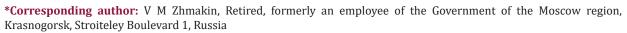


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Initial Components and Abiogenic Homochiral Organic Synthesis Ere the Origin of Life

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ABSTRACT

The nature of the appearance of carbon, initial components, molecules of organic and homochiral abiogenic synthesis on the early Earth and in carbonaceous chondrites has not been established. With the duration of preservation of such molecules in carbonaceous chondrites for more than 4.5 billion years, the reasons for their preservation from decay, racemization and hydration have also not been clarified. In the oxygen-free atmospheres of the nebula and the early Earth, hydrogen and hydrogencontaining gases were oxidized with carbon monoxide and carbon dioxide to form carbon and water, as well as intermediate products of these reactions, formaldehyde and methane acid. Together with ammonia, they were the initial components of organic synthesis. According to the Rebinder rule, carbon adsorbs hydrogen well, including in organic molecules. In experiments with the assumed conditions of the early Earth. R-(rectus, Latin) ribose from formaldehyde and S-(sinister) serine from formaldehyde, methane acid and ammonia were obtained by adsorption on carbon. For other S-amino acids, a stereo chemical justification of their formation based on S-serine is given. For carbonaceous chondrites, the results of the above experiments were confirmed by the correlation of an increase in homochiral excess with an increase in the amount of hydrogen in aldonic acids and lactic acid with a correlation coefficient of 0.94 and 0.85 in amino acids. The justification of the homochiral process will reduce the costs of searching for life on planets, for biochemical research, for the production of medicines, perfumes, food, and so on.

Introduction

The problem of preservation of organic substances during the fall of meteorites was resolved after the detection of a thin glassy heat-protective crust, and the reasons for the appearance of carbon, initial components, homochiral molecules and their preservation from racemization and decay for more than 4.5 Ga have not been established. In the absence of oxygen, H_2 and gases with H can be oxidized by CO_2 and CO to form H2O and carbon (reactions 1 - 4, 7, Table 1) or intermediates $H_2\mathrm{CO}$ (methanal) and HCOOH (methanoic acid) (reactions 5, 6, 8, 9, Table 1), which with NH_3 were the initial components of abiogenic organic synthesis [1]. In experiments, formaldehyde was a universal starting component in the synthesis

of amino acids and sugars. The formation of amino acids racemates has been established with a content from 10^{-4} to 5×10^{-3} mol in a solution with 2.5% H2CO (formaldehyde or methanal) and 1.0 - 1.5% NH₄NO₃ or NH₄C1 at a temperature of 313 - 318 K [2]. For the first time, sugars from H₂CO were obtained in 1861 by A.M. Butlerov. Ribose from H₂CO was synthesized with high yield in the presence of apatite as a catalyst in an Ar atmosphere at pH-7.3 and a temperature of 313 K and [3]. Previously, the possibility of the formation of fatty acids and nitrogenous bases was shown with the disproportionation of H2CO [1]. H₂CO is installed in the atmosphere of Mars [4] in the interstellar medium and comets [5] in carbonaceous chondrites [6] and in volcanic gases.

According to the Rebinder rule, carbon adsorbs hydrogen well, and inorganic molecules its adsorption properties [7] are similar to hydrogen. This made it possible to obtain R-ribose and S- serine (reaction 10, Table 1) in experiments conducted for the first time by adsorption on carbon with stereo chemical justification [1,8]. Other S-amino acids could formed on the basis of S-serine during the disproportionation of $H_2C(OH)_2$ (metanediol, H_2CO in water) molecules, as shown by the example of the formation of alanine (reaction 11, Table 1) and phenylalanine (reaction 12,

Table 1). Homochirality in other optical isomers (for example, in glucose) could arise during evolution. Adsorption on carbon provided for organic synthesis: the concentration of the initial components, the formation of only R-ribose and S-amino acids, hydrophobic-hydrophilic properties, protection from hydration, decomposition and racemization [1,8]. The main purpose of this work is to substantiate the abiogenic synthesis and preservation by adsorption on carbon of homochiral enantiomers in carbonaceous chondrites and on the early Earth.

Table 1: The most likely reactions the formation organic molecules and carbon.

| Nº | Reactions | lgKT* at T, K | |
|----|---|----------------|--|
| | | 298 323 348 | |
| 1 | COg+H ₂ Og=CO ₂ g+H2g | 5.1 4.6 4.1 | |
| 2 | CO ₂ g+2H2g =2H ₂ Oliq | 14.0 11.6 8.9 | |
| 3 | CO ₂ g+CH4g=2H ₂ Oliq+2Cgr | 5.1 3.7 2.5 | |
| 4 | 3/2CO ₂ g+2NH3g=3H ₂ Oliq+3/2Cgr+N ₂ g | 15.1 12.7 10.7 | |
| 5 | 3/2CO ₂ g+2NH ₃ g=H ₂ COg+2H ₂ Oliq+1/2Cgr+N ₂ g | 7.1 7.0 6.9 | |
| 6 | 2CO ₂ g+2NH ₃ g=HC00Hliq+2H ₂ Oliq+Cgr+N2g | 2.7 1.1 -0.3 | |
| 7 | CO2s+2H2Ss=2H ₂ Oliq+Cgr+2Scr | 5.7 4.6 3.7 | |
| 8 | HCO ₃ ⁻ +2HS ⁻ +H ⁺ =H ₂ COs+2H2Oliq+2Scr | 7.1 7.2 7.3 | |
| 9 | 2HCO ₃ ⁻ +HS ⁻ +2H ⁺ =HCOO ⁻ +CO ₂ s+2H ₂ Oliq+Scr | 8.7 8.6 8.5 | |
| 10 | CH ₂ (OH)CH(OH)2a+HCOONH ₄ =+NH ₃ CHCH ₂ OHOOa+2H ₂ Oliq | | |
| 11 | $- CH_2OH + H2C(OH)_2 + HCO_3^- = - CH_3 + HCOO^- + H_2Oliq + H_2CO_3$ | рН 6.3-7.5 | |
| 12 | $- \text{CH}_2\text{OH} + 12\text{H}_2\text{C(OH)}_2 + 12\text{HCO}_3^- = - \text{CH}_2\text{C}_6\text{H}_5 + 6\text{HCOO}^- + 13\text{H}_2\text{Oliq} + 12\text{H}_2\text{CO}_3$ | рН 6.3-7.5 | |

Note: g-gas, s-solution, gr-graphite, liq-liquid, cr-crystal a-adsorbed. *Approximate values. — chemical bond. — CH₂OH serine radical. (Thermodynamic potentials [25–27]

Methods

Initial Components and Homochiral Molecules in Carbonaceous Chondrites

The conditions in the nebula make it possible to study planetary regularities and the composition of carbonaceous chondrites. The differentiation of gases in the nebula caused the maxima of their relative content (mass of atmospheric gas/mass of the planet) for Venus (v) - CO₂, Earth (e) - H₂O (taking into account exospheres) and Jupiter (i) - H2 [9], which is confirmed mathematically by the ratios of the molecular masses of gases (M) and the distance to $Sun(R)(MCO_2/MH_2O)/(R_eR_v) = (MH_2O/(R_i/R_e) = 1.745 \pm 0.015)$ Carbonaceous chondrites are divided into several types depending on the composition and temperature of formation from ${\sim}1150$ -1200 K to liquid water [10,11]. At the same time, the classifications of carbonaceous chondrites take into account the increase in carbon content with the growth of water and organic substances [10,11], which may reflect the relationship of their formation during the reduction of CO₂ (reactions 1-7, Table 1). These chondrites contain carbonates [10,11] which, together with the atmospheres of the

terrestrial planets, indicate the presence of CO_2 in the nebula. The average contents of carbon and water in the main types of carbon chondrites [11] has an almost direct linear relationship. Such dependence could be realized in the nebula in the reactions of H2 and gases with H with C0 and CO_2 with their activation by optimal temperatures and solar radiation [12]. Carbon, $\mathrm{H}_2\mathrm{O}_2$, or intermediates of $\mathrm{H}_2\mathrm{CO}_2$ and HCOOH were the products of these reactions (reactions 1- 6, Table 1), the yield of which increased at temperatures below 970 - 940 K. Like reactions could occur in water (reactions 7 - 9, Table 1).

 $\rm H_2CO$ and HCOOH together with $\rm NH_3$ were the initial components for organic synthesis during adsorption on carbon [1] and detected in carbonaceous chondrites. $\rm H_2CO$ was found in 12 carbonaceous chondrites studied from 0.1 to 18 mg g $^{-1}$ [6,13], and HCOOH in chondrite of Lake Tagish from 209 to 378 mg g $^{-1}$ [13] and in the composition of monocarboxylic acids in other chondrites [5]. Ammonia was identified in 6 carbonaceous chondrites from 0.5 to 10 mg g $^{-1}$ [14] and in the composition of amino acids. Organophosphorus compounds with H3PO4 have not yet been detected. These initial components were in the nebula at

pressures from $\lg P = -3$ to -5 bar [10] in gaseous state or in solution in carbonaceous chondrites with condensed water. Carbonaceous chondrites contain pentoses, sugar alcohols, acids, and more than 100 types of amino acids, as well as other organic compounds. Among them, a homochiral excess is often found. For example, amino acids with S- excess are found in 24 out of 24 studied carbonaceous chondrites, and in one of them R-alanine predominates by 12% [5]. 33 amino acids were found in Lake Tagish chondrite, but homochiral S-excess was studied and revealed in 10 types, and its excess is in alanine (%) 22, in isovaline - 13 and in valine - 97.7 [13]. According to the review [15], out of 101 identified amino acids in carbonaceous chondrites S-excess was found in 23 from 1 to 99%

of 28 studied and 5 are represented by racemates, 13 were achiral, homochirality not studied in 60. In the composition of 101 amino acids, 75 types are characteristic only for meteorites. Homochiral R- excess found in 2 aldonic acids with 3 CH (or $\rm CH_2$) radicals from 33 to 55%, in 4 with 4 radicals from 47 to 100%, and in 6 with 5 radicals of 100% each. S-excess of 3 to 12% is noted in lactic acid. S-excess of 7% and an R-excess of 3% identified in the alanine of one meteorite. R- excess is present in allo-isotreonine up to 8% and from 12 to 60% in allo-isoleucine. The listed compounds with a biologically inverse homochiral excess contain an asymmetric CH3 radical relative to hydrogen at the central carbon atom (H1, Figure 1a) and two radicals in allo- isoleucine.

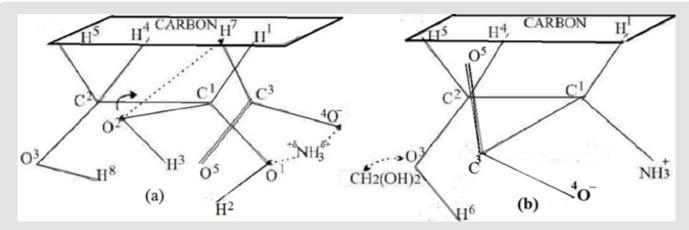


Figure 1: Block diagram of the formation of

- (a) S-serine and
- (b) S-alanine.

Dotted lines - the interaction of atoms at the time of the reaction, the arrow-the rotation of the group C1.

On the Cosmic Nature of Enantiomers with Homochiral Excess in Carbonaceous Chondrites

Organic substances are known in the interstellar medium and comets [5], so their extraterrestrial nature in carbonaceous chondrites, and hence homochiral molecules, is beyond doubt. Doubts about the nature of enantiomers with homochiral excess in carbonaceous chondrites arise most often due to the lack of clarity of the processes of their appearance, which is justified below. In addition, the extraterrestrial nature of these enantiomers is confirmed by the isotopic ratios of C, N₂ and H₂ [5,13-15]. The same nature is indicated by the complete absence of high-molecular organic compounds in carbonaceous chondrites (peptides, proteins, organophosphorus compounds, RNA, etc.). The ice of Antarctica in meteorite impact sites and beyond contains organic matter in trillionths, which is most likely due to meteorite dust [16], and the content of various amino acids in carbonaceous chondrites is millions of times higher than in ice. Carbonaceous chondrites contain about 15 sugar acids and about 75 amino acids not found

in the biosphere [10], as well as bio-inverse enantiomers. One meteorite may contain racemates, molecules with bio- and reverse excess, which is not typical of the biosphere.

Initial Components of Homochiral Synthesis and Conditions on Earth at the Origin of Life

Planetary regularities show that the Earth's atmosphere in the early Archean did not qualitatively differ from the atmospheres of Venus and Mars and consisted of $\mathrm{CO_2}$, $\mathrm{N_2}$ and other gases in descending order, as on Venus: $\mathrm{SO_2}$, CO , $\mathrm{Cl_2}$, HCl , $\mathrm{H_2S}$, $\mathrm{CH_4}$, $\mathrm{H_2}$, $\mathrm{NH_3}$ [17]. Martian atmosphere contains $\mathrm{H_2}$, $\mathrm{CH_4}$, $\mathrm{NH_3}$, and $\mathrm{H_2CO}$ [4]. The volcanic gases of the Earth have a similar composition. Of 108 samples for 6 volcanoes [18], the average content of gases without $\mathrm{H_2O}$ is (%): $\mathrm{CO_2}$ - 58.7, $\mathrm{SO_2}$ - 20.1, $\mathrm{H_2}$ - 7.7, HCl - 6.7, CO - 5.1, HF - 0.58, $\mathrm{H_2S}$ - 0.41, $\mathrm{NH_3}$ - 0.16, $\mathrm{CH_4}$ - 0.016, sometimes OCS, $\mathrm{CS_2}$, $\mathrm{H_2CO}$ and others. Carbon, carbohydrates, and amino acids were found in the products of volcanic eruptions with an abiogenic nature according to isotopic data [18]. Volcanoes during the degassing of

terrestrial planets determined the composition of the atmospheres in the Archean. The composition of volcanic gases is similar to the composition of comets [5], gases in carbonaceous chondrites and their minerals [10,11]. The composition of volcanic gases is similar to that of comets [5], gases in carbonaceous chondrites and their minerals [10,11]. Organic synthesis could have originated on Earth with the appearance of sedimentary rocks and water basins 3.7 - 3.8 billion years ago. As carbonaceous chondrites, typical sedimentary rocks of this period contain abiogenic carbon and carbonates [19], and homochiral enantiomers on Earth should have appeared before life, so the initial components and processes of homochiral abiogenic synthesis of the Earth and the nebula should have been identical.

The CO₂ pressure in the atmosphere was about 50 – 60 kPa, and the temperature was close to 310 - 320 K [20,21]. The height of the Archean troposphere could be about 6.5 km, which is 1.7 times less than its current average height of 11 km (from the comparison of the modern homogeneous atmosphere with the atmosphere from CO_2 at pressure of 55 kPa by the formula H=P0/ ρ x g, where P0 and ρ are the pressure and density at sea level, g - is the acceleration of gravity). These factors provided sharp daily temperature changes up to 25 - 35 K (as in tropical deserts) with change in the solubility of gases and pH (hydrogen index) in small pools. From the above, we can draw the main conclusions about the causes of biosynthesis on Earth. During the accretion of planets, the distance of the Earth from the Sun determined the maximum concentration of water on it and, together with the content of greenhouse gases in the atmosphere, the optimal temperature for its existence in liquid form. The appearance of H₂CO and HCOOH was facilitated by the mass of the planet, which prevented dispersion of H₂ into space, as well as the predominance of CO₂ pressure in the atmosphere over the sum of pressures of H₂ and gases with H (but at P₁₁₂, more than

the minimum for reaction with CO_2 , at $PCO_2 = 0.55$ bar, $P_{H2} > 10-6.8$ from reaction 2 of Table 1).

Results

Synthesis and Preservation of Homochiral Enantiomers by Adsorption on Carbon

The age of chondrites varies from 4.5 to 4.575 billion years [10]. The time of racemization of asparagine at 252 K is estimated at 20 million years, and in the shell of mollusks with an age of one million years, the ratio of R to S amino acids was: for asparagine - 0.88, alanine - 0.95, valine - 0.85 [10]. Half of the racemization cycle varies from 500 to 180,000 years for 11 amino acids from the composition of proteins [22]. Long-term preservation of homochirality in enantiomers from decay and racemization could be achieved by fixing them on a solid material and at low temperatures. The most accessible method of fixation is adsorption, and the most effective adsorbent in carbonaceous chondrites is carbon. An increase in the number of hydrogen atoms in 3 alkanes and 5 alcohols was accompanied by an increase in their adsorption from solutions (experiments with alkanes were performed 4 times on 4 brands of coal, and with alcohols 5 times on 5 brands of coal) [7]. Therefore, in the homochiral synthesis of molecules by adsorption on carbon, there must be a correlation between the number of hydrogen atoms and the homochiral excess. Significant correlation coefficients of 0.94 (with a significance level of 0.001) were established between the number of hydrogen atoms in aldonic acids and lactic acid and the average values of homochiral excess and 0.85 (with 0.05) for amino acids (Table 2). The correlation coefficients show that the homochiral synthesis of enantiomers is directly related with adsorption on carbon. Correlation is also noted for homochiral excesses and molecular weights of aldonic acids and lactic acid due to their proportionality to the amount of hydrogen, but in its absence there is in amino acids no correlation.

Table 2: Initial data for calculating the correlation coefficients and the regression equations.

| Nº | Enantiomers | Quantity H atoms | Average homochiral | Number determinations | References |
|----|--------------------------|------------------|--------------------|-----------------------|------------|
| | Lactic and aldonic acids | | % R-excess | | |
| 1 | Lactic acid | 3 | - 6 (S-excess) | 4* | [15]** |
| 2 | Erythronic acid | 4 | 54.0 | 1 | -//- |
| 3 | Threonic acid | 4 | 43.7 | 4 | -//- |
| 4 | Ribonic acid | 5 | 57.0 | 1 | -//- |
| 5 | Arabinonic acid | 5 | 53.5 | 2 | -//- |
| 6 | Xylonic acid | 5 | 91.0 | 2 | -//- |
| 7 | Lyxonic acid | 5 | 80.5 | 2 | -//- |
| 8 | Allonic acid | 6 | 100.0 | 1 | -//- |
| 9 | Altronic acid | 6 | 100.0 | 1 | -//- |
| 10 | Gluconic acid | 6 | 100.0 | 2 | -//- |
| 11 | Idonic acid | 6 | 100.0 | 1 | -//- |
| 12 | Galactonic acid | 6 | 100.0 | 2 | -//- |

| 13 | Talonic acid | 6 | 100.0 | 2 | -//- |
|----|--------------|----|------------|----|-----------|
| | Amino acids | | % S-excess | | |
| 1 | Serine | 5 | 46.4 | 8 | [5,15,13] |
| 2 | Aspartic | 5 | 57.5 | 14 | [5,15,13] |
| 3 | Alanine | 6 | 35.5 | 11 | [5,15,13] |
| 4 | Threonine | 7 | 68.0 | 3 | [15] |
| 5 | Glutamic | 7 | 67.4 | 16 | [5,15] |
| 6 | Proline | 8 | 65.0 | 2 | [15] |
| 7 | Valine | 10 | 96.0 | 2 | [13] |

Note: * Statistical samples: 24 lactic and aldonic acids and 53 amino acids. ** The accuracy was not determined, and the values were excluded if it was close to the racemates.

Enantiomers with homochiral excess could not appear and persist in the nebula from radiation and collisions without carbon protection. The absence of complex organic molecules (sugars, amino acids, etc.) in the interstellar medium and comets [5] is a confirmation of this. The increase in homochiral excesses and the number of hydrogen atoms in the molecules indicates the fixation of hydrogen on carbon, what ensured their preservation for more than 4.5 Ga in carbonaceous chondrites at low temperatures. The vast majority of meteorites (99.8%) originate from the asteroid belt, where the temperature varies depending on the distance of the orbit to the Sun from 165 to 190 K. It was even lower at the early luminosity of the Sun. At these temperatures, racemization of enantiomers could occur in molecules with weak adsorption adhesion after desorption, which depends on the number of adsorbed hydrogen atoms. Desorption could occur due to the penetration of high-energy cosmic rays and internal radioactive decay, or by heating in the frontal part and a strong impact during a fall. As shown in subsection 2.3, the conditions on Earth in the early Archean and in the Nebula during the formation of carbonaceous chondrites were similar, so the processes of synthesis of homochiral enantiomers were the same, but the time of their preservation on Earth was less.

Discussions

Stereo Chemical Substantiation of Homochiral Synthesis by Adsorption on Carbon

There are several ways to obtain amino acids, but their synthesis by adsorption on carbon has not been studied. Common to this synthesis are condensation reactions. Glycolic aldehyde hydrate was formed from two $\mathrm{CH_2(OH)_2}$ molecules in the reaction with the release of $\mathrm{H_2O}$. Glycine could be synthesized by adsorption on carbon in a condensation reaction from the $\mathrm{CH_2(OH)_2}$ molecule and ammonium formate (or from HCOO- and then NH4 $^+$, since ammonium could not react with glycolic aldehyde hydrate. Next-ammonium formate). S-serine appeared in the presence of glycolic aldehyde hydrate and ammonium formate (reaction 10, Figure 1a).

The structural features of glycolic aldehyde hydrate and ammonium formate determined the synthesis of S-serine at adsorption on carbon. Their convergence was due to the dipole-dipole interaction of O^5 with two protons H^2 and H^3 (Figure 1a). Hydrogen H^7 was adsorbed by carbon in situ with minimal influence of other hydrogen atoms. The valence angle of $H^4C^2H^5$ in glycolic aldehyde is 106.18 degrees according to experimental data, and the coordinates of the atoms were determined experimentally and by calculation [23]. The angle between H^1C^1 and H^4C^2 , calculated from the coordinates in the projection on the XOZ plane, is 46.0 degrees. Consequently, the C1H1 bond is shifted from the bisector of the angle $H^4C^2H^5$ to H^4 by 7.1 degrees. Hydration of the carbonyl group of glycolic aldehyde resulted in a clockwise rotation of C^1H^1 from the bisector of the angle C^2H^2 with a general deviation of C^1H^1 from the bisector of the angle C^2H^2 by 17.6 degrees in the hydrate of glycolic aldehyde.

The valence angle of H7C3O5 in the HCOOH molecule is about 118 degrees, almost the same as that of glycolic aldehyde hydrate $H^1C^1O^2$. The interaction of O^5 and O^2 during the approach caused structural stress, so the C1 group in the glycolic aldehyde hydrate could rotate clockwise by about 60 degrees to neutralize this stress with the formation of the C1C3 bond. Rotation of the group at C2 was prevented by stable fixation of two hydrogen atoms by adsorption on carbon. The amino group was formed after the conversion of ammonium into ammonia with an increase in pH. This is how S-serine was synthesized. The formation of other S-amino acids (Figure 1b) occurred during the disproportionation of CH₂(OH), molecules in reactions with hydroxyl O³H⁶ in serine. R-ribose was synthesized sequentially from five CH₂(OH)₂. The asymmetric position of newly formed CH3 radicals in the structure of enantiomers relative to H1, as in alanine (Figure 1b), could be accompanied by its inversion during adsorption with fixation of the reverse homochiral excess on carbon. Otherwise, only equilibrium racemates would be preserved. This process probably occurred as during racemization, but with the preservation of the new structure by adsorption on carbon.

Conclusion

The processes of abiogenic homochiral synthesis on the early Earth and in carbonaceous chondrites were carried out by adsorption on carbon and were identical but differed in a small amount of water and the absence of organophosphorus compounds in carbonaceous chondrites. The adsorption nature of abiogenic homochiral synthesis on carbon in carbonaceous chondrites is confirmed by significant correlation coefficients between the amount of hydrogen and homochiral excess, their preservation from decay and racemization for more than 4.5 Ga, the formation of only R-ribose and S-amino acids with a rare bio-inversion confirming this nature. Doubts about the abiogenic origin of life on Earth are most often associated with the unique phenomenon of the presence of homochiral molecules only in living organisms. This work will significantly reduce such mysticism.

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Author Declarations

Statement of Interest

There are no interests to declare.

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Authors' Contributions

The author V. M. Zhmakin is the only author of the manuscript who has carried out all kinds of work for its creation, including information collection, analysis, mathematical calculations, generalization, writing and editing of the text.

Competing Interests

Author has declared that no competing interests exist.

Ethics Declaration Statement

Not applicable. Living organisms were not involved for experiments.

Consent to Participate

Not applicable.

Consent for Publication

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