EPSC Abstracts
Vol. 10, EPSC2015-409, 2015
European Planetary Science Congress 2015

Determination of the effect of ammonia on clathrate hydrates at Titan' surface-subsurface conditions

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Abstract

The development and combination of two facilities, a cryogenic calorimeter and a cryostage coupled with a Raman spectrometer, allows for detailed studies of the chemical reactions/phase transformations that may take place in the upper layer of Titan's crust and the subsequent exchange processes with the atmosphere due to phase transitions related to the composition in icy materials and the thermal state of the upper few km.

1. Introduction

Titan is the only icy satellite known so far with a dense atmosphere, in which a hydrocarbon cycle (similar to Earth's hydrological cycle) between the atmosphere, the surface and the subsurface occurs [1]. Several data obtained in Cassini-Huygens mission, such as the high methane concentration and the origin of molecular nitrogen at the atmosphere, can hardly be explained by the current models [2, 3]. In order to resolve these uncertainties, experimental simulation of the surface and the subsurface is required, since the geochemistry in this zone is directly related with the atmospheric composition. In this work we focus in the "methane question". It is known that the surface is likely dominated by water ice I_h and ethane (C_2H_6)- methane (CH_4)-clathrate hydrates, beneath the ice shell is expected an ocean with dissolved ammonia (NH_3) [4]. NH_3 affects deeply the equilibrium regimes of the solid minerals mentioned, causing a depression in the melting temperatures. One plausible explanation for the methane amount observed is that a replenishment mechanism exists from the clathrate dissociation. Previous works have been performed on this regard, observing a temperature decrease in clathrate melting even around 75 K [5,6], but more experiments are necessary to pinpoint the geochemical processes that should be occurring. The study of the system H_2O-THF-NH_3 with the techniques mentioned lead a better understanding of the NH_3-H_2O interactions at clathrate conditions. Once stablished this effect we will be able to analyze with accuracy the system H_2O-CH_4-NH_3.

2. Experimental setup

With the Setaram BT 2.15 Calvet calorimeter, coupled with the Residual Gas Analyzer mass spectrometer, it is possible to run tests in a temperature range from 77 to 470 K and a pressure range from 0.1 to 10 MPa. While the Linkam CAP500 cryostage, which operates between 77 and 570 K at a gas pressures up to 20 MPa, allows the monitoring of the runs via Raman spectroscopy, using the Horiba Jobin-Yvon LabRam confocal dispersive spectrometer.

3. Expected results

The combination of calorimetry and spectroscopic analysis allows for a detailed study of the clathrare hydrates system. Inside the pressure-temperature zone of study (i.e. 0.1 – 10 MPa and 77 – 295 K), we are able to determine what chemical reactions take place, the temperature at which they occur, the character (endo/exothermic) and the energy required in these reactions, and the molecular environment changes during the processes.

Phase transitions, energy exchanges and eutectic, peritectic and *liquidus* temperatures have been already determined in perfect agreement with previous works [6, 7, 8] of the systems H_2O-NH_3, H_2O-THF and H_2O-THF-NH_3 at a NH_3 concentration range from 0 to 25 wt% and 0.1 MPa. Additional observations not observed before are being also assessed. Future runs under pressure of the system H_2O-CH_4-NH_3 will be conducted early.

Acknowledgements

This research was supported by an appointment to the NASA Postdoctoral Program at the Jet Propulsion Laboratory, California Institute of Technology, administered by Oak Ridge Associated Universities through a contract with NASA. Support from the NASA Outer Planets Research program and government sponsorship acknowledged.

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