PROTON IRRADIATION OF SIMPLE GAS MIXTURES:
INFLUENCE OF IRRADIATION PARAMETERS

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ABSTRACT

In order to get information about the influence of irradiation parameters on radiolysis processes of astrophysical interest, methane gas targets were irradiated with 6.5 MeV protons at a pressure of 1 bar and room temperature. Yields of higher hydrocarbons like ethane or propane were found by analysis of irradiated gas samples using gas chromatography. The handling of the proton beam was of great experimental importance for determining the irradiation parameters. In a series of experiments current density of the proton beam and total absorbed energy were shown to have a large influence on the yields of produced hydrocarbons. Mechanistic interpretations of the results are given and conclusions are drawn with regard to the chemistry and the simulation of various astrophysical systems.

To improve the understanding of the chemical evolution of planetary atmospheres, many laboratory simulation experiments have been performed which investigate the influence of UV and ionizing radiation on primitive gas mixtures (for references see1). However, the physical conditions in the laboratory simulations differ dramatically from those in the simulated systems: e.g., the total irradiation time is many orders of magnitude lower and the flux of ionizing radiation higher. Hence the aim of our investigations was to evaluate the relevance of this latter irradiation parameter and to explain how it influences the chemical reaction network.

We irradiated stationary methane targets at room temperature and atmospheric pressure with 6.5 MeV protons provided by a Tandem - Van de Graaff accelerator. After irradiation the gas targets were analyzed through gas chromatography for higher hydrocarbons like ethane or propane. In order to define exactly the current density of the proton beam (which is proportional to the particle flux), emphasis was given to the handling of the beam: many steering and focusing elements as well as apertures were applied. Measurements
of the current density distribution over the proton beam have shown that the beam current was constant over the defined cross section. Experimental details are given in ref. (2).

Current density (power density) was varied between 0.026 μA/cm^2 (1.5 mW/cm^3) and 28.7 μA/cm^2 (1640 mW/cm^3); to create the same total absorbed doses the irradiated time was varied between 1.1 sec and 121 min. Strong dependencies of the yield of produced hydrocarbons on the current density have been found and these dependencies differ for different products^2.

To explain these results in terms of a chemical mechanism, one has to consider the principal reaction pathway that leads to the formation of the detected substances. The primary processes are ionizations and excitations of the methane molecules by the high energy protons; this leads to the production of various ions, radicals, and electrons, which itself may cause secondary ionizations. These ions will undergo either ion-molecule reactions or neutralizations. The final products are most likely produced in neutral reactions.

The ion-molecule and neutral reactions can also be classified in another way: some are reactions of reactive species (ions, radicals) with target molecules (methane) like

\[ \text{H} + \text{CH}_4 \rightarrow \text{H}_2 + \text{CH}_3 \]  

some are reactions between two different reactive species, e.g.,

\[ \text{H} + \text{C}_2\text{H}_5 + \text{M} \rightarrow \text{C}_2\text{H}_6 + \text{M} \]  

The latter reactions are certainly strongly dependent on the concentration of reactive species and in concurrence to reactions of type (1). Hence it can be stated that the concentration of reactive species (ions, radicals) has considerable influence on the reactions that occur. This density of reactive species is determined by the power density, which was a varied parameter in our experiments. Because the reaction network is complex it is not surprising that different substances show different dependencies of their yields on the varied parameter.

The fluxes of ionizing radiation in the astrophysical systems to which our results shall be applied, are generally very low: e.g., the flux of MeV protons in the atmosphere of Titan is of the order of \(10^6\) cm\(^{-2}\)sec\(^{-1}\); the fluxes in our experiments are \((1.6 \cdot 10^{11})\) cm\(^{-2}\)sec\(^{-1}\) to \((1.8 \cdot 10^{14})\) cm\(^{-2}\)sec\(^{-1}\). Hence our experimental conditions could perhaps better be applied to auroras or lightening, where very high particle fluxes can occur.
One consequence can be drawn from our experiments which is relevant to the simulation of planetary atmospheres: In most laboratory experiments the particle fluxes of ionizing radiation are by orders of magnitude higher than in the planetary atmosphere. Thereby it seems possible to simulate, for example, 1000 years of the evolution of the planetary atmosphere within some hours in the laboratory, as a similar total energy is deposited. But as our investigations have shown, there are enormous differences in the yields of products already in the small interval of particle flux examined in our experiments. Hence the application of such simulations in a quantitative way to the understanding of the planetary atmospheres is very difficult.

References

3 T. Scattergood, P. Lesser, and T. Owen, Nature 274, 100 (1975)

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