

HYDROTHERMAL ORIGIN OF ORGANIC MATTER DIVERSITY IN CARBONACEOUS CHONDRITES: AN EXPERIMENTAL APPROACH

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Introduction: The organic molecules detected in primitive carbonaceous chondrites (e.g. amino acids, nucleobases and the insoluble macromolecule) are more complex than the molecules observed in the interstellar medium (ISM) [1]. Although large isotopic enrichments in deuterium may sign a connection between ISM chemistry and organic matter in carbonaceous chondrites, the hypothesis that organics in meteorites could derive from organics in the ISM is still debated. We have experimentally investigated the chemical evolution of interstellar organic matter submitted to hydrothermal conditions mimicking asteroidal alteration. We have also tested the potential catalytic role of clays minerals in the polymerization or degradation of organic matter on chondrite parent bodies.

Methods: Hexamethylenetetramine (HMT, an organic compound rich in C-N bonds) was chosen as an interstellar organic precursor. HMT is a simple molecule (22 atoms- C₆H₁₂N₄) that represents about 50 wt% of the typical organic residues recovered in interstellar ice analog experiments [2]. This molecule is indeed easily formed in ice (containing water, formaldehyde, ammonia and formic acid) submitted to thermal processing between 20 K and 300 K [3]. Although it remains stable in the solid phase until 400K, it has never been detected in the ISM so far, neither in carbonaceous chondrites.

Hydrothermal experiments were conducted at 150°C, under alkaline pH and for various durations up to 31 days [4]. Experiments were performed with and without clay minerals (laboratory synthesized organic-free montmorillonite and Fe³⁺-rich nontronite) in order to assess the influence of minerals on the chemical evolution of HMT. Organic products were characterized by GC-MS, FTIR, and synchrotron-based X-ray absorption near edge structure (XANES) spectroscopy. Inorganic material was studied by X-ray diffraction, FTIR and thermogravimetry (TGA).

Results and discussion: Under our hydrothermal conditions, HMT totally decomposes in less than 20 days, which may explain why HMT has never been detected in meteorites. Organic products consist in (1) a very diverse suite of soluble compounds dominated by N-bearing aromatic compounds (> 150 species after 31 days with m/z 73 to 250), including for instance formamide, pyridine, pyrrole and their polymers (2) an aromatic and N-rich insoluble material that forms after only 7 days of experiment and then remains stable through time [4]. This insoluble organic material represents less than 1wt% of the organic products and its elemental composition is H/C=1.2, N/C=0.26 and O/C=0.31. The reaction pathways leading to the soluble compounds likely include HMT dissociation, formose and Maillard-type reactions, e.g. reactions of sugar derivatives with amines [4].

When minerals are present, the organic diversity is reduced: 36 and 66 soluble organic molecules are identified by GC-MS in the 31 day experiment, in the case of nontronite and montmorillonite, respectively (to compare to 150 in the experiment with no mineral). XRD reveals that organic matter gets trapped within the phyllosilicate interlayers. At the end of the experiments, nontronite contains up to 2wt% of organic matter, montmorillonite 7wt%.

Our study reveals that simple organic compounds undergo complex chemical transformations during hydrothermal alteration on chondrite parent bodies, possibly forming complex insoluble macromolecule. Some of the diversity of organic compounds found in carbonaceous chondrites may thus result from asteroidal hydrothermal alteration. Phyllosilicates may actively influence these reactions, notably by concentrating a significant proportion of the newly formed organics. In addition, phyllosilicates may trap the newly formed organic compounds and protect them from subsequent alteration. The present study constitutes step forward to decipher the potential genetic relationship between the simple organic molecules detected in ISM and the complex organic molecules observed in carbonaceous chondrites.

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References: [1] Remusat L. (2015), EMU notes in Mineralogy, Vol 15, chapter 8. [2] Munoz-Caro G.M., Schutte W.A. (2003) *Astronomy & Astrophysics*, 412:121-132. [3] Vinogradoff V. et al. (2012), *Physical Chemistry Chemical Physics*, 14:12309-12320. [4] Vinogradoff V. et al. (2018) *Icarus* 305:358-370.