Cassini to be identified. However, we note that different discharges have different sources of ions/excited molecules and thus in discharges as a simulation mimic it is necessary to carefully define the plasma conditions and their relevance to specific regions of Titan's atmosphere.

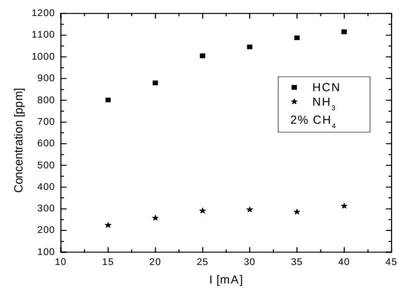

Figure 2: Dependence of NH_3 and HCN concentration on discharge current.

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