J.Mol.Evol.7,245-251 (1976)

Journal of
Molecular Evolution

Melanoidin and Aldocyanoin Microspheres: Implications for Chemical Evolution and Early Precambrian Micropaleontology

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Received November 18, 1975; January 1, 1976

Summary. Two new classes of organic microspheres are described. One of them (melanoidin) is synthesized from amino acids and sugars in heated aqueous solutions. The other (aldocyanoin) is formed in aqueous solutions of ammonium cyanide and formaldehyde at room temperature.

The general properties of these microspheres, including conditions of synthesis, size and shape, mechanical and pH stability, and solubility, are compared with corresponding properties of other "protocell" model systems. It is concluded that melanoidin and aldocyanoin microspheres are plausible candidates for precellular units in the primitive hydrosphere.

Since the bulk of the organic carbon in early Precambrian sediments is insoluble kerogen-melanoidin, it is suggested that some Precambrian "microfossils" may be abiotic melanoidin microspheres of the type described herein.

Key words: Abiotic Synthesis/Aqueous Systems/Organic Microspheres/Aldocyanoin/Melanoidin/Precambrian Sediments/Microfossils

INTRODUCTION

The presumed phase of prebiotic chemical evolution which bridges the gap between the synthesis of biopolymers and the appearance of the first living cells is the least understood aspect of the origin of life problem. Several model systems have been proposed for the prebiotic origin of microscopic units ("protocells") which might have evolved into the first

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cells. Among these are proteinoid microspheres (Fox & Dose, 1972), coacervate droplets (Oparin, 1965), "Jeewanu" (Bahadur, 1966), NH_4CN microspherules (Labadie et al., 1969), "sulphobes" (Herrera, 1940), and NH_4SCN-HCHO microstructures (Smith et al., 1968). The list was recently extended by Folsome et al. (1975) who described organic microstructures formed in a Miller-Urey type of electric discharge experiment.

Although each of the proposed model systems exhibits some rudimentary properties of chemical evolutionary interest, it must be emphasized that a very large gap separates the most complex model systems from the simplest contemporary living cells (i.e., the Mycoplasmatales). Moreover, the geochemical plausibility of many of these "protocell" models is open to serious question (Nissenbaum et al., 1976). One of the chief criticisms of the major current "protocell" model is that it employs a limited range of initial reactants, such as pure L-amino acids in the dry state (Fox & Dose, 1972). In spite of these limitations, comparisons between organic microstructures formed in the laboratory and putative Precambrian microfossils as well as "organized elements" in meteorites suggest that many of these structures may have been formed abiotically (Folsome et al., 1975; Smith et al., 1969; Fox, 1972). In this paper we describe two new classes of organic microspheres synthesized from compounds likely to have been abundant in the primitive hydrosphere. The ease of formation and the morphology of these structures must be taken into account in attempts to determine the biogenicity of some Precambrian organic microstructures.

We have argued that insoluble polymers similar to the melanoidin-type material which makes up the bulk of the organic carbon of sediments may have been synthesized on a large scale in the prebiotic hydrosphere (Nissenbaum et al., 1976). Melanoidins are readily synthesized in the laboratory in dilute aqueous solutions of an amino acid and a sugar (Nissenbaum et al., 1976; Hoering, 1973). They form over a wide range of temperatures, reactant concentrations, and solution *p*H's (Ellis, 1959).

Aldehydes and cyanides are known to have played a variety of roles in prebiotic chemical evolution (Miller & Orgel, 1974) Substances of one or both classes have been detected in comets (Ulich & Conklin, 1974; Marsden, 1974), meteorites (Breger et al., 1972), and deep space (Snyder & Buhl, 1973). In aqueous solutions these compounds can generate a rich variety of biomolecules (Oró, 1963; Oró & Kimball, 1961; Ferris et al., 1974; Ferris et al., 1973; Sanchez et al., 1967; Loew et al., 1963) including agents which promote dehydration-condensation of biomonomers (Steinman et al., 1965, 1966). Microscopic particles (Labadie et al., 1967) form in solutions of NH_4CN, and