

First steps of neutral hydrocarbon cluster formation in Titan's atmosphere: a laboratory kinetics approach

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Abstract

Titan's dense atmosphere is the siege of a complex photo-chemistry initiated by the dissociation of its two most abundant components, nitrogen N_2 and methane CH_4 . This cold chemistry generates a plethora of hydrocarbons and nitriles and takes part in the production of a thick orange haze. The detection of heavy neutrals and positive ions in Titan's upper atmosphere by the Ion and Neutral Mass Spectrometer (INMS) embarked onboard Cassini [1] and measurements of significant amounts of negative ions with masses up to 13 000 amu with the Cassini Plasma and Electron Spectrometer (CAPS/ELS) [2] suggest that low temperature chemical reactions and physical processes occurring at high altitudes near 1000 km could be the haze source. This haze material could act as a nucleus for the condensation of organic vapors in Titan's stratosphere and troposphere.

In any event, evidences to support this hypothesis are still partial. The pathways leading to the formation and growth of haze aerosols remain far to be well understood. From a chemical viewpoint, for instance, the low temperature kinetics and the nature of the products of negative ion-molecule reactions are poorly known. From a physical viewpoint, the precise steps of hydrocarbon condensation remain elusive too.

Hydrocarbons, which are formed in Titan's cold atmosphere, starting with ethane C_2H_6 , ethylene C_2H_4 , acetylene C_2H_2 , propane C_3H_8 ... up to benzene C_6H_6 , play not only some role in aerosol production, but also in cloud processes, rain generation and Titan's lakes formation.

Our goal is to study in the laboratory the kinetics of the first steps of condensation of these hydrocarbons. Several studies have investigated the phase of e.g. ethane and propane and their spectral signatures. At

the exception of our studies on the dimerization of benzene (C_6H_6) [3], pyrene ($C_{16}H_{10}$) [4] and anthracene ($C_{14}H_{10}$) [5] performed over the 15-300 K temperature range, there is no other work on the first elementary steps of the kinetics of nucleation of hydrocarbons. Rate coefficients however, are very sensitive to the description of the potential interaction surfaces of the molecules involved. Combined theoretical and experimental studies at the molecular level of the homogenous nucleation of various small molecules should improve greatly our fundamental understanding. This knowledge will serve as a model for studying more complex nucleation processes actually taking places in planetary atmospheres.

Here we present the first experimental kinetic study of the dimerization of two small hydrocarbons: ethane (C_2H_6) and propane (C_3H_8). We have performed experiments to identify the temperature and partial densities ranges over which small hydrocarbon clusters form in saturated uniform supersonic flows. Using our unique reactor based on Laval nozzle expansions [6], the kinetics of their formation has also been investigated down to 23 K. The chemical species present in the reactor are probed by a time of flight mass spectrometer equipped with an electron gun for soft ionization of the neutral reagents and products.

This work aims at putting some constraints on the role of small hydrocarbon condensation in the formation of haze particles in the dense atmosphere of Titan.

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