HYDROGEN ISOTOPE FRACTIONATION IN CH_4 PLASMA: COSMOCHEMICAL IMPLICATIONS.

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The deuterium-hydrogen isotope ratio (D/H) is commonly used to reconstruct the chemical processes at the origin of water and organic compounds in the early solar system [1]. On the one hand, the large enrichments in deuterium of the insoluble organic matter (IOM) isolated from the carbonaceous meteorites are interpreted as a heritage of the interstellar medium [2,3] or resulting from ion-molecule reactions taking place in the diffuse part of the solar disk [4]. On the other hand, the molecular structure of this IOM, suggests that organic radicals have played a central role in a gas phase organo-synthesis [5].

During the course of experiments aiming at reproducing this type of chemistry between organic radicals, we observed large hydrogen isotopic variations at a sub-micrometric spatial resolution in the IOM isolated from the black organic residues deposited from a microwave plasma of CH_4.

The plasma is generated by a microwave discharge in CH_4 at 1-3 Torr and the black residue is deposited on the glass walls of the reactor. The soluble organic molecules were removed from this residue by organic solvent extraction and the H isotopic composition of the remaining IOM was investigated with a NanoSIMS ion microprobe. The molecular structure of this IOM was also investigated by GC Mass-spectrometry and through various spectroscopic and degradative techniques [5]. The aromatic fraction of this structure is similar to the *chondritic* IOM isolated from the Murchison meteorite and also similar when octane is used in place of CH_4. This indicates that the chemical reactions with the "small" radicals $CH_3^\bullet$, $CH_2^\bullet$ or $CH^\bullet$ govern the chemical structure of the IOM.

In the plasma, the dissociation of CH_4 is achieved by electron impact and a high density of neutral molecular radicals $CH_x^\bullet$ (with the subscript x from 0 to 3) is maintained in steady state at 1600 K. These radicals are produced by two types of reactions:

$$CH_4 + e \rightarrow CH_3^\bullet + H + e \qquad \text{and} \qquad CH_x + H \rightarrow CH_{x-1}^\bullet + H_2$$

with x between 3 and 1. We will show that the distribution in the D/H ratio observed in 100 μm^2 IOM regions, likely reflects the differences in the isotopic fractionations associated with these types of reactions. Interestingly, these isotopic heterogeneities are commensurable with those observed in the meteorite IOM hot spots. Their δD range between -700 to +1200‰; at a sub-micron spatial resolution, δD can reach values up to +4000‰.

A possible interpretation of this isotope effect may hinge on the differences between the probabilities accounting for the interaction between distiguishable and indistinguishable isotopes (i.e. H-D compared to H-H). As a consequence, the appearance of organic radicals in the ionized regions of the T-Tauri solar disk may have triggered the formation of organic compounds.

References

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