

N-heterocyclic compound synthesis from aldehydes and ketone with ammonia: A simulation of organic reactions on the meteorite parent bodies

*Junya Miyazaki¹, Hiroshi Naraoka¹, Akira Tsuchiyama²

1. Department of Earth and Planetary Sciences Kyushu University, 2. Division of Earth and Planetary Sciences Kyoto University

[Introduction]

Carbonaceous chondrites have the most primitive chemical compositions in the solar system and contain volatile components such as water and carbon. In spite of their primitive character, most carbonaceous chondrites have had experienced aqueous alteration on parent bodies, which was revealed by the occurrence of hydrous minerals. The meteoritic carbon are present mainly as organic matter, in which soluble organic matter (SOM, ~1 to ~30 wt%) consists of relatively low-molecular-weight compounds such as carboxylic acids and amino acids. The rest major carbon exists as insoluble organic matter (IOM) having high-molecular complicated structures. Therefore, the aqueous alteration must have influenced the organic-mineral interactions. In order to study the chemical evolution of organic matter in the solar system, the role(s) of minerals should be clarified. In this study, we performed simulation experiments of organic compound synthesis using ammonia (NH₃), formaldehyde (HCHO), acetaldehyde (CH₃CHO), propionaldehyde (C₂H₅CHO) and acetone (CH₃COCH₃), which are simple molecules found in molecular clouds, under the aqueous environment in the presence or absence of minerals.

[Materials and Methods]

The several aqueous mixtures with various ratios (by mol) of NH₃ (1 to 10) /HCHO (0.1 to 1) /CH₃CHO (0.01 to 0.1) /C₂H₅CHO (0.01 to 0.1)/CH₃COCH₃(0.01 to 0.1) were heated in the presence of powdered mineral including forsterite (San Carlos), magnetite (Utah), synthetic forsterite or amorphous silicate (Mg₂SiO₄ in composition) at 60 to 80°C for 144 to 192 hours in N₂-purged glass ampoules. A total of 20 runs of simulation experiments were performed. The reaction product was extracted with methylene chloride/methanol (2/1, by volume) and analyzed by high performance liquid chromatography/mass spectrometry.

[Results and Discussion]

Many compounds were observed at the range of m/z 100-400 in all reaction products. Of the identified compounds, alkylpyridines (C_nH_{2n-5}N), alkylimidazoles (C_nH_{2n-3}N₂) and hexamethylenetetramine (C₆H₁₂N₄, HMT) were the main products for each run. The compounds that have not been identified but represented by compositional having C_nH_{2n-1}N₃O in composition were also identified as a main product. Although alkylpyridines and alkylimidazoles have been detected from carbonaceous chondrites (e.g. Yamashita and Naraoka, 2014). Although HMT has been reported as a main product during interstellar analog experiment by UV irradiation at low temperature (Vinogradoff et al., 2011), HMT has not been found in meteorites. HMT is relatively abundant in the products with the absence of minerals. However, the HMT amount decreased in the presence of the amorphous silicate and forsterite (San Carlos), instead of increasing production of alkylpyridines and alkylimidazoles. The minerals could control the reaction pathways as a catalyst. In particular, as amorphous silicate is the main solid phase in the interstellar environments (F. Kemper et al., 2004), it may have important roles for the evolution of extraterrestrial organic matter. Further experiments are needed to identify the intermediate compounds as well as to observe the change in mineral phases during the reactions.

[References]

Yamashita Y. and Naraoka H. (2014) Two homologous series of alkylpyridines in Murchison meteorite. *Geochemical Journal* 48: 519-525.

F. Kemper, W. J. Vriend, and A. G. G. M. Tielens(2004) THE ABSENCE OF CRYSTALLINE SILICATES IN THE DIFFUSE INTERSTELLAR MEDIUM. *The Astrophysical Journal*, 609, 826–837

V. Vinogradoff, F. Duvernay, G. Danger, P. Theulé, and T. Chiavassa New insight into the formation of hexamethylenetetramine (HMT) in interstellar and cometary ice analogs. *Astronomy & Astrophysics* 530, A128

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