

Chapter 7

THEORETICAL AND EXPERIMENTAL PREREQUISITES
OF EXOBIOLOGY¹

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**EXOBIOLOGY AND
THE ORIGIN OF LIFE:
THE INSEPARABLE CONNECTION**

The world we live in is constantly changing through a process of evolutionary development that is gradual, and which leads to emergence of ever newer, more complex, more diverse, and richly differentiated forms of reality. This process may not occur simultaneously and identically everywhere, but in different ways at different rates in various places in the universe and on various objects in the celestial world. Thus, evolution must not be diagramed as a single direct line, but rather as a cluster of various paths, whose separate branches may lead to very complex and complete forms of organization and movement of matter. Nothing is known about many of these forms, and in numerous instances, their existence is not even suspected. Any one form should not be considered prematurely as a special category of life, only because of its complexity or perfection. Life is far from a result

of all the numerous branches in the development of matter; its specific qualities and ways of emerging and improving are inherent. Only a single example of life, terrestrial life, is known at present, from which judgments must proceed of other possible forms of biological organization. The Earth and the events which originated on it must, to a great extent, serve as a model for broader judgments of life in the universe.

As long as the origin of life on Earth was considered a kind of rare "happy accident," there was little stimulus to discuss life beyond our planet, apart from calculations of the degree of probability of such an "improbable" event, which was scarcely intellectually satisfying. However, there is every reason now to see in the origin of life not a "happy accident" but a completely regular phenomenon, an inherent component of the total evolutionary development of our planet. The search for life beyond Earth is thus only a part of the more general question, which confronts science, of the origin of life in the universe. Study of the conception of life on Earth amounts to an investigation of only one example of an event which must have occurred countless times in the world. Therefore, an explanation of how life appeared on Earth should strongly support the theory of existence of life on other bodies in the universe.

The problem of the origin of life, almost completely ignored by the scientific world at the

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beginning of this century, now attracts many investigators with different specialities and from various countries. The related scientific literature has thus increased greatly. It is expedient to cite from the literature those monographs [40, 41, 75, 76, 261, 305, 368, 369, 371, 374, 375, 376, 440, 486, 487] and thematic collections [71, 120, 148, 246, 263, 320, 373, 381, 438, 502, 511] published in recent years which contain generalized data and broad summaries.

Terrestrial life originated specifically through the evolutionary development of hydrocarbons (organic compounds). The proposal is often made in scientific, and particularly in popular literature, that life could have emerged on other bodies of our celestial world based on compounds of other elements (silicon, germanium, and so forth). Since signs of the existence of such forms of life have in fact not been observed, this proposition is general and purely theoretical, but even in such form it meets very solid theoretical objections. Quantum chemistry, which provides an understanding of the specific electron structure of molecules [459, 463, 464, 625], nowhere favors the possibility of a noncarbon form of life. The most important vital functions are impossible without conjugate or resonant molecular systems, which are rich in electrons and highly delocalized [460, 461]. No other element may replace carbon in this respect. In particular, silicon—close to carbon in Mendeleev's table—could not serve as the basis for the structural components of life despite its abundance on the surface of the Earth [115, 232, 624]. This prediction, according to modern quantum chemistry, is based on electronic delocalization, which is inherent in carbon compounds but lacking in silicon compounds [282, 462]. Thus, in the summary works cited and in this review, attention is concentrated on the evolutionary development of carbon compounds, which led to the origin of life on Earth.

EMERGENCE AND EVOLUTION OF CARBON COMPOUNDS IN THE UNIVERSE

Modern astronomical data reveal surprising interconnections within the galactic system as

a whole. The evolution of the universe appears at present as a single development, where each subsequent stage is inseparably bound to the preceding one and understood only in that light. An arbitrarily selected starting point of evolution, which has lasted many billions of years, is the process of nucleosynthesis, i.e., formation of the atomic nuclei of all the known elements. The evolution of nucleosynthesis is normally set in parallel with the evolutionary development of the stars [64, 144, 145, 162, 208, 555], since it has become clear that the process of star formation has occurred and continues without interruption in the universe [17, 131, 184, 514].

That our galaxy consists of five different types of star population [329] is conditionally accepted. Stars of an age close to that of the galaxy [146, 236, 240] belong to the first and most ancient type—from 12 to 20 billion years old. The Sun, a younger star of the third generation, is calculated to be about 5 billion years old, i.e., it originated after our galaxy had existed for a long time [58]. Even younger star populations are in our galaxy, among which, in particular, are stars located directly in the vicinity of the spiral branches of the galaxy [11, 65]. The elemental composition of stars of the first generation differs from that of younger stars in having a much higher ratio of hydrogen over metals. This graphically indicates that the initial medium from which stars of the first generation were formed consisted almost exclusively of hydrogen, which even now is the most prevalent element in space (98% of all atoms in the universe are hydrogen atoms) [144, 566]. The stars of later generations (in particular the Sun), however, were formed from a mixture of light and heavy elements, which must have originated long after the galaxy began to form [239, 248].

Nucleosynthesis

Modern data show that two paths of nucleosynthesis are possible: one, stable, is related to the mechanism of stellar radiation in stable stars; and the other, unstable, is related to supernova bursts [77, 79, 558, 559]. The nucleosynthesis of light elements is exoergic; the energy liberated supports the stellar radiation. Be-

cause electrostatic repulsion during the joining of hydrogen nuclei is relatively small, a conversion of hydrogen into helium may occur at temperatures of a million degrees. Nucleosynthesis of light elements is even possible in a contracting gas sphere which consists of only hydrogen, and it may begin immediately after this sphere reaches the indicated temperature due to its compression and the liberation of gravitational energy. For the origin of heavier elements, however, a temperature of several billion degrees is necessary, which is lacking in the interiors of stable stars and arises only during supernova bursts.

Carbon did not require supernova bursts for its nucleosynthesis, but originated long before the formation of the heavy elements in the stable process of stellar radiation. The carbon-nitrogen reaction, along with proton-proton transformation, is the basic thermonuclear source of the stars' energy. Hydrogen, helium, carbon, nitrogen, and oxygen comprise the group of nucleogenetically related elements. Hydrogen is the most primordial chemical element in the universe, and all other elements are formed from it. The nuclear fusion of hydrogen, as a result of the action of the proton-proton cycle, led to the formation of helium at the beginning of stellar evolution.

The carbon-nitrogen cycle, during the process of "combustion" of helium and the capture of α -particles, leads to the formation of carbon, nitrogen, and oxygen in correspondingly more developed celestial bodies, particularly in the so-called carbon stars [64, 65]. Therefore, carbon, like other light elements (H, He, N, O), is extensively distributed in the universe. It could have been detected comparatively long ago in studying the spectra of all classes of stars, including the most ancient generations [272]. In stellar atmospheres which have the highest surface temperatures, carbon is present in the ionized or neutral atomic state [210, 433], but even beginning with type-A stars, g-bands appear in their spectra, which indicate the origin of primary carbon compounds—hydrocarbons (methane, CH, [627]). In the spectra of subsequent types of stars, the hydrocarbon bands appear with increasingly greater clarity as the surface temperature

of the stars decreases, and reaches maximal clarity in the M, N, and R spectra. The spectra mentioned also show, in the atmosphere of the stars, the presence of compounds of carbon with nitrogen (CN₁) and the so-called Swan bands, which are determined by molecules consisting of two connected atoms of carbon (C₂) [497, 526], and the presence of (C₃) and (CH₂) [217].

The formation of water, molecular hydrogen, nitrogen, and oxygen, and of these six diatomic compounds—C₂, CN, CO, CH, NH, OH [11]—is an unavoidable consequence of the interaction of carbon with nitrogen and oxygen which are produced by it in stellar atmospheres which are exceptionally rich in hydrogen. Thus, in the atmospheres of relatively cold stars [11, 217], including the Sun [4], hydrogen, carbon, nitrogen, and oxygen exist mainly as atoms and as homo- and heteronuclear diatomic combinations. The triatomic molecules H₂O, C₃, and C₂H are found in relatively large quantities in the atmospheres of carbon stars, which, it has been proposed, were the sources of interstellar carbon kernels or graphite particles [241]. The presence of HCN, HCO, CH₄ and NH₃ in dwarf stars was proposed by Vardya [596] on the basis of his calculations and a number of theoretical concepts. However, not one of these compounds has actually been detected so far. The stars of various generations must be viewed, therefore, as the starting place for the synthesis of original carbon compounds which are predecessors of organic substances.

Star Formation

Stars are in constant interaction with their surrounding interstellar space. This interstellar medium is far from uniform. Turbulent processes are observed in it, including local warmings due to the effect of gas ionization. The bursts of supernova stars create shock waves, at which time the gas density in such a wave increases by several orders, and this may be highly significant for the formation of gravitationally related condensations and subsequent formation of stars [134]. If a high-density gas-dust cloud appears on the path of the shock wave which characterizes the propagation of an ionization front, it is com-

pressed due to the pressure of hot gas and converted into a dense dark formation—a globule. This is the embryo of a star, which then, in a quite short time, may be compressed even more as a result of gravitational collapse and be converted into a self-luminous body. A large number of such globules, normally connected to nebulae, are known at present [132]. A graphic example of such a process is represented by filaments in the constellation Cygnus. Studies of various photographs of this region, taken during half a century, show that these filaments, which include a certain amount of stellar chains, are fleeing at great speed from some general center, shown to be the source of radio emissions. A phenomenon of such a type is interpreted by Fesenkov as proof of a supernova star burst in the past at a given point of the galaxy [132]. Many similar filaments have been photographed by Shayn [527] in various regions of the sky.

Quickly developing condensations, in which noticeable changes have appeared for several years, are also encountered in the region of the Orion nebula, where even now an intensive star formation is occurring [32, 212]. In the spectrum of this nebula, narrow interstellar absorption bands of CH, CH⁺, CN, and a number of other molecular bands have been detected. Solid particles with dimensions in fractions of microns are also scattered in these condensations. The composition of these particles is unknown, but in their spectral characteristics they identify with graphite and, as recently suggested, even with diamonds [498, 605].

Star formation from interstellar matter occurs constantly in our galaxy [17, 131, 184, 514]; but in line with this, the stars are constantly giving off part of their substance into the surrounding medium, enriching it particularly with carbon compounds. They create favorable conditions in space for the synthesis of various organic compounds, radiating cosmic rays and plasma clouds which contain ionized atoms of different elements. For example, the Sun creates the so-called solar wind, which washes the upper layers of its atmosphere with intense radiation at great velocity. They comprise a gaseous component of the solar corona which, however, contains solid particles of varying origin [135].

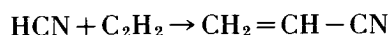
N-type red giants, on an enormous scale, generate a very fine carbon dust which is constantly expelled into space by radiation pressure and forms the chief component in space clouds [109]. The total mass of these particles does not exceed 1% of the gas composition of the interstellar environment. However, these particulates are responsible for the very high light absorption which determines the overall appearance of the Milky Way. These particulates may also give rise to molecular systems, because they are highly irradiated by cosmic rays and stellar emanations [238, 241, 247]. Therefore, it is not surprising that radiometric determinations give evidence of the presence, here, of different organic compounds in various regions of interstellar space [29, 49, 107, 114, 217, 465, 529].

Interstellar Space

Thermodynamic data show that in the presence of hydrogen, carbon is reduced to methane, nitrogen to ammonia, and oxygen to water [345]. Actually, recent radioastronomic observations have shown ammonia [82] and water vapors [83, 540] to be in interstellar space. Carbon monoxide (CO) has been detected in various regions of the Milky Way [249, 636]. Molecular hydrogen in interstellar space was predicted by Knaap et al before its actual detection [273]. HCN was also observed in Milky Way emissions [59, 541], and formaldehyde was the first organic molecule detected in space [411, 542, 654]. The region where it was found is considered dusty, and it has been suggested that this organic substance is a consequence of the particulate protection of molecules from ultraviolet light; but it is more probable that this is related to an increased rate of formation of formaldehyde.

Complex chemical processes must undoubtedly occur in interstellar dust clouds, which prompted the suggestion that large particles of carbon capture other atoms (H, N, and O) in interstellar dust and form more complex organic molecules [58]. One of such molecules detected in interstellar space is the molecule of cyanoacetylene [584, 585]. Its formation consists of 5 atoms of 3 different elements, which cannot be explained by simple gas-phase reactions capable of syn-

thesizing diatomic molecules; however, cyanoacetylene could be formed in the reaction between HCN and acetylene:



Hodgson [222] suggests a possible reaction for the formation of pyrrole from an unsaturated four carbon chain and ammonia. The interaction of pyrrole with formaldehyde could be the mechanism for formation of porphyrins in plasma reactions [226]. There are also indications here of the presence in interstellar space of other polymer organic substances, interpreted by some authors [251, 252, 253] as porphyrins, and by others as polyaromatic hydrocarbons [106, 107, 211, 626].

Comets

Studies of comets provide a great deal of information on carbon compounds in interstellar matter. There is not yet a completely clear concept of the mechanism of origin of comets; however, there is no doubt that such unstable small bodies could not be generated within the solar system. Condensations of similar composition, obviously, arise universally in space from typical interstellar material. Comets, in their chemical composition, reflect principally the composition of primordial solar nebulae [291], containing large amounts of carbon compounds, and thus may be considered space models for the abiotic synthesis of biochemically important compounds [393, 395]. The nucleus of comets is seen as a discrete mass of solid particles which could be formed from typical interstellar material at a temperature on the order of 10° – 100° K [631, 632]. When comets approach the Sun at a distance less than 3 astronomical units (AU), it is possible to observe their spectrum, which reveals fluorescent emission bands corresponding to molecules of CN, CH, CH_2 , C_2 , C_3 , NH, NH_2 and OH, and also to radicals CH^+ , OH^+ , CO^+ , N_2 and CO_2^+ [32, 313, 475, 573, 574, 641]. These compounds supposedly exist in the comet nuclei in one of these forms:

1. As frozen free radicals [108, 174].
2. As "ice"–water, ammonia, and methane [631, 632].
3. As clathrate-type hydrates [342].

A fourth probable form consists of the so-called parent compounds, which comprise the internal parts of the comets and must be complex organic compounds such as HCN, NH_2 , H_2O , CO, C_2N_2 , CH_4 , C_2H_2 and other hydrocarbons [200, 217, 234, 574].

The importance of these comet substances for formation of biochemical compounds at the preplanetary and planetary stages of the Earth has been particularly emphasized by Oró [393, 395, 397]. Comets can, even at present, "fatten" the Earth with organic substances to some degree. Of course, their role in this connection was more significant in the past when the population density of a near-solar comet cloud was much greater. Even with the present abundance of comets, it could be expected that the Earth, in the course of its history, collided with comets no less than a hundred times, thereby receiving various organic compounds from them [394].

Thus, the atmospheres of stars, the accumulations of gas-dust matter, the comet clusters and interstellar space contain an abundant amount of carbon and its compounds—organic substances which alone are completely capable of supporting all chemical evolution toward living substances similar to those on Earth [303]. These organic substances may originate, exist, and evolve in a wide range of temperatures from 3000° K and greater which are prevalent in the atmosphere of carbon stars [11, 434, 573] down to 1.8° K in dust clouds [58]. However, this evolution could scarcely lead to the emergence of life here, for which the necessary condition is the formation of planetary systems. Heavenly bodies with a sufficiently large mass, such as stars, may be formed almost entirely from pure hydrogen. However, the existence of bodies the size of Earth with only this element is inconceivable because the mass of these bodies would not be sufficient to prevent its dissemination in space. Accordingly, it is possible to conclude that during the first billions of years of the existence of our galaxy, when heavy elements had not yet been formed in a sufficient quantity, planetary systems similar to ours could not exist. Therefore, life could not exist during the evolutionary process of these planets, which makes it possible to conclude that propagation

of life in the universe is limited to stellar populations of sufficiently late types. However, it cannot be assumed that our solar system is extraordinary in any way. A great multitude of similar systems must exist in our galaxy. Of course, not all planets of this or any system may be abodes of life, for which they must meet a number of conditions in terms of dimensions, chemical composition, distance from a central light source, temperature, illumination, and so forth. These conditions can evidently be fulfilled by only one or two planets in a given system. After considering all these conditions, Shapley [524] calculated that there are 10^8 places in the universe suitable for the emergence and existence of life. However, Huang increases this figure to 10^{18} [242]. Brown's calculations show that a great number of the visible stars may have planetary systems with one or two planets with necessary parameters for the existence of life. Even if stricter limitations are applied to the conditions for spontaneous emergence of life than have been considered normal until the present, life could have evolved at least 10 billion times in our galaxy, according to Brown's calculations [55].

EVOLUTION OF CARBON COMPOUNDS IN THE SOLAR SYSTEM

The formation of the solar system and the mechanism of this process cannot, at present, be regarded as completely explained. However, starting with Russell [485], it has been accepted that the basis for the formation of the Sun and its surrounding planets is the process of fusion of diffuse space matter particles [94, 163, 237, 289, 557, 564, 580, 587, 588, 630, 640]. According to these concepts, the starting point for formation of the solar system was the formation of a cloud of gas-dust matter with a size commensurate with the scale of a modern planetary system, a temperature on the order of 50° K, and a weak magnetic field. It contained approximately 1000 hydrogen atoms per 1 cm^3 , a significantly smaller amount of helium, and other inert elements, which together with hydrogen formed the gas component of the cloud. Methane, which is preserved in a gaslike state even at comparatively low temperatures, may also have been

present [29]. The remaining elements of the Mendeleev table were present mainly as components of the dust, comprising only thousandths or even smaller fractions of a percent of the total mass of the accumulation.

The gas-dust cloud was gravitationally unstable; it had a known turbulence and certain central condensation which finally led to its general disintegration. When the mass of the central body precipitously increased, it quickly became very dense and hot due to gravitational energy. The hydrogen-helium reaction then caused it to be transformed into our Sun. The rest of the matter which did not become part of the Sun formed into a discoidal cloud which gave rise to formation of the "protoplanets" [9, 78, 290, 307, 525, 592, 594, 633]. Because of this method of formation, the atomic composition of the Sun is in direct proportion to the composition of the gas-dust nebula which gave birth to it. Hydrogen comprises the main mass, followed by helium, oxygen, nitrogen, and carbon. Subsequent heavier elements are represented in thousandths, tens of thousandths, and smaller fractions of a percent [177, 330, 524].

All substances which were part of the original gas-dust solar nebula can be divided into three groups [53]. In the so-called *gas group* belong hydrogen, helium, and inert gases which preserve their gaslike state at temperatures close to absolute zero. Next comes the *ice group*, which includes compounds formed from carbon, nitrogen, oxygen, and hydrogen, such as water, ammonia, and methane (the latter might also be partially assigned to the first group). Finally, a group arbitrarily called the *earth group* composed of compounds of silicon, magnesium, iron, and other heavier elements. The ratio of the relative mass of these groups in the Sun may be approximated as:

If the *earth group* is taken as unity, then the substances in the *ice group* will contain 4-7 times more and those in the *gas group* 300-600 times more mass [53, 56, 565].

Different ratios were created in the protoplanetary discoidal cloud [590, 591, 633]. Its temperature depended strongly, first, on the ever-increasing solar radiation, and second, on

the loss of heat from the cloud by radiation into interstellar space. Its temperature at the periphery thus differed strongly from the central regions of the gas-dust disk. This gradient must have determined the nature of distribution of protoplanetary material in the disk [54]. Nearer the periphery, where intensive cooling occurred, substances of the ice group condensed at a great rate and froze to the earth group particles, forming the so-called *comet clusters*. In the region of Jupiter's orbit and farther towards the periphery, gas, dust, and the comet clusters quickly fused into large planets, thus determining their chemical composition [633].

Formation of Planets

The planetary building processes must have developed differently in those regions of the discoidal cloud closer to the Sun and in the region of the Earth-type planets. Here the gas component of the cloud was almost entirely lost. The ice group was preserved only partially, and the principal material was the earth group. The accumulation of live dust particles consisting of this material led to formation of planet clusters—small bodies whose composition included all the nonvolatile substances of a primary dust cloud—silicates and their hydrates, metals (in particular iron), their oxides, sulfides, carbides, and live particles of natural carbon, plus components from the ice group—water in the form of hydrates of ammonia salts and organic substances (hydrocarbons). The later fusion of planet clusters led to formation of Earth-type planets and asteroids [230, 328, 536, 565, 593].

Planets which emerged in this way already differed from one another in the initial material of which they were formed. This difference was magnified later by each one having had its own subsequent complex history before becoming what it is now observed to be [469, 489]. This also influenced decisively the evolution of carbon compounds belonging to the planets. Jupiter in its very deep and turbulent atmosphere contains large quantities of ammonia, methane, hydrogen, and possibly also water vapor [6, 389, 556, 634, 635]. Based on the presence of these substances, Jupiter might serve as a natural model for study

of the evolution of organic substances [488], especially because within its enormous cloud layer there must be a zone where relatively mild temperatures prevail [338].

Recent laboratory tests simulating conditions possible for Jupiter have shown that under these conditions, coupled with electrical charges and ultraviolet light, complex and polymerized organic substances are formed within corresponding gas mixtures [437, 451, 499, 637]. However, Lewis and Prinn [310] conclude that the results of these laboratory tests cannot be directly applied to the modern atmosphere of Jupiter. Conditions actually existing there contradict possibilities of the preservation of organic substances and, even more so, preclude the emergence of life. Conditions similar to those on Jupiter evidently exist on Saturn and on other large planets, but for them, it should be taken into account that as the distance from the Sun increases, their illumination by solar radiation decreases [207, 288, 349, 481, 489].

Different composition and different evolutionary paths for the carbon compounds are inherent on the Earth-type planets. Mercury is the least studied planet. It is bare rock, devoid of atmosphere, and thus is open to the entire spectrum of solar radiation. Its surface facing the Sun is heated to 350° C, while the opposite side is cooled to -170° C, which not only prevents the emergence of life, but also makes doubtful any further evolution of carbon compounds which would be blown away by the solar wind [5, 36, 105, 235].

Venus. In contrast to Mercury, Venus has a very extensive atmosphere, closed at the top by a dense cloud layer which has prevented direct telescopic study of its surface. Only radio-astronomical investigations have permitted some degree of knowledge on composition of the atmosphere and temperature which prevail [50, 254, 276, 327, 388, 431, 535]. Such data show that although Venus was formed from the same planet cluster material as the Earth, is approximately the same size, and is in an orbit located next to the Earth's, it cannot serve as a natural model for the study of evolutionary paths of Earth (see, however, [354]). Venus, initially a cold body, did not undergo internal stratification, according

to Fesenkov. Therefore, in contrast to what took place on Earth, the long-lived radioactive elements on Venus remained distributed more or less uniformly throughout the entire mass, which caused a gradual heating which has continued to this day [133].

Mars. This planet has greater promise, with more similarities to Earth [442, 652]. It has long attracted attention because of possible emergence and existence of life; consequently, the scientific literature on Mars is exceptionally large [175]. However, numerous early suggestions had to be rejected, for example, the proposed biologic indications of seasonal changes resulting from intense summer development of vegetation on the surface of Mars. A summary of the pertinent data [28, 35, 81, 175, 214, 268, 304, 337, 361, 430, 491, 539, 571] permits characterization of the planet Mars:

The principal component of the martian atmosphere is carbon dioxide (CO_2), which is judged to comprise 50–100% of the atmosphere [35, 270, 355]. Nitrogen does not exceed 5% [92], and very small amounts of water [409, 583] and CO [255] have been observed. The presence of argon in amounts comparable to CO_2 is theoretically probable; however, there are no observational data.

The martian atmosphere is rarefied, approximately 100 times less dense than the Earth's atmosphere at its surface [213, 267, 269, 470]. The mean temperature over the planet at the bottom of the atmosphere is approximately equal to 200°–210° K. In the summer at the Equator, the temperature may reach +30° C in the daytime, but may fall the same night to –70° or –80° C. Ultraviolet radiation from the Sun with a wavelength of 1900 Å reaches the surface of Mars if its atmosphere is not obscured by sandstorms. The surface layer of Mars is formed of a sand-dust cover, which contains a significant amount of limonite. The external appearance of Mars greatly resembles the Moon. Basically, it is a desert devoid of water with significant elevations and depressions, entirely covered by craters of various sizes, ages, and origins, which in many cases have more gentle slopes than those

on the Moon, indicating their active disintegration by winds and great thermal extremes.

The poles of Mars are covered with white caps which consist of solid CO_2 with a water-ice underlayer [214, 361]. The white caps thaw in summer, which is related to the darkening of so-called seas which extends to the Equator. The small amount of water, complete permeability of the atmosphere for shortwave ultraviolet light, and the low temperatures with their great even drops would create very severe conditions for life of terrestrial organisms. But these conditions do not exclude completely the possibility of the existence of life on Mars. Some authors have suggested that Mars has undergone substantial changes in the course of its planetary history. In the initial periods, it may have been more like Earth, and the emergence of life occurred there the same as on our planet. Once emerged, the organisms may have gradually adapted to the increasingly bleak conditions of the martian surface [528, 590].

Meteorites

Recent extensive studies of meteorites have provided the most significant information on the material from which the Earth was formed and, in particular, on those initial carbon compounds whose evolution in our planet led to the emergence of life. This is true since until recently, meteorites were the only extraterrestrial objects which could be subjected to direct chemical and mineralogical analysis. Their composition must be very similar to that of the planet clusters which serve as initial material for formation of Earth-type planets. Extensive reviews of modern works on meteorites comprise hundreds of articles and books [12, 13, 263, 356, 603].

The origin of meteorites is a problem that cannot be considered completely solved. But it is accepted as highly probable that they were formed in our solar system in the zone of the asteroids which apparently are the "parent bodies" for them. The time of their formation is near that of the solar system. All meteorites can be divided basically into two groups—iron and stone. The first ones supposedly originated from the central part of the "parent bodies" and the

second ones from the peripheral part. All contain some amount of carbon and its compounds [293, 331]. Iron meteorites contain carbon in its natural form or in a compound with metal (cohenite—FeNi, CO_3C). Stony meteorites contain an average of about 1% carbon. Carbonaceous chondrites, in which the content of carbon may reach almost 5%, are even richer; a significant part of chondrite carbon is in the form of organic substances [206, 601, 612] which may amount to 7% of the carbon; approximately 10% are soluble in organic solvents. These are called bitumenlike substances—basically a complex mixture of hydrocarbons [600].

The hydrocarbons of carbonaceous chondrites were studied by Studier [562] and Oró [293, 366, 407] with the aid of a combination gas chromatograph and mass spectrometer. They showed that the distribution of hydrocarbons in carbonaceous chondrites is somewhat similar to their distribution in ancient sedimentary rocks, also to products of abiogenic synthesis according to Fischer-Tropsch. In all the carbonaceous chondrites which Oró studied, isoprenoid hydrocarbons were detected in a small amount, in particular pristap, thytat, and norpristap, which at one time were considered indicators of the biogenic origin of given mixtures of organic substances [75]. However, using the meteorite Groznaya, Vdovykin [603] and Oró [400] showed that these substances have an abiogenic nature and are not the result of contamination under terrestrial conditions. In a model experiment, these substances may even be formed by means of inorganic synthesis [357]. Other organic substances have been detected along with hydrocarbons in the carbonaceous chondrites [202, 203, 206, 224, 299, 308, 365, 366, 398, 560, 561, 600, 606, 617, 650], in particular aromatic and fatty acids, sulfur- and chlorine-containing organic substances (phiophenes and alkylchlorides), cyclic nitrogen compounds, purines and triazines, porphyrins [223] and amino acids [256]. Detection of the latter substances evoked especially heated discussions in the literature, because they could easily have been terrestrial contaminants—in particular as traces from the fingers of the experimenters. In numerous instances the amino acid composition of meteoritic

samples corresponded to that of such contamination [188, 540].

The extraterrestrial origin of amino acids has been proved often. Particularly persuasive are studies of the Murchison meteorite which fell in Australia and was immediately subjected to analysis. Along with the predominant amino acids normally encountered in chondrites—glycine, alanine, glutamic acid, and proline—small amounts were found of 2-methylalanine and sarcosine, which are not component parts of Earth proteins, but arise during abiogenic syntheses. Twelve nonprotein amino acids were later found which could have originated only in the meteorite itself or, more accurately, even before its formation during the action of solar radiation on primordial dust [296, 297]. All the amino acids found in meteorites are optically inactive, with the exception of those that were injected with terrestrial contaminants. A number of other signs distinguish meteoritic organics from substances of the most ancient deposits on the Earth. The age of meteoritic substances is calculated at approximately 4.5 billion years [332]; consequently, they are 1 billion years older than the carbon compounds found on Earth.

On the basis of morphologic similarity in carbon chondrites, Claus and Nagy [85, 359], in the early 1960s, described microinclusions which luminesce in ultraviolet rays, which might be expected of residues from space microorganisms which could have been generators of organic substances in meteorites. However, now it can be considered sufficiently well-established [137, 602] that the so-called organized elements of carbonaceous chondrites are sometimes morphologically very similar to the residues of microbes, or in some cases, are particles of mineral origin or small clumps or carbonaceous matter, or in other cases, the results of terrestrial contamination.

Almost all carbonaceous compounds which have been identified in carbonaceous chondrites may be produced in the Fischer-Tropsch reaction from a mixture of CO , H_2 and H_3 in the presence of nickel, iron, or a magnetite catalyst [562]. A number of organic compounds of great biological interest, detected in meteorites, were also synthesized by Hayatsu, Studier, and Anders

from CO, D₂ and ND₃ in the presence of nickel-iron and aluminum catalysts. They were able to identify p-alkanes, iso-, anti-iso, and dimethyl-alkanes, adenine, guanidine, xanthine, glycine, alanine, asparaginic acid, glutamic acid, tyrosine, histidine, pyrrolic polymers, and other organic compounds [202]. It is very important that the substances obtained in this manner show carbon-isotope fractionation of the same sign and of the same amounts as observed in carbonaceous compounds of meteorites. These studies show that the organic substances in meteorites could have been produced in a catalytic reaction between CO, hydrogen, and ammonia in the solar nebula [299]. Thus, the organic substances of carbonaceous chondrites differ from biogenic organic substances of rocks by their carbon-isotope composition, absence of optic activity, different ratios of separate components of their compounds, presence of chlorine, and by a number of other peculiarities [604]. All characteristics point to their abiogenic nature and that they were formed during agglomeration of the initial material for meteorites.

Lunar Samples

The second nonterrestrial objects, after meteorites, subjected to direct chemical and mineralogical study were lunar soil samples supplied by US astronauts and the Soviet automatic stations Luna 16 and 20 [80, 118, 294, 308, 554, 570, 616, 639]. The age of the lunar dust samples from the Sea of Tranquillity lies within 3.7 to 4.6 billion years. In this respect, they occupy a place between the oldest samples of rocks on the Earth and meteoritic material. Thus, lunar organic substances, if they were found, would be of special interest, because they would belong to the period which has not been studied so far. They could have either an abiotic character [653] (abiogenetically formed on the Moon itself) or have accumulated with meteorites which have fallen on the lunar surface, or, they might have originated as a result of the vital activity of organisms which exist or did exist on the Moon [170]. Unfortunately, analyses of the lunar material yielded disappointing results in this regard.

The total carbon content in lunar dust is not slight; on the average, it is expressed as 150 ppm [127, 257, 350]. For comparison with terrestrial rocks, this amount is approximately equal to 200 ppm on Earth [333]. However, per portion of organic substances in lunar dust, it is less than 2 ppm. No sugars, nucleic acids, fatty acids, or amino acids could be detected [167, 295, 408]. However, with the aid of a very sensitive amino acid analyzer, Fox et al determined the presence of a minimal amount of amino acids (20–70 parts per billion of lunar soil) in samples taken by Apollo 11 and Apollo 12 from trenches, among which the presence of glycine, alanine, and so forth can be presumed [157, 195, 358]. Traces of porphyrins were also detected in lunar dust (Apollo 11), but it turned out that these did not belong to the Moon but were synthesized in the rocket gases [225, 226]. The hydrocarbons detected in lunar dust during acid hydrolysis could have been generated from carbides, which are important components of the carbonaceous complex of the lunar surface [15].

No microstructures were detected on the Moon which would confirm the presence of organisms living at present or in the past [24, 504]. The isotopic composition of the lunar carbon is very unusual; evidently lunar carbon is a mixture of carbon of local origin, meteorites, and solar wind [299, 323]. The abnormally high content of ¹³C in lunar dust cannot be assigned to a carbonate phase because the amount of carbonates, if present at all, is exceptionally small. Present-day carbon on the Moon, on the basis of the chemical and isotopic composition, evidently did not originate from carbonaceous chondrites, which have continuously fallen onto the lunar surface for billions of years. Possibly lunar carbon compounds came from iron meteorites which contained these substances. However, mineralogical studies show only very small inclusions of nickel-iron meteoritic material [258]. Since carbon on the Moon is represented not by carbon-hydrogen heteroatomic compounds, but principally by carbides, oxides, elementary carbon, and a high enrichment of ¹³C, there are grounds to propose that it had a unique history on the surface of the Moon. In this connection, the organic substances of the original matter and of

the carbonaceous chondrites were strewn on the completely unprotected lunar surface by solar wind and were not retained. Only the more stable and heavy compounds of carbon were preserved.

Thus, both interstellar space and clouds of gas-dust matter are quite rich in carbon and organic substances, which were also present in the protoplanetary discoidal solar nebula. But in those regions where prevailing higher temperature formed, the gaseous components—hydrogen, helium, and other inert elements—must have escaped to the peripheral regions of the nebula in significant amounts. Methane also must have met such a fate.

The content of inert gases on the Earth is very small in comparison with their content in space [52, 563]; their abundance was highly depleted as a result of volatilization from that part of the protoplanetary cloud in which the Earth was formed. This volatilization occurred even before the Earth was formed as a planet, under a very low gravitational field. According to Urey [589], this explains depletion of the neon content on our planet to a significantly greater degree than the heavier xenon. But if the depletion proceeded entirely by means of fractionation by mass, i.e., if volatilization from a gravitational field took place according to molecular weight, then CH_4 would be expected to be as depleted as neon, but in fact carbon is preserved on the Earth to a much greater degree. This indicates that it was retained during the formation of our planet not as methane, but in the form of heavier chemical compounds—in particular as organic substances which had been synthesized under conditions of the interstellar medium in the protoplanetary nebula. These cosmic organic substances, according to their origin, entered the composition of the planet clusters which served as material for the formation of Earth-type planets and asteroids. Each of the planets which arose in such a manner from a similar material later had its individual, long, and complex thermal history, during the course of which the initial organic substances evolved in a specific manner for each planet. This history determined the composition of its modern carbonaceous compounds. The thermal history

of the asteroids, because of their small weight, was less significant and therefore the meteorites which arose during their crushing evidently retained their original composition and structure to a great degree.

Space research provides information, to a great degree, on those abiogenic carbon compounds which were starting compounds for the further evolution of Earth organics on the path to the emergence of life. However, research is still insufficient to understand these paths well. In order to understand them, the modern geochemistry of carbon compounds may be studied and the behavior of organic substances modified under conditions which might have taken place on the Earth at the beginning of its existence.

EVOLUTION OF ORGANIC SUBSTANCES ON THE EARTH

The formation and initial stages in the evolution of the Earth as a planet are being extensively studied at present, from both the geological and geochemical points of view [22, 29, 49, 117, 130, 309, 487, 587, 588, 593, 594, 607, 608, 614, 615]. Nevertheless, this stage of evolution is still far from adequately understood and evokes strong differences of opinion. According to current widely held concepts, our planet was formed by accumulation of cold solid bodies with varying contents of iron and silicates, but basically lacking such free gases and volatile compounds as molecular hydrogen, light inert elements, and methane. They were lost previously in that region of the gas-dust nebula in which the Earth was formed. Only a small number of these gases could be preserved in the Earth's composition to the degree that they were adsorbed by solid components of the nebula. They could have formed the primary atmosphere of the Earth. However, this atmosphere could not have been long-lasting because its component gases could not be retained by the Earth's gravitation. Because of such loss, the Earth developed its modern mass and the composition which is now characteristic of it—one which is close to the average composition of meteorites.

The remaining compact mass of the Earth continued to evolve through its internal energy,

at which time the mass divided into the core, mantle, and crust. The mechanism for such differentiation is quite characteristic of our planet, but remains far from clear. A hypothetical secondary melting due to the initial gravitational heat and radiogenic warming is advanced most frequently as an explanation. However, complete melting of the Earth by such means would require several billion years. Actually, the differentiation occurred within an incomparably shorter time. It has been proposed that "short-lived" and presently "frozen" isotopes, such as ^{10}Be , ^{26}Al , ^{129}U , ^{244}Pu , and so forth, played an important part in the complete or partial melting of the Earth. In any case, the entire subsequent course of the evolution of the planet was determined by its thermal history. During its formation, the principal sources of internal heat were gravitational energy given off during agglomeration and compression of the Earth's matter, and energy formed during the decay of radioactive elements which were not distributed throughout the entire mass, but basically displaced toward the Earth's periphery.

The most important boundary in the Earth's development was the surface formation of a crust, atmosphere, and hydrosphere. The modern Earth crust consists of a granite and basalt shell. The so-called mantle, of low-grade silicon and ultrabasic rocks (dunites), underlies the crust. On the basis of tests on the zonal fusion of the substance of stony meteorites, Vinogradov [609] became convinced that the dunites of the mantle are residues from melting of the original substance of the Earth, which is similar to the composition of chondrites. Light basalt rocks melting from a mantle stratum due to the effect of radiogenic heat should have been accompanied by a discharge of various vapors and gases due to their evaporation as the temperature increased, or due to their formation in solid ground shells during radioactive, radiochemical, and chemical processes. Therefore, the formation of aqueous and gaseous shells of the Earth (its hydrosphere and atmosphere) was closely connected to processes in the lithosphere from the very beginning.

The amount of water on the surface of the Earth

during its early existence was not great [483, 588]. The basic mass of water which formed the world oceans appeared only at the very beginning of the Archean era, immediately after formation of the Earth's crust. It developed gradually during formation of the lithosphere from silicate hydrates or generally from bound constitutional water of the Earth's interior [482, 613]. Archean waters differed substantially in chemical composition from modern waters. Acid gaslike products—HCl, HF, boric acid, H_2S , CO_2 , CH_4 , and other hydrocarbons which had been discharged by volcanoes were dissolved in them, thus they had an acid reaction. The composition of the water of the ancient oceans began to approximate modern composition only toward the middle of the Archean age. The acids became neutralized after being subjected to the action of silicates of sedimentary deposits and carbonates of K, Na, Ca, Mg [553] (see, however, [484, 534]). In spite of the Earth not having free, molecular hydrogen during its formation, it retained this element in the form of diverse compounds which imparted a regenerative character to both the lithosphere and to the secondary atmosphere which had arisen.

Molecular oxygen, characteristic of the modern atmosphere, was practically lacking [87, 230]. Only a very small amount of it could appear during photolysis of water vapors by short-wave ultraviolet rays [199]. However, even in this case, oxygen could not accumulate in the atmosphere to any significant amount, because it was absorbed very quickly and completely by rocks which were unsaturated with respect to it [302]. Such a process of O_2 absorption can be observed even now. For example, lava masses ejected onto the Earth's surface during modern volcanic eruptions are normally very rich in metal oxides, so that lava and basalts are of black and green color. The sedimentary formations from them (red or yellow sands or clays) are rich in the higher oxide form of iron. Thus, when free oxygen is abundant in the modern atmosphere, it oxidizes only the very surface film of the Earth's crust; if it were not for the constant biogenic formation of this gas, it would have disappeared from our atmosphere in a very short time, since it is absorbed by rocks [179].

Thus, before the formation of biogenic oxygen [47, 610], the secondary atmosphere had no free oxygen and was reducing in nature. This may be ascertained from direct geologic data obtained by studying ancient Precambrian deposits. Rutten [486] divides the history of our planet into two basic periods: actualistic, similar to the modern, and preactualistic, the period preceding it. During these periods, geologic processes which occurred on the Earth's surface differed a great deal. For example, during the modern actualistic era, in an atmosphere rich in free oxygen, chemical processes play the chief role in weathering basic rocks. Minerals already oxidized were transported and deposited elsewhere. On the contrary, if the atmosphere had a reducing nature (proposed for the preactualistic era), minerals of the basic rocks were only worked over physically and deposited in a chemically unchanged form. Studies from this point of view of a number of deposits more than 2 billion years old show convincingly that they formed under conditions which lacked free oxygen [230, 260, 306, 467, 468].

Studies of gases discharged by modern volcanoes and fumaroles provide some concept of the possible enrichment of the ancient atmosphere of the Earth by CO_2 and CO . In these discharges, carbon is represented principally in its oxidized form, and methane comprises only a small admixture [324, 543, 544]. Their source could have been graphite scattered in basalts, also organic substances of cosmic origin. Carbon could oxidize due to H_2O . The gases CO_2 , CO , CH_4 and H_2 which are frequently detected in cavities of ejected rocks also originated in this way. At high temperatures, the equilibrium shifted in favor of CH_4 , and at lower temperatures, in favor of CO_2 . Therefore, ancient volcanoes ejected CH_4 as well as CO_2 and CO . Methane and carbon monoxide shifted principally to the atmosphere, and CO_2 was distributed between the atmosphere and water [611, 613]. However, a significant part of the carbon monoxide dissolved in ocean water interacted with silicates and then shifted to residues in the form of carbonates. This was in contrast to what might have happened in the atmosphere of Mars, which lacked open water.

In summary, it is possible to conclude that the Earth, from the very beginning of its formation, was enriched by both natural and carbide carbon as well as by cosmogenic organic substances—frequently by quite complex, heavy molecules. Miller and Urey [346] noted that these organic substances, during gravitational and radiogenic warming of the Earth, had to be subjected to pyrolysis to some degree. The equilibrium mixture which originated on this occasion (CH_4 , CO_2 , CO , NH_3 , N_2 , H_2O , H_2) was sublimated onto the Earth's surface and served as material for secondary synthesis of organic compounds under the action of radiant energy and other energetic factors. However, it has become obvious that even in the very depths of the Earth there was not only decay (which evidently continues), but also synthesis of organic substances as a result of the Fischer-Tropsch reaction [202, 299] and similar abiotic processes [110].

Abiogenic Synthesis

To understand the totality of these complex, abiogenic, organochemical transformations which occurred at some time on the Earth has been rendered extremely difficult. Life originated and developed luxuriantly on the surface of our planet, where it forms a zone of life—the biosphere. Within the limits of this zone, the prebiochemical evolution of organic substances under natural conditions cannot be directly observed.

After emergence and further development of life, an abrupt change of conditions on the Earth's surface excluded any possibility of prolonged evolution of organic substances in the same ways that this evolution had occurred during the abiotic epoch. If abiotic organic substances could somehow now emerge, they would quickly be destroyed, eaten by microorganisms which populate the soil, water, and air everywhere. Charles Darwin pointed this out in one of his letters [93]. No matter how paradoxical it may sound, the main reason that life originates on Earth is the fact that it has already done so [367] (see, however, [262]). Since life developed hundreds of millions of years ago, the possibility is eliminated of studying abiogenic

syntheses in a pure form under natural conditions not only within limits of the biosphere, but also in the most ancient deposits of the Earth's crust. Organic substances of biogenic origin accumulate here, resulting from the vital activity of organisms—a process which is exceptionally vigorous and completed at much faster rates than abiogenic synthesis.

The basic difficulty is that there are no available chemical methods to distinguish reliably between biogenic and abiogenic organics. Only a few years ago, the detection of porphyrins or other complex organic compounds, not only on the Earth but on any space object, was considered convincing proof of the presence of life. For example, in 1966 Urey wrote concerning organic substances found in carbonaceous meteorites: "evidently, it is safe to say that if such material would have been found on the Earth, nobody would have raised the question of biological origin" [595].

This position has changed radically. At present, not one organic compound discovered in ancient terrestrial deposits could, by itself, serve as a reliable indicator of its biotic or abiotic origin. This concerns the isoprenoids in particular, e.g., pristane or phytane, which at one time Calvin [73] considered indicators of biologic origin. These substances may be synthesized in an abiogenic way [33]. The more ancient the rocks which are studied, the clearer the indication of abiotic processes should be. In a study of petroleum samples from different deposits, Robinson noted that crude petroleum of more ancient origin contains substances which lag far behind biochemical compounds, but arise easily by abiogenic means [477, 478, 479].

However, this position is now far from being universally convincing. The lack of reliable chemical criteria for the biogenic or abiogenic origin of natural organic substances has caused serious arguments between proponents of the mineral and biologic origin of petroleum [119, 209, 285, 286, 453, 454, 575, 598, 599]; both viewpoints are extreme and cannot be considered acceptable. It is understood that organic substances which are synthesized by organisms and entered the composition of sedimentary rocks, after having undergone a number of changes in

the process of geochemical decay and secondary synthesis, provide a beginning for the formation of petroleum sources. It cannot be denied that under the complex conditions of the Earth's crust, numerous abiogenic syntheses occurred (and continue), which are based on mineral, catalytic processes for which both inorganic forms of carbon as well as products of the decay of modern and ancient organics have been, and are, the initial raw material.

As life developed, biogenic processes acquired increasingly greater importance during the synthesis of organic substances; but in the ancient periods of the Earth's existence, in particular before the emergence of life, abiogenic processes had an exclusive importance and only later were obscured by biosynthesis. From the viewpoint of the origin of life, organic substances which formed in basic rocks could be considered examples of inorganic synthesis that are particularly interesting. Works on the geochemistry of organic substances are reviewed extensively in *Organic Geochemistry* [120], so that only supplementation is needed with references to the proposed inorganic syntheses. Specific finds in this area are fully described in the scientific literature; only summary mention of them will be made here.

Kropotkin and Shakhvarstova [283] describe petroleum occurrence in basic rocks of various countries and regions—Cuba, Mexico, California, South America, Turkey, New Caledonia, France, Brazil, Australia, South America, and the Soviet Union. Florovskaya et al [140, 143], give data on the distribution of carbon substances in different formations of the Earth's crust, including magmatic rocks, pegmatites, and hydrothermal veins. There are important monographs on hydrocarbon gases and bitumens in rocks of the khibinite alkaline massif, on the inclusion of organic substances in minerals, of carbonaceous compounds in rocks or crystalline shields and of the crystalline foundation of plates [141, 142, 245, 284, 428, 429]. Hydrocarbons from Lake Trinidad asphalt and formations of Mount Sorrel, which evidently do not have a biological origin, are of great interest [446, 447]. Most of these authors consider such finds proof of the abiogenic origin of the organic compounds

detected; but since there are no reliable chemical features in the substances found, these conclusions are made only on the basis of geological concepts and in certain instances can be interpreted otherwise.

Biogenic Synthesis

Solving the problem of emergence of life on Earth involving attempts to trace the consecutive stages of chemical evolution and its transformation into a biological form of organization appears to be a good approach, and is on the same basis as that which paleontologists follow to study the Darwinian evolution of life. Such investigations seek simpler forms of organic materials which have been preserved in the more ancient zones of the Earth's crust. The basic purpose of all related work is to link the increasingly complex chemical formation with the most primitive forms of life. Serious difficulties are encountered in this method. On the one hand, the assumption is that the further back in paleontological time, the more primitive will be the morphological structure of the formations discovered. In addition, many colloid substances which have no relationship to life might, under artificial and natural conditions, easily form structures which externally are very similar to microorganisms. The "artificial cells" of Herrera [215, 216] are examples. This author mixed solutions of sulfo-cyanates and formaldehyde, then fixed the colloids obtained with alcohol. Residues were formed which even expert cytologists recognized as fixed microorganisms belonging to various classes of living substances. Therefore, in examining the most ancient structural formations, great care must be exercised to determine whether they are residues of primitive organisms or artifacts which originated from organic or inorganic colloidal mixtures. On the other hand, when any organic substances in the most ancient deposits of the Earth's crust are discovered, it can never be said with complete certainty whether they originated abiogenically or are products of the vital functioning of organisms which might have lived at one time. Extensive reviews are devoted to the most ancient fossils [25, 75, 88, 261, 487, 505, 619, 620].

Petrified remains of animals and plants preserved from the beginning of the Cambrian period clearly picture the irreversible process in development of the organic world, ongoing continuously over 550 million years. Not long ago, it was still thought that the paleontological chronicle had been broken off during the Cambrian age, and that the remains of organisms which had populated the Earth more than a half billion years ago had not been preserved to the present because the rocks which contained them had been subjected to profound alterations and metamorphosis which completely destroyed all biological structures. However, it has been possible recently to establish that in many places on the Earth, for example in Southern Australia and Africa, on such platforms as the Russian, Siberian, and Chinese, and beneath layers with the most ancient Cambrian complexes, there are strata closely related to them which contain more ancient fossil remains.

Investigation indicated that late Precambrian life was a direct predecessor of the Cambrian. The fossils discovered here point to the existence, at that time, of not only a rich flora, but also fauna, represented by comparatively highly developed unicellular as well as multicellular organisms. In particular, in deposits 650 million years old, in the region of Ediacara Hills (South Australia), impressions and petrified fossils were discovered of frozen embryonic jellyfish, soft corals, platyhelminthes and other extinct forms of animals [171, 173, 178]. In the Precambrian deposits of the USSR and China, Vologdin found the most primitive needles of silicon sponges, impressions of worms, and so forth [619]. However, in deeper layers with an age of 800-1000 million years, animal remains are not present, but, in contrast, the vegetative world is represented by numerous fragments and accumulations of varied algae and spores of primitive plants [23, 171].

Stromatolitic Periods

All periods preceding the Proterozoic were evidently an era of bacteria and algae, the most important of which were the so-called stromatolitic formative periods. Stromatolites are unique

limestone formations in the form of relatively rectilinear cupolas consisting of a number of cones built on top of one another. They are products of the vital activity of ancient photosynthesizing organisms, principally blue-green algae [172, 198, 259, 492, 618]. Early stromatolite-formers played an essential role in the gradual enrichment of the Earth's atmosphere with oxygen. However, in their initial existence, the oxygen they formed did not necessarily go into the atmosphere but could have been entirely absorbed by chemosynthetic iron bacteria also present in the stromatolite, which were in symbiosis with the blue-green algae. As a result of seasonal fluctuation, a laminated structure of stromatolites formed in which limestone alternates with iron hydroxide [621].

Deposits of limestone secreted by organisms which have been found in the Sahara are classified as the beginning of stromatolites, which are about 1.5 billion years old [42, 181]. Formations similar to the stromatolites were found by MacGregor in dolomite series of Southern Rhodesia [317], with age determined at 2.7 billion years [363]. Actual remains of organisms were not present. The conclusion that these formations developed from algae is based only on external similarity to stromatolites. However, it is completely possible that the producers of limestone secretion in Rhodesia had no definite morphological structure but were only masses of abiogenic colloidal material or probionts which preceded life.

The most representative accumulation of fossil remains of authentic organisms of the Precambrian period were discovered in layers of black shale near the foundation of Gunflint iron ore deposits around Lake Ontario [23, 27, 86, 90, 586], whose age has been estimated at 1.9 billion years [243]. Numerous unicellular and colonial forms were detected (both branched and unbranched), which closely resemble modern blue-green algae. Numerous spherical formations resembling spores, as well as the remains of organisms are found which have no similarity to modern living creatures. Bacterialike formations, in particular, similar to modern iron bacteria, were also discovered electron-microscopically with the aid of the replica method [506].

The Sudan formation (Minnesota, USA)

belongs to carbon-rich ancient sedimentary formations, with an age of about 2.7 billion years [16]. Structures which might be taken for the remains of algae or bacteria were found here in gray pyrite globules, but these could be simply structures of abiogenic colloidal material [89]. Even more ancient finds (3.1 billion years old) were found in Fig Tree deposits (Eastern Transvaal). Although the fossil forms found here are very sparse, nevertheless some of them recall modern small bacteria when shown singly on replicas [26]. Barghorn called them *Eubacterium isolatum*. However, there is no certainty that these formations are true fossils and not artifacts. Thus, Rutten [487] cautiously suggested calling them "organized elements." There is even more doubt about the carbon structures found in the Onverwacht deposits (approximate age—3.7 billion years) [126, 360].

All the material presented on the development of life before the Cambrian age shows that it took place according to the same principle (from simple to complex) as during the later epochs which have been well studied by paleontologists. The forms of life discovered are simpler and more uniform the further back in time one goes. During the late Precambrian age, life was represented by various multicellular creatures, by both plants and animals (Ediacara Hills). A flourishing development of stromatolite producers—blue-green algae and bacteria—was found in the earlier epoch of the Proterozoic (1–1.5 billion years ago). During the Gunflint epoch, various representatives of these organisms were both unicellular and confervoid in shape. However, at the beginning of the Archean era, there were evidently only unicellular forms. Much earlier finds—"organized elements"—may have represented probionts or even accumulations of abiogenic colloidal material. Thus, the beginning of life is considered a transition from the preceding chemical evolution to a biological one.

Molecular Paleontology

It is tempting to link those investigations based only on morphological data with a chemical study—the so-called "molecular paleontology." Such investigation in the area of molecular

paleontology has begun comparatively recently, but numerous works already exist in the scientific literature in which both the oldest deposits mentioned above, as well as the many other formations of later origin, have been subjected to detailed chemical analysis for the purpose of establishing the intramolecular structure of the organic substances contained within them [3, 23, 34, 37, 48, 66, 67, 68, 74, 116, 119, 160, 187, 197, 227, 228, 250, 336, 406, 432, 455, 500, 519, 520, 521, 522]. In this way, many classes of organic compounds have been subjected to analysis: carbons, fats, amino acids, with special attention paid to hydrocarbons as the substances which have been preserved for the longest time.

In *Chemical Evolution* [75, 76], Calvin summarizes extensively works on the molecular paleontology of hydrocarbons, giving the results of analyses with gas chromatography and mass spectroscopy of many hydrocarbons extracted from comparatively ancient objects:

Site	Age, years
San Joaquin petroleum	30 million
Green River shale	60 million
Nonesuch ancient rocks	1 billion
Gunflint	1.9 billion
Sudan	2.7 billion
Fig Tree	3.1 billion
Onverwacht deposits	3.7 billion

Various compounds related to normal hydrocarbons, isoalkanes, anti-isoalkanes, cyclohexylalkanes, isoprenoids, and polycyclic isoprenoids were found in these deposits.

Isoprenoid phytane (C_{19}) and pristane (C_{20}) were given particular attention since they were once considered substances of exclusively biological origin similar to the so-called biological markers. However, it has since become known that these isoprenoids may also originate in abiogenic reactions and even in space objects. In general, it is not known at present if the molecular structure alone of such hydrocarbon compounds may be reliable proof of their biogenic origin. In order to discover the biogenic or abiogenic nature of hydrocarbon mixtures which have been isolated from objects of various geological age, Calvin uses the following approach.

For modern living creatures, it is characteristic that they synthesize organic sub-

stances selectively. Of a large number of thermodynamically possible isomers in the process of biological metabolism, only very few are synthesized, and that is determined by the specificity of the action of enzymes. In analyzing hydrocarbon mixtures which have been obtained from known biological objects, gas chromatograms are produced with clearly pronounced sharp peaks, each of which corresponds to a specific compound. In contrast, a nonspecific formation of all possible isomers might be expected during inorganic syntheses. Thus, the gas chromatogram of such a complex mixture would be expected to have the appearance of a continuous curve characteristic of the statistical distribution of hydrocarbons.

Such a chromatogram is actually produced from a mixture of hydrocarbons generated in carbide solution in hydrochloric acid [229] or during a spark discharge in methane. A similar continuous chromatogram is also characteristic of the natural abiogenic hydrocarbons of Lake Trinidad and the formation of Montsorrel [447].

The specificity of hydrocarbons found in ancient rocks could, conceivably, confirm convincingly their biological origin [336]; however, the total fraction of alkanes obtained in a Fischer-Tropsch reaction, i.e., by abiogenic means, also gives a gas chromatogram with clearly pronounced discrete peaks for hydrocarbons, which might be judged as biogenic. Calvin obtained data on the composition and properties of hydrocarbon mixtures isolated from comparatively young deposits (San Joaquin and the Green River) of clearly biogenic origin, and uses these data as the standard of biological formation for more ancient formations as well. In particular, he believes indicators of biogenic nature are: the discrete character of the chromatogram; prevalence of *p*-alkanes with an odd number of hydrocarbon atoms; interrelationship of cyclic and open isoprenoid chains; presence of pristane and phytane; and others.

This similarity with the younger rocks is obvious for the shales of Nonesuch and Gunflint, but the matter is slightly more complex because the Sudan and the Fig Tree shales did not reveal any predominance of "odd" over "even" hydro-

carbons. In addition, Oró and Nooner discovered here the presence of the isoprenoid C_{17} , which is lacking in the younger deposits of biological origin (for which there are biochemical reasons); but this hydrocarbon is formed in the Fischer-Tropsch reaction [405]. This raises doubt about the biological origin of the Fig Tree hydrocarbons, although the discovery there of optically active amino acids favors their biogenic origin [298]. Finally, there is a comparatively continuous chromatogram for the extracts from the Onverwacht deposits, saturated with hydrocarbons, which is characteristic of abiogenically synthesized substances [318].

These studies in molecular paleontology, despite their significance, unfortunately cannot provide a clear picture of molecular evolution or the process of making organic substances more complex. This was accomplished in the earliest periods of the Earth's existence which led to the emergence of those ancient organisms whose residues we now discover in deep deposits of the Earth's crust. Model experiments, however, which reveal the chemical potential of organic substances in various degrees of complexity, and the possible realization of these potentials under conditions near those which existed on the primitive Earth, will help in solving this problem.

MODEL TESTS FOR REPRODUCING ABIOGENIC SYNTHESIS

Many works at present on laboratory abiogenic syntheses of organic substances lead to the subsequent complication of these substances and, thus, to modeling of those processes which might lie at the basis of chemical evolution, which preceded life on Earth. Summaries of these articles are in recent monographs, collections, and review articles [71, 75, 122, 148, 261, 263, 305, 320, 381, 421, 438, 450, 511], and characterize chemical and thermodynamic potentials in carbonaceous compounds from the most primitive representatives to the most complex polymers. These potentials have remained unchanged both in the past and present, but developed differently under varied conditions. Therefore, in the model tests, there is normally considerable effort to reproduce conditions which supposedly existed

on the primitive Earth. However, abiogenic syntheses of biologically important substances could take place within very wide limits of these conditions, in particular at the first stages of chemical evolution. For example, α -amino acids are formed in different gaseous mixtures, with varied energy sources on various space objects. As further evolution moved toward complication of organic compounds, the effect of the external conditions was evermore significant; and in different local areas of the Earth's surface, this evolution proceeded differently. It is generally possible now to enumerate those general conditions which must always be taken into consideration in attempts to reproduce, in model tests, abiogenic syntheses of primary organic substances on the surface of the still lifeless Earth.

Initial carbonaceous compounds. The Earth, during its formation, obtained a significant reserve of abiogenic organic substances of varying degrees of complexity which originated in space. Some, of course, were subjected to pyrolysis during the process of warming of the Earth's interior, and the gaslike products of this pyrolysis were sublimated onto the Earth's surface and entered the composition of the secondary (preactualistic) atmosphere, and partially the hydrosphere of our planet [346]. These gaslike products were CH_4 , CO_2 , CO , N_2 , NH_3 , H_2O , and H_2 , which in one combination or another, serve principally as the starting material for model abiogenic syntheses. Processes which occur in an aqueous medium are studied in smaller dimensions. These investigations are frequently connected to more complex organic compounds which originated on the basis of the primary abiogenic syntheses. The possible abiogenic syntheses which could have taken place in the Earth's crust as a result of catalytic reactions, for example the Fischer-Tropsch reaction, have been even more poorly reproduced [204, 642].

Reductive conditions on the surface of the preactualistic Earth. The Earth's atmosphere was devoid of free oxygen for a long time, which is shown by the geologic data presented [486]. This is confirmed by anaerobic processes being the basis for the energetic metabolism of all modern living creatures. Oxygen-respiration units in aerobes were added on to these processes later

[51, 321, 368], "Among all natural systems," correctly wrote Tsukerkendl and Poling, "only living matter differs by the fact that, in spite of the very essential transformations which it has undergone, it preserves by means of recording within its own organization, a large quantity of information which reflects its history" [581]. Analysis of the modern organization of life shows convincingly that its beginnings were under anaerobic conditions.

Source of energy needed for syntheses. Interaction of organic substances could occur under the preactualistic conditions of the Earth spontaneously, due to fundamental potential energy or in conjugate reactions. In particular, this is probable under conditions of the Earth's crust at increased temperature and in the presence of corresponding catalysts [43, 110]. However, external sources of energy must have played an important role in the organochemical syntheses of that time. Shortwave ultraviolet rays, which reach the Earth's surface due to the absence of an ozone screen in the atmosphere of that time, are predominant [103, 186, 345, 425, 445, 577]. Later, according to Calvin, radioactive emissions of a number of elements and in particular of ^{40}K (in which the primitive Earth crust was very rich [572]) must have been an important source of energy for the abiogenic syntheses. A number of authors have achieved the synthesis of many biologically important substances by bombarding a mixture of primitive gases with very high-energy electrons and, in this manner, simulating radiation by ^{40}K [73, 164, 410, 443]. In this connection, radiation of uranium and thorium might also have played an important role both in syntheses in the Earth's crust and in the hydrosphere [357, 450].

The energy of electrical discharges in the Earth's atmosphere, although relatively small, has been most widely used to this day for abiogenic syntheses. This form of energy was first used in Miller's basic tests [339], then in different variations by others [1, 185, 219, 396, 425]. Biologically important compounds also form during simple heating of exhaust gases and solutions [190, 192, 193, 316, 392, 402, 404]. Undoubtedly, such local heating took place in the primitive Earth on a wide scale, and to a great degree, continues in volcanic activity and hydrothermal

phenomena. To these basic sources of energy, it is possible to add the shock wave in gas during the fall of meteorites [182, 221] and ultrasonic waves which can be an aqueous medium [123, 124, 125, 545].

Decomposition of organic substances. The decomposition of organic substances as well as the synthesis must have been caused by such energy. Thermodynamic equilibrium is eventually established but does not work to the advantage of synthesis. For example, calculations for the surface layer of ocean waters subject to prolonged action of shortwave ultraviolet light would indicate that during onset of thermodynamic equilibrium only negligible amounts of organic substances could be preserved [533]. However, under natural conditions, thermodynamic equilibrium could not become established on the Earth's surface because of a continuous displacement of substances synthesized in one place to another. For example, under hydrospheric conditions, these substances easily pass from the decomposing action of shortwave rays to deeper layers of the water and become absorbed in ground particles, and so forth [379]. Therefore, an indispensable condition for model tests is displacement of substances which are synthesized in one place and accumulate in another. This condition was met in Miller's device [339], in the synthesis of proteinoids by Fox [149], and in Young and Ponnampuruma's model tests [652].

Abiogenic Synthesis of Biomonomers

Various organic substances have been synthesized in laboratory tests—sugars, fats, organic alcohols, aldehydes, acids, and a number of heterocyclic compounds. Particular attention was paid to the formation of biomonomers, i.e., those molecules which are component parts of biologically important polymers—proteins and nucleic acids. Some are amino acids, purine and pyrimidine bases, nucleosides and nucleotides.

Amino acids. Amino acids are formed from primitive gases more easily than any other biomonomers. In particular, this means that α -amino acids are synthesized in significantly larger quantities than β -compounds (for example, β -alanine) [192, 316, 339, 340, 341]. Amino

acids, which are not incorporated in proteins (and mentioned in connection with the Australian meteorite [296, 297]) are synthesized in even smaller quantities. Several chemical means for their abiogenic formation have been proposed in the literature. Miller proposed a cyanohydrin mechanism, which is confirmed by the presence of aldehydes and HCN in its reaction products. Ponnampertuma and Weller [452] show α -aminonitrile as an intermediate reaction product. Sanchez et al [494] grant that cyanoacetylene plays an important role; they emphasize the leading role of oligomers of HCN, like other authors [2, 190, 334, 335].

Experiments were carried out in modeling abiogenic synthesis in a mixture of gases of CH_4 , NH_3 , H_2O and H_2 , in certain cases with the exclusion of H_2 or with the addition of CO , CO_2 , N_2 , ethane (in place of CH_4) or H_2S . With this method, the following amino acids were synthesized: cysteine, serine, threonine, α -aminobutyric acid, N-methylglycine (sarcosine), asparagine, asparaginic acid, glycine, alanine, β -alanine, glutamic acid, valine, leucine, isoleucine, alloisoleucine, lysine, phenylalanine, and tyrosine [84, 104, 397, 424, 427, 580]. There are other works in the literature where the synthesis of amino acids was carried out in an aqueous medium with the use of other types of compounds (for example, HCHO , HCN , NH_4Cl , NH_4^+ , NO_3^- , formamide, paraformaldehyde, malonic acid and urea, N-acetylglycine, and so forth [19, 112, 147, 201, 218, 274, 364, 415, 426, 472, 530, 531]).

Components of nucleic acids. In the synthesis of purines and pyrimidines, achievements are less significant than successes with the synthesis of amino acids. In an experiment simulating the primitive Earth atmosphere, Ponnampertuma et al discovered adenine after irradiating a mixture of CH_4 , NH_3 , H_2O and H_2 by an electron beam [443]. No other purine or pyrimidine bases have been synthesized under these conditions, which is interesting to compare with the preeminent position of adenine in all biological systems. Molecular orbital calculations indicate that adenine, of all nucleic bases, has the most resonance energy [459], which, of course, makes its abiogenic synthesis highly probable.

Oró was the first to report on adenine synthesis in 1960 [392]; this work was illustrated in more detail in subsequent publications [402, 403] and confirmed by Lowe et al [316]. The synthesis was carried out under moderate heating (from 30° to 100° C) of a mixture of hydrogen cyanide, ammonia, and water. Oró and others studied the mechanism of this reaction and showed that this condensation occurs through aminomalonitrile (a trimer of HCN) and diaminomalonitrile, or 4-amino-4-cyanoimidazole, (a tetramer of HCN) [129, 402, 495, 496]. Adenine, like another nucleic purine, guanine, was synthesized during irradiation of a weak solution of HCN by UV light [436]. Both these purines are reaction products of 4-amino-4-cyanoimidazole with HCN or cyanogen in an aqueous solution [493, 495], which emphasizes the great importance of HCN in primary abiogenic syntheses. Of the three pyrimidines—cytosine, uracil, and thymine—the first two were obtained in model tests, which simulated primitive Earth conditions to some degree.

Uracil was synthesized during heating of malic acid, urea, and polyphosphoric acid up to 130° C [156]. Oró shows the formation of uracil during heating of urea with acrylonitrile [397]. Sanchez et al discovered the formation of cytosine during the heating of ethylenedinitrile in an aqueous solution of KCN [494]. In a number of these syntheses, other bases were also detected which normally do not enter the compositions of nucleic acids—hypoxanthine, diaminopurine, and xanthine [493].

The carbon components of nucleotides—ribose and 2-deoxyribose—have been synthesized under conditions similar to primitive [159, 323, 399]. Ponnampertuma et al were able to carry out fusion of nucleotide components in an aqueous medium by using ethyl metaphosphate. The syntheses were made in aqueous solutions of adenine, adenosine, and adenylic acid of ribose and ethyl metaphosphate; mixtures were illuminated by ultraviolet light with a wavelength of 2537 \AA at 40° C. In this way, conversion of adenine into adenosine, adenosine into AMP, AMP into ADP, and finally ADP into ATP was established [441, 444, 449]. Even more promising results were obtained by the use of ionizing radiation [436]. There are also other indications

of model tests on the synthesis of varied nucleosides and nucleotides [31, 244, 622] and because of the important role of polyphosphates in model tests, other works should be mentioned [20, 292, 343, 344, 439, 466, 507, 508, 510, 512].

Other monomer substances. The possibility of abiogenic synthesis of other biologically significant compounds has been demonstrated in model tests on a much smaller scale than for amino acids and nucleic components. The syntheses of fatty acids [10, 164, 552] might be indicated here—imidazole heterocyclics [532], isoprenoids [357], and porphyrin [128, 226, 279, 280, 576]. The latter are of great interest to researchers because of their importance in the syntheses of chlorophyll and cytochromes. Optically inactive (racemic) mixtures of organic substances are formed in all abiogenic syntheses in model tests. In contrast, the biogenically formed compounds are always optically active [166], which has attracted attention since the time of Pasteur. At present, asymmetrical syntheses are carried out in the laboratory under the action of circularly polarized ultraviolet light [287] and on the surface of nonsymmetrical quartz crystals [39, 578, 579]. However, Wald doubts that any of the indicated abiogenic factors may create conditions for the emergence of stable asymmetry. In his opinion, this could occur not during the synthesis of monomers, but particularly during the process of higher order structures forming; for example, during selection of proteins and polypeptides which form spirals. The problem of asymmetry has been extensively studied [8, 191, 264, 265, 266, 476].

Abiogenic Synthesis of Biopolymers

The evolutionary step toward the emergence of life following the formation of polymers was their uniting into biopolymers, in particular, into compounds similar to proteins and nucleic acids, which are exceptionally important in organization of living substances [30, 121]. In a number of modern model tests, this association has been carried out by means of dehydration condensation, in which two methods are used to verify the possibility of such condensation under primitive Earth conditions.

The first method uses high temperatures in corresponding nonaqueous solutions; and the second, in searching for compounds whose free energy (also whose affinity for water) might enable them to carry out dehydration condensation of amino acids or mononucleotides even in diluted aqueous solutions. Basically, attention will be concentrated here on model abiogenic syntheses of polypeptides and polynucleotides, although there are similar experiments in the syntheses of polyglycosides, melanine, and so forth [44]. There are extensive summaries of the abiogenic synthesis of biopolymers [75, 261].

Polypeptides and Proteinlike Substances

Fox et al, simulating volcanic conditions, heated a mixture of 18 amino acids to 170° C on a piece of lava for 6 h. An excess amount of dicarboxylic amino acids melted the mixture until it was carbonized. Heating, also in the presence of polyphosphates, permitted reducing the heating temperature to 100° C [155, 158, 194]. Under this temperature, an amber-colored polymer formed with molecular weight of several thousands (from 5000 to 10 000) which contains all amino acids used in the test typical of protein (it lost its optical activity, however). Fox called this polymer proteinoid because a number of features were similar to proteins.

Two very interesting properties of proteinoids were revealed in later studies: first, a high internal molecular uniformity which is repeated by the coherence of amino acid residues (which will be treated in more detail later); and second, a completely, although weakly, detectable experimentally enzymatic activity (catalysis of hydrolysis, decarboxylation, amination, and deamination) [150, 151, 152]. Among numerous works on the synthesis of polypeptides in an aqueous medium, these should be noted: polycondensation of glycine during its heating in an aqueous solution of ammonia [541], during γ -irradiation of N-acetylglycine [112], during thermal treatment of solutions of ammonia cyanide [316], during heating of the mixture of HCN, NH₃ [271, 335], during polymerization of aminoacetonitrile [7, 8, 189, 473], cyanamide [448, 501, 550] and dicyanamide [183, 548, 549, 551].

The sequence in which amino acid residues are bound in a polypeptide chain is a very important problem in the synthesis of polypeptides and proteinlike compounds. In modern biological synthesis of proteins, this sequence, which is characteristic of each individual protein (primary structure), is determined by the nucleic code, but, as shown, even during Fox's thermal synthesis, a certain constant order of proximity of amino acids was created in the absence of nucleic acids [153, 194]. Steinman [547] attached the first amino acid in the row to granules of a high-molecular polymer (chloromethylated polystyrol) and, in that way, obtained a resin to which one amino acid was attached. He then studied the relative effectiveness of attaching to it a number of other amino acids, with a protected amino group. After determining experimentally the yields of different dipeptides, Steinman compared these results to the rate of amino acid pairs as defined in the "atlas of protein structures" [95, 96, 523] in modern proteins, and found a significant conformity.

In *Chemical Evolution* ([76], p. 175), Calvin comments on this subject: "There are reasons to suppose that the mechanism which determines the sequence of amino acids in polypeptides includes some component for which the presence of a code system was at first not necessary." He then gives a model for a reproduction system of a polypeptide without a matrix based on control from the growing end [582]. It is interesting to compare these views with the remarkable synthesis of cyclopeptide (gramicidin S) carried out by Lipmann in the absence of a nucleic code [314, 315].

Nucleic Acids

The possibility of synthesizing nucleic acids in vitro has been shown [180, 275]. However, these syntheses may take place only in the presence of a corresponding specific enzyme, therefore cannot serve as a model for abiogenic processes on the primitive Earth. The probability of such a synthesis was first mentioned by Schramm [508] in his works with polyphosphorus ether. It was later shown that cytidine-phosphate may condense into oligonucleotides with the aid of polyphosphoric acid [513]. The capacity of carbo-

diimide to induce the formation of di- and trinucleotides from a mixture of nucleosides and nucleotides in dilute aqueous solution was shown in model tests, which more closely approximated conditions of the primary Earth [352, 567, 568, 569]. Thus, current model tests on abiogenic synthesis of organic substances under conditions which simulate primordial Earth permit (with great probability) imagining the sequential course of chemical evolution from the simplest carbon compounds to high-polymer substances from which were formed those "organized elements" or primitive organisms whose residues are discovered in deposits of the Earth's crust. At this stage of the development of matter, the transition from chemical evolution to biological evolution occurred on our planet when individual whole systems—living organisms—separated from the homogeneous solution of organic substances. A concept of this transition can be formed by comparing modern data of comparative biochemistry [138] to laboratory models.

MODEL TESTS WITH INDIVIDUAL OPEN SYSTEMS

Life on Earth is now represented by various organisms—individual complete systems with a specific complex structure and metabolism, and a definite order for mutually compatible biochemical reactions. Naturally, such complex systems could not arise suddenly in the simple, more or less homogeneous, aqueous solution of the hydrosphere, in the so-called "primordial soup." The systems must have been preceded by much simpler formations which, in the evolutionary process, gradually acquired newer and newer features of organization specific only to the living world [380]. First and foremost, they must have acquired the capacity to counteract the growth of entropy—a characteristic of all living creatures. Second, there must be a mechanism for adaptation to existence under given conditions of an external environment. In fact, all parts of the organism (its molecules, cellular organoids, and organs) must adapt to the functions they carry out in the process of living. Third and finally, there must be a special, perfected form of preserving transmission of information

based on the intramolecular organization of high-polymer compounds.

At first glance, it seems that the capacity of living organisms to counteract the growth of entropy contradicts the second law of thermodynamics [38], although this apparent contradiction is known. This was solved by Schroedinger (descriptively, although not completely accurately), who showed that the living organism "attempts" in a way "negative entropy" due to the external environment. In this way, the local reduction of entropy in any individually viewed living object is compensated for by increased entropy in the surrounding environment. Actually, this matter is much more complex within organisms. But in order to carry out even such a simplified arrangement, an organism-and-environment complex is necessary, that is the most primitive formation of phase relationships in a homogeneous solution [325, 418, 419, 471]. The emergence of multiple phases, separation from the starting uniform solution by individual systems which have separated from the surrounding medium by definite interfaces, is extremely widespread in nature and constantly encountered during laboratory studies. Without doubt, such separation on a vast scale also occurred repeatedly on the surface of a lifeless Earth.

In different sections of the hydrosphere with varied chemical composition and natural conditions, countless variegated individual systems could form, which would differ greatly from modern living creatures [72]. Numerous models of such phase-separated systems cannot only be imagined, but also reproduced in laboratory tests. For example, Goldacre [176] described the formational process for small closed vesicles which are contained in a protein-lipoid membrane on the surface of modern bodies of water. They emerged simply because of the effect of wind on protein-lipid film which had formed from products of decomposed bodies of modern organisms which had collected on the surface of the water [98]. In essence, these are simply sections of water removed from the surrounding environment by a definite interface. Even in such simple systems, conditions are created for potential independent interaction with the external environment [21, 97, 99, 413, 414].

In another example, Fox dissolved proteinoids produced by heating a mixture of amino acids in hot water. After cooling this solution, there was a formation of numerous microscopic spherules 2–7 μm in diameter, which he called microspheres. These proteinoid microspheres have an electron-microscopic structure, particularly a double membrane, with a number of biologically interesting properties [149, 153, 651]. Bahadur, the Indian scientist, observed the synthesis of polymers in sunlight. In his tests under sterile conditions, small spherical bodies which had separated from the surrounding solution were formed with an external shape, internal structure, and a number of other properties which, in his opinion, make them like living organisms. Thus, he called them "jeewanu," which translated from Sanskrit means "particles of life" [18, 19].

In working with such artificial systems which automatically separate from solution, particular attention is often paid to their external morphologic similarity to living objects. The problem of counteracting the growth of entropy in phase-separated systems is not to be solved by this, but by interaction with the surrounding medium — as open systems do, whose thermodynamics differ principally from classical thermodynamics. The thermodynamics and kinetics of open systems have been well-developed [70, 353, 416, 417, 456, 457, 458].

In closed systems (on which classical thermodynamics was principally developed), only substances included in the system are capable of chemical reaction. The constancy of the system's properties is characterized by a state of equilibrium in which the reaction rate goes in one direction equal to the rate of the reverse reaction. A thermodynamic criterion of this equilibrium is the minimum role of free energy and the maximum importance of entropy (in other words, its transition to the most probable of all possible states). Spontaneous processes in a closed system are not capable of transferring to a less probable state; that is, they can only keep the entropy of the system constant or increase it, depending on whether reversible or irreversible processes are being dealt with. When the entropy in the system increases, equilibrium is absent. On the contrary, during the onset of equilibrium, the rate

of increase of entropy is equal to zero. Conversely, in an open system, substances entering from the external medium are balanced somehow by chemical compounds returning to the external medium. Therefore, the constancy of such an open system is not characterized by thermodynamic equilibrium (as observed in closed systems), but by the onset of a steady state in which rate is achieved unilaterally by the chemical changes going on and the diffusion of substances into and out of the system.

Thermodynamic equilibrium and the steady state are similar in that in both, the system preserves its properties constant in time. A radical difference is that generally there are not changes of the free energy during equilibrium ($\alpha F = 0$); but during the steady state, this happens continuously, at a constant rate ($\alpha F = \text{const}$). Thus, the steady state is not constant because the free energy is at a minimum (as during the formation of a crystal), but because the system constantly receives free energy from the external medium in an amount which compensates for its decrease internally. The kinetics of the processes in open systems are also complex and unique. This is particularly important in the study of living objects, inasmuch as they are chemical open systems in which, for example, the introduction of a catalyst changes not only the reaction rate, but also the steady-state concentrations of components (hydrodynamic model [69], chemical models [101, 220, 417, 420]).

To solve the problem of reproducing systems which have the capacity to counteract the growth of entropy, it is important that models for this purpose pass easily from the static to a steady, continuous state characteristic of open systems [377]. The most promising (but not the only possible) models in this regard may be coacervate droplets produced at the beginning of the 1930s by Bungenberg-de-Jong which were studied later in detail [45, 46, 60, 61, 62, 63]. In initial tests, dilute aqueous solutions of gelatin were mixed with gum arabic, and observed under normal temperature and acidity. Droplets visible under an optical microscope, in which the molecules of both polymers had concentrated, separated by phase from this solution, while they were lacking entirely in the surrounding solution.

A number of subsequent investigations showed that the coacervate droplets might be formed from various substances, especially from organic polymers and lipoids.

Coacervate Systems

About 200 examples of coacervate systems formed from varied proteins, nucleic acids, polyglycosides, phosphatides, chlorophyll, and so forth, are given by Yevreinova in *Concentration of Substances and the Action of Enzymes in Coacervates* [646]. In the formation of coacervates, the degree of polymerization of the substances chosen is of great importance. They can be obtained not only from natural biochemical compounds, but also from artificial polymers, for example, polyadenine and polylysine [370, 385]. The volume of droplets (from 5 to several thousand 10^{-12} cm^3) fluctuates greatly as a function of the components and coacervation conditions selected. The concentration of polymers in the droplets exceeds their concentration in the starting solutions by many tens and hundreds of times. The study of droplets in interference, ultraviolet, or electron microscopes, reveals a certain structure and a clearly pronounced external interface (surface film) [386, 643, 644, 645, 647, 648].

Coacervate droplets can selectively adsorb and concentrate interiorly different substances from the surrounding solution (dyes, amino acids, mononucleotides, and so forth [649]). A colloidal-chemical study of cellular protoplasm shows that it and a number of its structures are found in the coacervate state lying between salts and gels [311, 312, 319, 515]. By including different catalysts (particularly enzymes) in the coacervate droplets, a number of varied reactions can be obtained (oxide reduction, polymerization, photochemical transformation, and so forth) because of substances entering from the external medium. Thus, substances of the external medium, undergoing chemical changes, either accumulate in the drops as polymers or are destroyed (freeing the energy enclosed in them), while products of their decomposition are liberated to the external medium. A continuous stream of substances is thus created through the droplets which simulate

biological metabolism in an elementary form. The droplets, interacting with the external medium according to type of open systems, take on a dynamic stability or a capacity for prolonged existence. Under certain conditions, they even increase in volume and weight, which can be directly observed under a microscope and established by chemical analysis (specifically, this was observed in the synthesis of amylose in protein-glycosylic coacervates or of polyadenine in protein-nucleic droplets) [370, 377, 382, 383, 387, 420, 422, 516, 518].

In coacervate droplets in which several inter-combined reactions take place, the more complete the union of the catalysts included in the droplets, the faster the droplets will grow. In comparing the growth curves of two differently organized kinds of drops in the same medium, the more perfect drops grow according to a rapidly rising curve, while the less perfect ones are suppressed then halted in growth [384, 597]. These tests made it possible to demonstrate the rudiments of a new law—prebiological natural selection of the predecessors of life probionts—which originated at some time on the surface of the Earth. The second of characteristic properties of life could have arisen only on the basis of this law—the “expediency” adaptability of a living system to conditions of its existence and adaptability of its parts (molecules, organoids, and organs) to the functions carried out by them in the life process.

In the scientific literature, the widespread concept of the initial formation of living systems is still encountered. There must be organized and functional nucleic acids and proteins within the aqueous solution of the Earth's hydrosphere. An automatic collection of their molecules would then have led to the formation of primary organisms similar to the assembling of a machine from previously manufactured parts [347, 348]. Such a concept echoes former mechanistic views which liken the organism to a machine. The adaptability of a machine part to the work it carries out is apt, because the part was manufactured according to a previously thought-out plan or drawing. It is in no way possible to imagine how, in the absence of such a plan, polymers could have been formed with a molecular structure

geared to carrying out specific functions in the complete living systems formed from them.

This adaptability could originate only as a result of natural selection which only complete, phase-separated, open systems could have undergone. These systems had to be capable of overcoming the increase of entropy on the basis of their interaction with the external medium. Individual molecules of the primordial soup could not have done so [351]. Model tests with coacervate droplets have shown that such individual systems may also arise from chaotically or uniformly constructed polymers [385]; these polymers may sometimes have different catalytic functions [151] or the capacity for replication. Efficient functions could be formed only in complete systems which have been subjected to selection. It is precisely from these systems (and not simply in the primordial soup), from the multiplicity of possible variants of polymer structures, that those chosen were best adapted to carrying out functions important to the preservation and more rapid growth of a system (under given conditions of the external medium). Later, they were able to reinforce the properties of these systems during the process of their proliferation and simulation [378].

Probionts

The evolutionary development of probionts is based mainly on data of comparative biochemistry. It is predominantly concerned with their catalytic apparatus as the most important factor in the organization of metabolism and is based on the relationships of the rates of its constituent reactions. At the stage of evolution under discussion, the only available catalysts for the probionts were inorganic salts and organic substances in the primordial soup and whose catalytic activity is very small. However, in mutual combination, this activity may be increased by many hundreds and thousands of times [300, 301].

A colossal number of different atomic groupings and their combinations can be supposed which, to one degree or another, were able to catalyze the reactions necessary for the existence of probionts. As a result of natural selection always

having eliminated the less perfect complexes, only very few, the most perfect—the coenzymes widely known to biochemistry—have survived. While their number is comparatively small, they are universal catalysts for all living beings, which indicates very early formation during emergence and evolution of life.

The essential constancy of concentration of coenzymes in the proliferating probionts could easily be supported by means of the simple entrance of these compounds (or the components which form them) from the external medium (such as modern vitamins, which also act as coenzymes). However, the capacity of the probionts to form these catalysts through their metabolism had to be developed later [231, 233]. Proteins—enzymes whose catalytic activity is closely related to their intramolecular organization, and above all to their primary structure, with regular location of amino acid residues in their polymer chain—only began a decisive role substantially later [102, 111, 372, 423, 538]. Even during simple thermal production of proteinlike polymers (proteinoids), their primary structure may have a specific order in the arrangement of the component amino acids. Moreover, they manifest a specific catalytic activity [113, 154, 196, 277, 290].

The possibility of such abiogenic synthesis of polypeptides with an ordered chain of amino acid fragments in the absence of a nucleic code is confirmed by Lipmann's work [315] on non-ribosomal synthesis of gramicidin and tyrocidine. Steinman [322] also found in the synthesis of peptides that specific amino acid fragments adjoin one another with the same degree of frequency as in modern proteins [96]. The possibility of fixed repetition in syntheses by replication based on control from the growing end was shown by Calvin [75], who advanced the proposition that the modern system of coding amino acids has its beginning, to some extent, in an ancient synthesis system of polypeptides in which the growing amino acid sequence determined itself. The existence of purely protein evolved systems can, then, be theorized.

The characteristic correspondence of the intramolecular structure of modern enzymes to the catalytic functions which they carry out arose

because of prebiological selection of complete systems, not of individual protein molecules. In systems of this type, numerous and varied combinations of amino acid fragments could be generated which would determine their various functions. The system gained an advantage, under conditions of the external medium, only when the internal protein structures which arose accelerated those reactions, which, in harmonic combination with other processes occurring in the system, were beneficial for the whole and promoted survival and continued proliferation of the entire system. If such were not the case, the system was destroyed by natural selection. Thus, it is understandable how at a given stage of evolution, that efficiency of organization of living systems arose which has been designated the second characteristic property of life.

The third peculiarity of life, the capacity for transmission of hereditary information, was only rudimentary because the direct replication of proteins was still very imperfect and easily disturbed. In modern organisms, enzymatic proteins synthesize by an extremely complex and perfect mechanism through which amino acids are sequentially linked to the polypeptide chain, in precisely that order. This is required for a specific, rigorously regular combination of mononucleotide fragments in DNA and RNA molecules. Such a mechanism could arise only during the process of prolonged evolution of probionts and the living systems which were then formed [474]. The first link in this evolution was the abiogenic formation of polynucleotides, a capability which was shown in a number of model syntheses [508, 568]. Of course, polymers arose in these which were much more primitive in intramolecular structure than those which now exist. They had to evolve, the same as the polypeptides. In examining this evolution, facts must be taken into consideration:

Polynucleotides have a clearly pronounced capacity for direct or complementary replication, and consequently for comparatively accurate self-reproduction of a specific intramolecular organization [628].

Polynucleotides, in contrast to polypeptides, do not have catalytic activity which would

permit them to participate directly in the organization of metabolism of probionts or of any other initial systems. They mediate this only by means of proteins.

The evolution of nucleic acids discussed by Orgel [390], deals with two problems:

The possibility of replication of polynucleotides without enzymes, and the evolution of polynucleotides without proteins.

While current model tests give a positive answer to the first question [391], the second is much more complex, which concerns the possibility of an evolution of polynucleotides oriented to the emergence of life in the absence of proteins. The "natural selection without function" proclaimed by Orgel is internally contradictory, because natural selection may only be compared to definite functions. With the capacity for quite perfect replication (although only in the presence of proteins and enzymes), nucleic acids which are found simply in aqueous solution might accumulate only in definite local places on the Earth's surface in the form of evolutionarily immobile organic deposits. In addition, the combination of proteins with catalytic functions and nucleic acids with perfect capacity for replications is extremely favorable for evolution. Union of these substances into phase-separated formations occurs (in model tests) even during simple mixing of intramolecularly poorly organized polypeptides and polynucleotides (which still do not carry genetic information). Under primitive Earth conditions, interaction of originally independent protein and nucleic systems and the subsequent evolution of the "genetic code," which later determined the preservation and ideal transmission of hereditary information, must have occurred also in such formations.

In generalizing much material in the literature,

Crick [91] proposes two possible pathways for the origin of modern nucleic code:

1. This code was derived simply on the basis of purely stereochemical relationships between triplets of bases (by codes) and amino acids. (In this case, its current universality has been determined by purely chemical structure and in principle it cannot be otherwise.)
2. Originally, the code was more primitive and became perfected during the process of its evolution on the basis of natural selection, i.e., by the attraction of biological regularity. It is now universal not because no other code could exist for purely chemical reasons, but "because at the present time *any change in it would be fatal or strongly counterselective.*"

A number of factors and concepts favor the second solution of the problem, and Crick provides a possible picture for subsequent evolution of the code which could have taken place only in complete systems formed from primitive polypeptides and polynucleotides subjected to natural selection, but which were capable of perfecting their organization on the basis of prebiological selection of the systems, not of the individual molecules. Thus, at a comparatively late stage in the evolution of the probionts, these systems reached a new, previously unachieved height of precise self-reproduction which is characteristic of all modern living beings [165, 638]. Further improvement was on this basis, with regard to both metabolic organization, and intracellular structure. This matter has been discussed on the basis of comparative biochemical and cytological data [75, 96, 100, 139, 161, 168, 169, 261, 278, 279, 281, 321, 322, 368, 371, 412, 435, 502, 523, 546, 629].

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