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Formation and solubility of organic aerosols in the atmospheres of Titan and early Earth

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Titan, the largest moon of Saturn, might provide clues for the chemical evolution toward life in early Earth's atmosphere, as Titan has a thick atmosphere mainly composed of N_2 and CH_4 probably resembling that of early Earth. Titan is covered with thick haze layers of organic aerosols produced from chemical reactions initiated by the dissociation of N_2 and CH_4 caused by solar UV irradiations and Saturn's magnetospheric electrons bombardments. Previous studies performed laboratory experiments simulating Titan's organic chemistry by irradiating cold plasma to gas mixtures of N_2 and CH_4 in order to produce solid organic materials called tholin. Recent laboratory experiments also irradiated cold plasma or UV light onto gas mixtures of N_2-CH_4-CO_2-CO simulating early Earth's atmosphere. However, there are only few studies focusing on the mechanisms and formation processes of Titan and early Earth tholin. Moreover, organic aerosols formed in the atmospheres of Titan and early Earth would have undergone further reactions with liquid on the surface. Nevertheless, solubility of early Earth tholin to the organic solvent has not been studied.

We have conducted laboratory experiments simulating the formation of organic aerosols in the atmospheres of Titan and early Earth by irradiations of cold plasma onto gas mixtures of N_2-CH_4, N_2-CH_4-CO, or N_2-CH_4-CO_2. With the objective of revealing the processes of the tholin formation, mass spectrometry and emission spectroscopy were performed to identify the gas-phase reaction products. Chemical structures of the tholins were analyzed by infrared and UV-VIS spectroscopy. Production rates of tholin were examined by measuring the thickness of tholin with ellipsometry. We also performed solvent extraction of the tholin and analyzed the chemical structure by UV-VIS spectroscopy for the solvents.

Our experimental results show that the formation of Titan tholin from gas mixture of N_2-CH_4 occurs efficiently, compared with early Earth tholin formed from N_2-CH_4-CO or N_2-CH_4-CO_2. The formation rate of early Earth tholin decreases with CO_2 introduced in the initial gas mixture, whereas tholin forms efficiently from gas mixtures of N_2-CH_4-CO. Results of emission spectroscopy revealed that CN radicals are produced by plasma irradiations. Mass spectrometry of gas species demonstrated that HCN and cyanides, such as CH_3CN, are produced in the experiments in addition to hydrocarbons. These results suggest that the formation of Titan tholin is initiated by the production of CN radicals and subsequent cyanide formations. Regarding early Earth tholin, our results suggest the production of CN radicals as an important process for the tholin formation as well as Titan tholin. Though, the results from infrared spectroscopy suggests existence of C=O bonds in early Earth tholin. Thus, the production of CO and incorporation of oxygen into the tholin are also a significant process for the formation of early Earth tholin.

Our results of solvent extraction show that early Earth tholin dissolve effectively in polar solvents, such as methanol. We also found that early Earth tholin displays a dissolution to low-polar solvents, such as CH_2Cl_2, suggesting the existence of both non-polar and polar structures contained in early Earth tholin. Chemical structures that contain both hydrophilic and hydrophobic parts may have played an important role in the origination of bio-membrane structure (lipid bilayer). Also, our results from UV-VIS spectroscopy of CH_2Cl_2 solution of early Earth tholin display an absorption band typical to fused ring aromatic compounds or heterocyclic compounds including porphyrin. These results suggest that organic aerosols produced in the atmosphere of early Earth might have played a key role in the chemical evolution possibly by providing nitrogen-containing bio-related heterocyclic compounds as well as bipolar complex organics to pre-biotic oceans.

Keywords: Titan, early Earth, reducing atmosphere, organic aerosol, chemical evolution