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The Influence of Admixtures on the Composition of Products by Nitrogen-Methane Atmospheric Glow Discharge

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This work extends our experimentally studies with simulation of Titan's atmosphere by atmospheric glow discharge. This work is devoted to estimate the influence of CO₂ and/or CO on reactivity in the Titan's atmosphere. The exploration of planetary atmosphere is being advanced by the exciting results of the Cassin-Huygens mission to Saturn and Titan, its most famous moon. Most of the studies were mainly interested in the reactivity of the N₂-CH₄ gaseous mixture and with the primary products of reactions, but the atmosphere of Titan also contains oxygenated volatile species.

Keywords: Titan's atmosphere, atmospheric glow discharge, FTIR spectroscopy

1 INTRODUCTION

Atmospheric pressure glow discharges are of significant interest for a wide range of applications such as pollution control, material processing or surface treatment. Among the many different types of atmospheric pressure discharges the DC glow discharges is one of the most deeply studied [1, 2]. The gliding arc configuration has been shown to be a good mimic of planetary atmospheres [3] being used to replicate physical and chemical conditions in Titan. Titan is the largest moon in Saturn's lunar system and the only one with a dense atmosphere-atmospheric pressure is approximately 1.5×10^5 Pa [4-6]. It is the only lunar body with a substantial containing significant quantities of methane (CH₄) and nitrogen (N₂) in its atmosphere [7]. Chemical processes in Titan's atmosphere are therefore able to create complex molecules containing C, N and H. This makes Titan our only planetary-scale laboratory for the synthesis of complex organics molecules [8]. In Titan's atmosphere, the dissociation of N₂ and CH₄ by solar ultraviolet radiation initiates chemical reactions that result in the formation of complex organic molecules.

The present work is focused on the experimental study of gaseous products produced in an atmospheric pressure glow discharge fed by

N₂-CH₄ gas mixtures with CH₄ concentrations in the range from 1 % to 4 % plus admixture of carbon dioxide (CO₂) from 1% to 4 %. The gaseous products were analyzed by Fourier-Transform-Infra-Red spectroscopy (FTIR).

2 EXPERIMENTAL APPARATUS

A simplified schematic drawing of the experimental set up is presented in Fig. 1. An atmospheric pressure DC glow discharge was created between two stainless steel electrodes separated by a 2 mm gap. The electrodes system had standard configuration of the gliding arc discharge but due to low applied power and low gas velocity the discharge is not moving along the electrodes. The discharge was formed in the stable abnormal glow regime with plasma channel of 1 mm in its diameter. The stainless steel vacuum chamber (volume of 1 litre) was evacuated by a rotary oil pump to maintain an oxygen free system. The discharge was operated with an applied power in range 4 to 15 W in pure nitrogen enriched by 1-4 % of CH₄ with admixture of 1-3% of CO₂ total flow rate of 200 sccm. The flow rates through the reactor for both methane and nitrogen were regulated using MKS mass flow controllers. The exhaust gas was analyzed in-situ by FTIR spectroscopy.

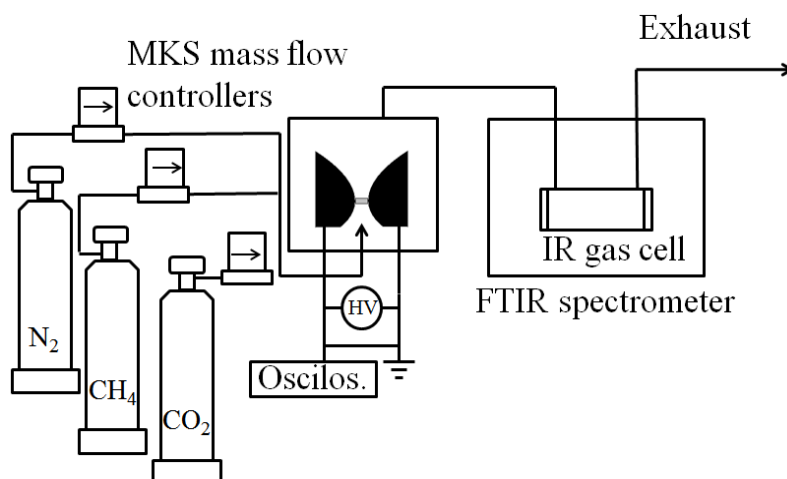


Fig.1: Schematic diagram of the experimental setup used for FTIR analysis of gaseous products of a DC glow discharge fed by various mixtures of N_2 - CH_4 - CO_2 .

3 RESULTS

A typical FTIR spectrum revealing the products formed in the nitrogen discharge fed by 2% of CH_4 and 1% of CO_2 is shown in Fig. 2. Similar spectra were observed for other N_2 - CH_4 - CO_2 molar ratios. The HITRAN spectral data individual absorption features was used for recognizing of specific compounds. Hydrogen cyanide (HCN) was found to be the most abundant product at wavenumbers of 1430 cm^{-1} and of 720 cm^{-1} . The other major

products were ammonia (NH_3) -966 cm^{-1} and acetylene (C_2H_2) -729 cm^{-1} as well as carbon monoxide (CO) and water. These products were recognized in all N_2 - CH_4 - CO_2 gas mixtures. The products concentrations are strongly depended on compositions of the gas mixture. Increasing the initial CH_4 concentration from 2% to 4% and also increasing admixture from 1% to 3% of CO_2 lead to increase in the products yield.

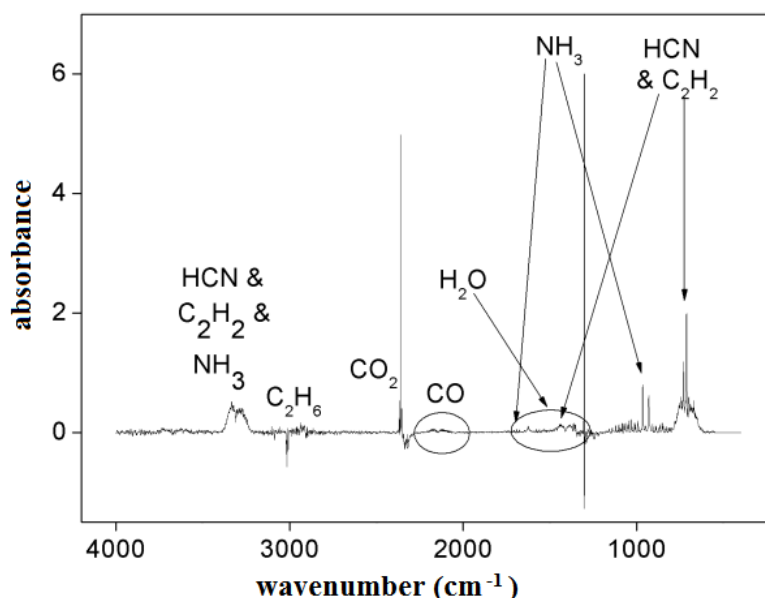


Fig.2: Sample spectrum of analysed products formed in a gas mixture 2% of CH_4 + 1% of CO_2 in N_2 .

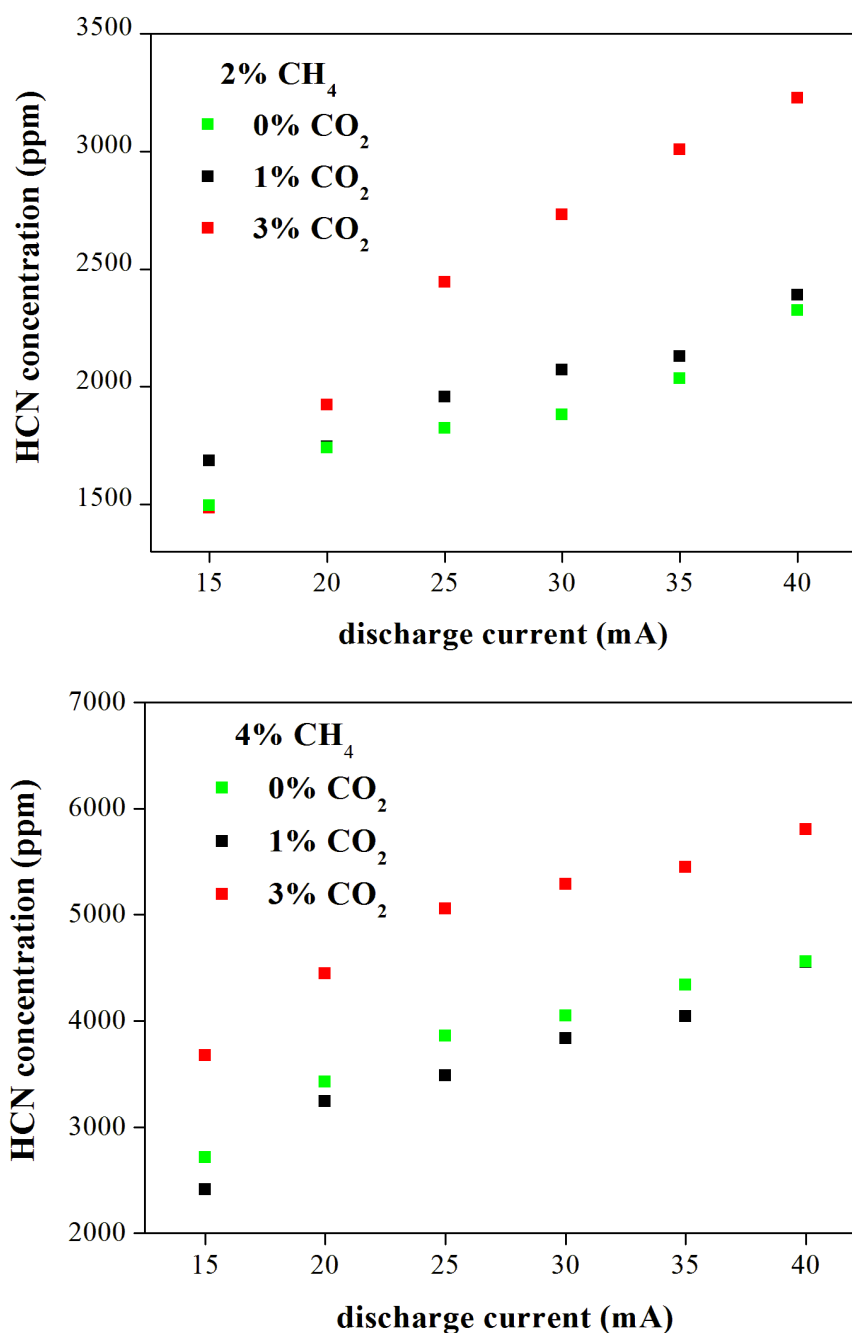


Fig.3: Concentration of hydrogen cyanide dependence on initial concentration of methane and also admixture of carbon dioxide.

Fig. 3 shows the quantitative analysis of HCN at 1430 cm^{-1} formed under different experimental conditions. Since these experiments have been carried out for different CH₄-N₂ mixture ratios and the same gas flow rates, the evolution of different product concentrations was found to depend strongly on the initial gas ratios. The dependence of product concentrations is strongly dependent on the different CH₄-N₂ gas mixtures. On the other hand it was proofed [9] that the kinetics of HCN formation

slows down in presence of CO, showing an inhibiting role of CO on HCN formation. In our experiments this influence was not recognize.

4 CONCLUSION

The gaseous phase products formed in the atmospheric glow discharge fed by different mixtures of N₂ plus CH₄ (2 % and 4 %) and admixture of CO₂ (1 and 3%) were determined by in situ FTIR analysis. The discharge was

operated in the flowing regime at different discharge currents at laboratory temperature. In situ FTIR technique for the exhaust gas phase sampling was successfully used for chemical analysis to deduce the gas composition in the N₂-CH₄-CO₂ reactive gas mixture mimics of Titan's atmosphere.

Various nitrile compounds and hydrocarbons were observed in all experiments. HCN was identified as the major gas phase product in all of measurements. Others minor products detected were C₂H₂, NH₃, CO₂, CO and also some nitrile oxides. These results are consistent with the Titan's atmospheric composition because the same compounds were detected during the Cassini-Huygens space mission. This fact clearly demonstrates that laboratory experiments can be used for prediction of both the presence and possible concentrations of compounds which have not been detected, yet. These simple organics should be tracers of the chemical groups constituting the dusty products.

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