NITROGEN ISOTOPE FRACTIONATION IN N_2-CH_x PLASMA: COSMOCHEMICAL IMPLICATIONS.

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The $^{15}N/^{14}N$ isotope ratio shows large and unexplained isotope variations at the scale of the whole solar system. On the one hand, all solid materials are enriched in ^{15}N compared with the protosolar value ($\delta^{15}N$ = -400‰) [1] and, on the other hand, the Insoluble Organic Matter (IOM) isolated from the carbonaceous chondrites is highly heterogeneous in ^{15}N exhibiting the so-called cold and hot spots ($\delta^{15}N$ = >-100‰ and <+400‰, respectively) [2]. Although the systematic solar system ^{15}N enrichement has been ascribed to a massive UV photodissociation of N_2 followed by a chemical reaction between the atomic N radicals and the protosolar H_2 to form NH_3 [3], the origin of the organic spots are not understood.

The molecular structure of this IOM, suggests that organic radicals have played a central role in a gas phase organo-synthesis [4]. Recently [5] we have shown that, in a plasma generated by a microwave discharge in CH_4, the black residues deposited on the glass walls of the reactor exhibit hydrogen isotope variations commensurable with those observed in the deuterium hot and cold spots present in the IOM. Therefore, following the previous study by Kuga et al [6] we have attempted a series of experiments with aliphatic and aromatic molecules, containing, or not, a N atom in their structure and in the presence, or not, of N_2.

The isotopic analyses were performed with the NanoSims at the Museum in Paris. The $\delta^{15}N$ was determined at a ROI scale of 3x3 µm with a typical ±20‰ reproducibility (2 sigma, ^{15}N cps ≥ 5000). Without N_2, the organic residues produced from N-bearing molecules exhibit homogeneous isotopic compositions within a ≈ 40‰ range (i.e. ± 2 sigma). In the presence of N_2, large areas (i.e. ≥ 25x25 µm) are either enriched or depleted in ^{15}N (variations lying between -180 and +120‰). In one case, in a mixture of Octane / N_2, small (1x1 µm) hot spots exhibit $\delta^{15}N$ up to +170‰. This last observation will be examined and discussed at the meeting. Correlation between $\delta^{15}N$ and the C/N ionic ratio allows to derive the isotopic composition of one of the end members in the simple situation of a two end member mixing model.

The relation at the origin of the isotopic fractionation has not been formally identified theoretically or experimentally. However, following the theoretical interpretation of ozone or that of the hydrogen isotope fractionation in a similar situation [5-7], the formation of a $[CN_2]^*$ complex having two decomposition channels ($[CN_2]^* \rightarrow C + N_2$ or $[CN_2]^* \rightarrow CN + N$) seems a possible interpretation for these Nitrogen isotope effects.

References:

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