High pressure reactivity of astrochemical model ices

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Ice mixtures are extremely common in our universe: mixtures of simple gases can be found in the cometary comae [1] besides that in the atmospheres and within the first crust layers of several planets and satellites in our Solar system [2].

Among these mixtures, the ternary system constituted by H_2O, NH_3 and CH_4 is of particular interest. First of all, these three gases are among the major constituents of Titan atmosphere and surface, being deeply involved in the chemical equilibria that characterize the evolution of the largest satellite of Saturn. Moreover, the mixture of H_2O, NH_3 and CH_4 can be considered a prototypical system in the framework of prebiotic chemistry. Since the Urey-Miller experiment [3,4], demonstrating that the biochemical constituents of living organisms could be synthesized by inorganic molecules under certain conditions, that replicate those of the early Earth, it is well known that the combination of reducing atmospheres and primary energy sources like electrical discharges, UV irradiation or shock waves may result in the synthesis of more complex molecules or precursor of biological interest. This research has produced during the years a lot of intriguing experimental and theorethical results [5,6].

Unexpectedly, the high pressure behaviour of the ternary mixture of H_2O, NH_3 and CH_4 has been poorly studied, both from structural and reactivity point of view: only one study is reported about the influence of free NH_3 in the dissociation of methane clathrate hydrates [7]. Here we report the results about the high pressure (below 1 GPa), photoinduced reactivity of $H_2O/NH_3/CH_4$ and $NH_3/H_2O/CH_3OH$ mixtures in diamond anvil cell (DAC). The reactivity was induced by means of two-photon (TP) absorption of near-UV radiation (355 nm) from a pulsed Nd:YAG laser, exploiting respectively the dissociative and pre-dissociative characters of the low-energy lying excited states of H_2O and NH_3. The reaction led to the synthesis of intriguing mixtures of products, involving the photodissociation of water as the initial, fundamental step of the process: besides the formation of H_2 and N_2, that demonstrate the photodissociation of both NH_3 and H_2O, we observed the formation of new solid products featuring amidic bonds like ammonium carbamate, thus suggesting the possibility to obtain complex organic precursor also starting from a very simple reducing mixture with respect to those reported in literature.

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