

Scanning electron microscopic observation of organic microspherules formed by Maillard-type reaction

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It has been suggested that organic microspherules played a role as a physical container to maintain catalytic molecules and their reaction intermediates at concentrations high enough to sustain catalysis in prebiotic chemistry on the early Earth (Weber, 2005). Experimental studies on the formation of organic microspherules from a variety of organic compounds, such as amino acids (Fox and Harada, 1958), gelatin and gum arabic (Oparin, 1976), organic extracts from meteorite (Deamer, 1985; Deamer and Pashley, 1989), interstellar organic analogue (Dworkin et al. 2001), fatty acids and polycyclic aromatic hydrocarbons (Groen et al. 2012), formaldehyde and ammonia (Cody et al. 2009; Kebukawa et al. 2013) have been reported. However, the formation process and stability of these organic microspherules have been unexplored. In this study, sizes, shapes, and distributions of organic microspherules formed during the progress of Maillard-type reaction of formaldehyde and ammonia were investigated.

Experimental:

Paraformaldehyde (120mg), glycolaldehyde (120mg), ammonium hydroxide (54ul), calcium hydroxide (30mg) in 2ml of water in a glass tube was heated at 50-90 degrees C for 71-720 hours. For comparison, the samples without ammonium hydroxide were heated under the same conditions. After heating, the sample solutions were centrifuged. The precipitated material were rinsed with 2N HCl to dissolve calcium, and dried at 50 degrees C to obtain organic solids. The organic solid samples were pressed on a indium plate, gold-coated, and observed by a scanning electron microscopy (SEM).

Results and discussion:

After several minutes in heating, all the sample solutions turned yellow and eventually turned brown to black. Organic solids were produced at 90 degrees C but 50 degrees C. The yields of organic solids from sample solutions with ammonia were 10 times higher than those without ammonia. The yields gradually increased during heating. While distorted-shaped aggregates are produced from the samples heated for 71-120 hours, micron-sized organic microspherules (0.4-4.0 um) were observed from those heated for 240-720 hours. The samples with ammonia show perfectly round shapes of microspherules. Some microspherules are large and oval in the sample heated for 480 hours. The sizes of the microspherules increased with heating time. Organic solids produced by the same reaction as this study's at 90 degrees C for 72 hours consist of approximately equal abundances of aromatic and aliphatic carbons (Kebukawa et al. 2013). This molecular composition could result in amphiphilicity that is related to formation of the stable microspherules observed in this study. Formaldehyde and ammonia are thought to have been commonly present on the early Earth, and thus the organic microspherules formed by these molecules which proceed polymerization efficiently under mild conditions, could have played a role as a precursor of prebiotic cell membrane.

Keywords: organic microspherules, Maillard reaction, prebiotic cell membrane