July 16-21, 2017 at UC San Diego, CA, USA

Formose reactions with ammonia prevailing for the synthesis of meteoritic soluble organic matter

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Introduction: Chemical evolution of extraterrestrial organic matter has been investigated mainly using carbonaceous meteorites, which are the primitive materials of the solar system. The carbonaceous chondrites contain a few weight % of carbon mainly as organic matter. A wide range of organic compounds including amino acids, carboxylic acids, sugars and nucleobases has been found in the meteorites [1]. The origins and comprehensive formation mechanism(s) of these compounds, however, have not been clarified due to the incomplete understanding of molecular occurrence [2]. The carbonaceous chondrite also contains water mainly as hydrous minerals resulting from aqueous alteration on the meteorite parent bodies. The aqueous activity and minerals could have important roles in chemical evolution in the early solar system. Therefore, further meteorite analyses and simulation experiments are needed to pursue the formation pathways of extraterrestrial organic matter.

Materials and Methods: The Murchison meteorite was extracted with methanol and hot water. The methanol extract was analyzed by high performance liquid chromatography/high resolution mass spectrometry (HPLC/HRMS) using an Orbitrap MS ($m/\Delta m > \sim 100{,}000$). The hot water extract and its extract residue were subjected to acid hydrolysis followed by derivatization to analyze by gas chromatograpy/MS. The simulation experiments were performed using formaldehyde and acetaldehyde with ammonia in aqueous solutions [3, 4]

Results and Discussion: Extensively alkylated N-containing cyclic compounds were revealed in the methanol extract of the Murchison meteorite. More than 600 positive ions were assigned to $C_nH_mN^+$ and $C_nH_mN_2^+$ in elemental compositions, in which saturate- and unsaturate-alkylated pyridines ($C_nH_{2n-5}N$ and $C_nH_{2n-7}N$, respectively) and alkylimidazoles ($C_nH_{2n-2}N_2$) were predominant [3]. In the water extract, totally 30 amino acids between C_2 and C_6 were identified including a new family of nine C_3 and C_4 hydroxy amino acids in addition to the most abundant glycine [4]. The simulation experiments gave various alkylpyridines and alkylimidazoles as well as amino acids including the hydroxy amino acids. Both the N-heterocyclic compounds and the amino acids could be produced from aldehydes and ammonia through aldol condensation and imine formation under an alkaline environment. The presence or absence of minerals affected the compound occurrence in the simulation experiments. Therefore, these results indicate that formose reactions with ammonia in the presence of minerals are an important process to produce meteoritic soluble organic matters including sugars [5] through aqueous alteration on the meteorite parent body. The soluble N-heterocyclic compounds may have a genetic relationship with the meteoritic insoluble organic matter, which also could be produced from aldehydes and ammonia through the similar mechanism proposed by Cody et al. [6]

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