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On the Origin of Biological Functions

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Abstract

We consider the problem of structure and functions of the first forms of living matter and present a hypothesis that they were formed through a physico-chemical process known as dendritic crystallization. According to this hypothesis the branching, dendritic structures helped build living systems by lending them functions so that organic chemical evolution is just one natural consequence of the evolution of matter in the universe. We conclude that a self-replicating biological system with adaptation emerged from simple molecules using completely abiotic mechanism of formation, which acted simultaneously or intermittently at different places on the early Earth and created similar structures everywhere. The dendritic hypothesis of origin of the functions explains similarities in the living systems and supports the assumption of a 'second genesis of life'. The dendritic scenario does not need carbon/phosphorus-based solutes in water-based solutions, which may have important implications for exobiology and extraterrestrial origin-of-life scenarios. An experiment to test the hypothesis is suggested.

Keywords: Origin of Biological Functions, Dendritic Growth, Prebiotic Chemistry, Protobiont

1. Introduction

Classification of tenable origin of life theories may be based on different principles. Davies and McKay [1] divided them into the categories of Extraterrestrial and Terrestrial origins. Bada [2] classified the Terrestrial theories into two categories "The prebiotic soup theory" and "The metabolist theory" and tried to build a consensus by incorporating them into a general scheme of "the transition from abiotic organic compounds to autonomous self-replicating molecules capable of evolving by natural selection into ones of increasing efficiency and complexity …" The unified theory culminated in the RNA World. Below I will review only works which are essential for the present discussion and did not find a way into the aforementioned reviews.

Prigogine and Nicolis [3] analyzed the problem of presence of spatial order and functions in biological structures and pointed to chemical evolution of matter in the universe as a necessary prerequisite of life. They concluded that "spatial dissipative structures", attained under nonequilibrium conditions in open systems, "have contributed in an essential way to the first biogenetic steps" and that emergence of biological order may be seen as the "consequence of far from equilibrium thermodynamics applied to certain types of non-linear systems". Prigogine [4] noted that the "dynamical instabilities … are … at the root of complexity that is essential for self-organization and the emergence of life". Kauffman [5] introduced an origin-of-life hypothesis, which assumes that the order of the first living systems was the result of spontaneous self-organization, rather than of a selection process. He also proposed that "Life is an expected, collectively self-organized property of catalytic polymers" and suggested that it appeared as "a phase transition leading to connected sequences of biochemical transformations by which polymers and simpler building blocks mutually catalyze their collective reproduction". Morowitz and Smith [6] introduced a hypothesis of "the collapse to life" under the geological stress, which explains stability of the core biochemistry by using the concept of the phase transition between biotic and abiotic states.

Almost all workers writing about the origin of life had at least some model of compartmentalization to overcome the concentration gap problem, but the problem of division persisted. Oparin [7] proposed a model of a protocell based on the properties of a coacervate, a droplet composed of mixtures of colloidal particles formed by the process of phase separation. He identified the mechanisms of fragmentation and competition as neces-

sary for the protocell formation and growth. Morowitz *et al.* [8,9] discussed "… the chemical logic of a minimum protocell … as an entity thermodynamically separated from the environment and able to replicate using available nutrient molecules and energy sources". They found [8, p. 100] "a very real similarity between crystallization and some aspects of self-replication". The authors came to the conclusion that the minimum protocell was a vesicle of amphiphiles and chromophores. To explain division of protocells they used Rashevsky's [10] idea that "at some point, the size of the membrane vesicle forming the protocell increases to the point that stabilizing forces are no longer able to maintain integrity, and the vesicle breaks down into two or more smaller vesicles". Unfortunately, the authors had never presented the driving force for such division, which is not trivial because the surface energy of the vesicle drives the small ones to coalesce into a large one that is, backwards. Recently significant progress was made in the area of spontaneous growth and replication of fatty acid micelles and vesicles with simple lipid membranes [11]. Hanczyc *et al.* [12] showed that clay accelerates spontaneous formation of vesicles of lipids. However, to induce vesicle division the authors had to invoke the process of extrusion—forceful drag of the material through a small-pore filter. They admit that the "use of membrane extrusion to mediate division is artificial, and the possibility of a natural analog of this process seems remote." To inspire growth and division of micelles and vesicles Rasmussen *et al.* [13] used energy of light and Stano and Luisi [14]—the surfactants. Although these results are impressive, it should be realized that even simple fatty acids are complicated materials for prebiotic conditions.

Based on an observation that natural minerals have many of the properties of living organisms, e.g. crystals can grow and store information in the form of crystal defects, Cairns-Smith [15-17] put forth "the clay hypothesis" of the mineral origins of life and the subsequent "genetic takeover". He introduced the concept of a genograph "as a kind of 'picture' of imperfections in a crystal … that held … the primitive genetic information" instead of a molecule. According to his hypothesis "A mineral genetic material might hold information in the form of a particular complex stacking sequences of layers and replicate it through an appropriate alteration of growth and cleavage" [18]. Later on minerals (e.g. clay or barium ferrite, Turner *et al.*, [19]) become templates for more complicated materials which "gradually 'takeover' the control machinery" in the process of genetic metamorphosis. In a "genetic staircase" scenario the "multiple overlapping genetic takeovers" led to appearance of sophisticated biological materials capable of their own survival and propagation.

Chirality, as manifested by the preponderance of L-amino acids and D-sugars in living matter, is another property of life, which, together with the cellular organization, should be addressed by the theories of life origin. Kondepudi *et al.* [20,21] and Buhse *et al.* [22] demonstrated the mechanism of spontaneous chiral symmetry breaking (SCSB), where cooling and stirring of highly concentrated aqueous solution of achiral molecules, e.g. sodium chlorate, yields chiral crystals. This mechanism involves random formation of a single crystal of arbitrary chirality, from which 'secondary crystals' of the same chirality were broken off by the external achiral process of stirring and convection. This mechanism works only in strongly supersaturated solutions (far-from-equilibrium). Viedma [23] added glass beads as the reinforcement of stirring and was able to induce SCSB in slightly supersaturated solutions (near-to-equilibrium). The obvious problem of SCSB for the origin of life is that biochirality is based on chiral molecules, not on chiral crystals of achiral molecules. Hazen *et al.* [24-26] considered the mechanism of chiral selectivity of mineral surfaces, according to which equally represented chiral mineral surfaces selectively adsorb chiral biomolecules, e.g. amino acids or nucleotides, from racemic prebiotic soup. SCSB mechanism may be used to describe the appearance of the chirally adsorbing mineral environments out of achiral geomaterial with the help of an external process, e.g. convection in molten Earth, Earth's magnetism, or the Coriolis Effect. Yet, to achieve the biochemical homochirality these mechanisms need a frozen accident scenario.

Shinitsky *et al.* [27] observed "unexpected difference in the solubilities of D- and L-tyrosine in water, which could be discerned by their rate of crystallization and the resulting concentrations of their saturated solutions". This effect is neither due to the difference in D- and L-equilibrium crystal structures, enantiomeric impurity, surface of the vessel, nor due to secondary nucleation. The authors conjectured that high cooperativity of crystallization enhances minute difference of energies of the enantiomers caused by the parity violation of weak nuclear forces. Based on this conjecture they suggested the mechanism of the origin of biochirality: one enantiomer was selectively removed from the racemic prebiotic soup leaving behind a concentrated solution of the other enantiomer; then biopolymerization took place in the leftover solution, not in the crystal of the first enantiomer. Kojo *et al.*, [28,29] attempting to answer the question "Why and how L-amino acids were selected in biosphere?" found that "racemic D, L-asparagine induces asymmetric resolution of co-existing racemic amino acids during recrystallization". Their data also show that crystalliza-

tion of racemic D, L-Asn yielded preferential formation of L-crystals over D-crystals, the fact that was left unexplained. In essence, this is another manifestation of "unexpected difference between D- and L-" enantiomers discussed by Shinitsky *et al.* [27].

The fact that complex biochemical features are shared by all forms of extant life made the origin-of-life scientists assume that all organisms originated from the same entity (single cell or a macromolecule), called Last Universal Common Ancestor (LUCA). However, recent observations and speculations forced many researchers to reexamine this paradigm. For instance, careful analysis of the geological records [30] shows that environmental conditions conducive to the origin of life were intermittent on early earth [31]. Wolfe-Simon *et al.* [32] found that a bacterial strain can replace phosphorus in its key macromolecules, including DNA, with arsenic. These and other similar observations led researchers to an assumption that "life may have arisen more than once" [33]. The hypothesis of the 'second genesis of life' becomes even more important in the context of organic material swap between the planets in the solar system [34].

2. Motivation

Almost all hypotheses of the origin of life on Earth describe the transformation from geochemistry to biochemistry, which brought about the material of life, a DNA-RNA-protein combination, and cellular organization of that material. Living organisms, however, are distinguished from a mixture of organic molecules by their high level of complexity, which allows them to carry out certain *functions*. Shapiro [35] pointed to a "missing fragment in our picture of the origin of life … a principle that governs the gradual evolution of simple chemical systems into more sophisticated ones capable of replication and Darwinian natural selection". The transition from a disorganized biomass to an organized system capable of reproducing itself and adapting to changing conditions represents the most puzzling problem in the study of the origin of life. The question of 'Why did life adopt these particular functions?' is left off the discussion in these theories. They imply that the right material will automatically take care of the functions problem as soon as it appears, e.g. RNA molecules 'know' how to reproduce; a metabolist 'knows' how to metabolize, etc. In fact, the question of the origin of the functions may be separated from the question of the origin of the material; the biomolecules may even vary in the make-up (e.g. Wolfe-Simon *et al.* [32]), but not in the functionality. The question of the origin of the functions deserves special attention and is the prime focus of the present publication. In the Bada model [2], chance plays a large role as the appearance of the first self-replicating molecules, their functions, and some of their properties, e.g. chirality, are assumed to come about by accident. If the problem of origin of functions is not addressed, an impression will remain that the extant form of life happened completely by accident, because other biomaterials are also possible. The problem of the origin of functions gains additional significance in the context of exobiology because we may soon be dealing with new forms of organic materials where silicon replaces carbon, arsenic—phosphorus, hydrogen-sulfide—water.

As pointed out above, in this article I am not concerned with the question: which biopolymer came first—protein or nucleic acid. Rather I am concerned with the problem of the origin of functions of the living organisms. Although the definition of functions of life is not a trivial subject [36-39,94] most of the researchers in the field agree that all biological (living) systems are characterized by the following basic functions: *growth and metabolism*, *division and replication*, *mutation and evolution* [40]. Notice that not all apparent functions of life are included into the list of the basic biological functions; for instance, motility is not one of those.

3. Hypothesis

Many observations and speculations have led me to conclude that the life functions have their roots in the physical process of crystallization as opposed to a chemical reaction. When crystallization in nonliving systems is taking place far from equilibrium, it results in the formation of branching, dendritic, patterns which are also ubiquitous and omnipresent in the biological systems.

Protobiont is a term that represents the first forms of living matter [41,42]. Protobiont is a self-replicating structure that carried some genetic information and could multiply inside the complex primordial environments e.g., slimy layers of molecules that had accumulated on the rocks. According to the principle of continuity postulated by Morowitz *et al.* [8,9] the protobionts had to have some of the biological functions, although enzymes and macromolecules had not yet arisen. In many different ways, protobionts are equivalent to Oparin's coacervates, Fox's proteinoids [43], Orgel's citroens [40], Wachtershauser's surface metabolists [44], Morowitz's vesicles of amphiphiles, Martin-Russell's protocells [45], Dawkins' replicators [46], or Woese-Fox's progenotes [47]. However, in this paper I prefer to use the term protobiont.

I hypothesize that protobionts were formed through the process of dendritic crystallization. The rest of this article is an attempt to substantiate the hypothesis and find useful applications of the latter. In this article I am not attempting to pinpoint the material that underwent the primordial crystallization, although a few candidates

will be suggested. I am trying to analyze the functional relationships and show that dendritic structures possess all characteristic functions of the living systems mentioned above.

Dendrites are branched microstructures of crystal growth; they bring to mind pictures of snowflakes, see **Figure 1**. Dendritic structure formation is an intrinsically nonequilibrium thermodynamic transformation in an open or closed system that occurs during crystallization of many pure substances, including biologically important ones, and their aqueous solutions [48-50, 51, p. 206]. Yet, not all crystals grow dendritically: dendrites appear during crystallization of substances with low entropy of fusion, $\Delta S_f < 2k_B$, where k_B is the Boltzmann's constant [52]. Dendritic morphology includes a primary stem, secondary, tertiary, and sometimes even quaternary branches growing approximately in crystallographic directions.

Crystals grow from melts or solutions under certain conditions because they present thermodynamically more favorable configurations in these conditions. For instance, lowering temperature of the melt below the equilibrium one makes it less favorable—contain greater amount of the free energy—than the crystal of the same mass at the same temperature, see **Figure 2**. In the process of phase change, there will be a certain amount of heat and/or species released. Crystals will grow only if the latent heat and/or excess of species are removed from the growing entity. To make this process the most efficient, dendrites develop fingered branching structures with high surface-to-volume ratio. Dendrites form through the mechanism of morphological instability of smooth crystals: fluctuations on the front of a growing globular crystal increase and turn into small, but visible bumps of different shapes, sizes, and velocities of growth (**Figure 3(a)**). Growing bumps communicate with each other through thermal and diffusional fields and eventually select a particular spacing through the mechanism of competitive growth. Then the bumps turn into needles (primary stems) with selected shape and speed of growth (**Figure 3(b)**); later on the primary stems are overgrown by sidebranches (**Figure 3(c)**). The sidebranches select their spacing using the same mechanism of competitive growth: some branches of a dendrite go extinct (passive branches) and some survive down to the latest stages (active branches) (**Figures 1** and **3(d)**) [53-55]. Dendritic morphology includes rapidly moving convex tips of the needles and sidebranches and non-changing concave regions, called necks (**Figures 1** and **3**). The former are surrounded by the supersaturated melt; the latter, due to the negative curvature, are surrounded by unsaturated melt which does not support further growth of the crystal (**Figure 4**). Specific details of the dendritic structure and the rate of its growth depend on the driving 'force' that is, the free energy change after crystallization (**Figure 2**). The latter is proportional to the supercooling of the melt or, if solute additives are present in the melt, supersaturation of the solution before crystallization.

Dendritic growth is a highly cooperative mode of crystal growth controlled by the long-ranged diffusive field, thermal and/or species. It produces complex structures with the measure of complexity intermediate between that of a simple crystal and a DNA-like polymer. On the lowest level one has to specify the symmetry of the crystalline lattice and composition of the substance.

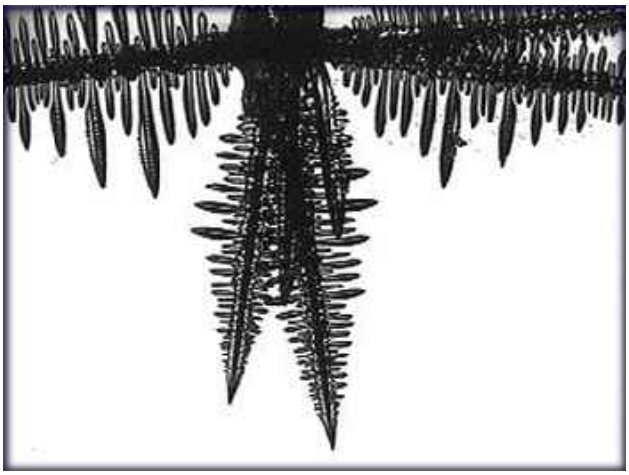

Figure 1. Dendritic structure of crystallization of pivalic acid (from LaCombe *et al.*); reproduced with permission.

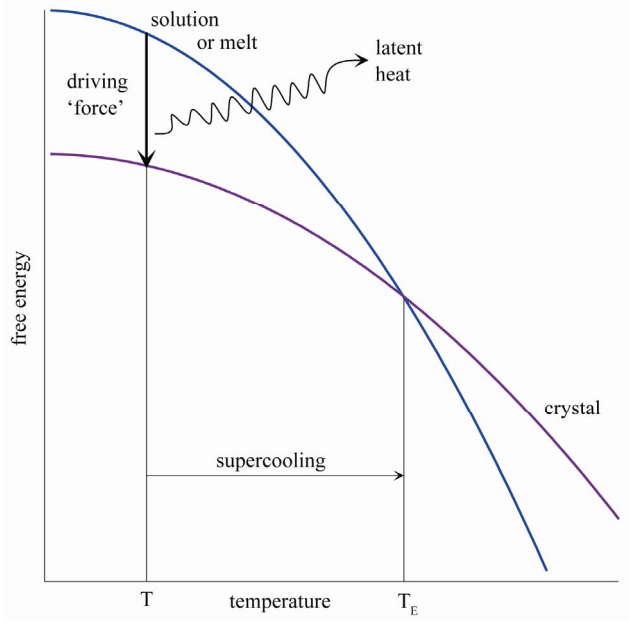

Figure 2. Free-energy versus temperature diagram of crystallization.

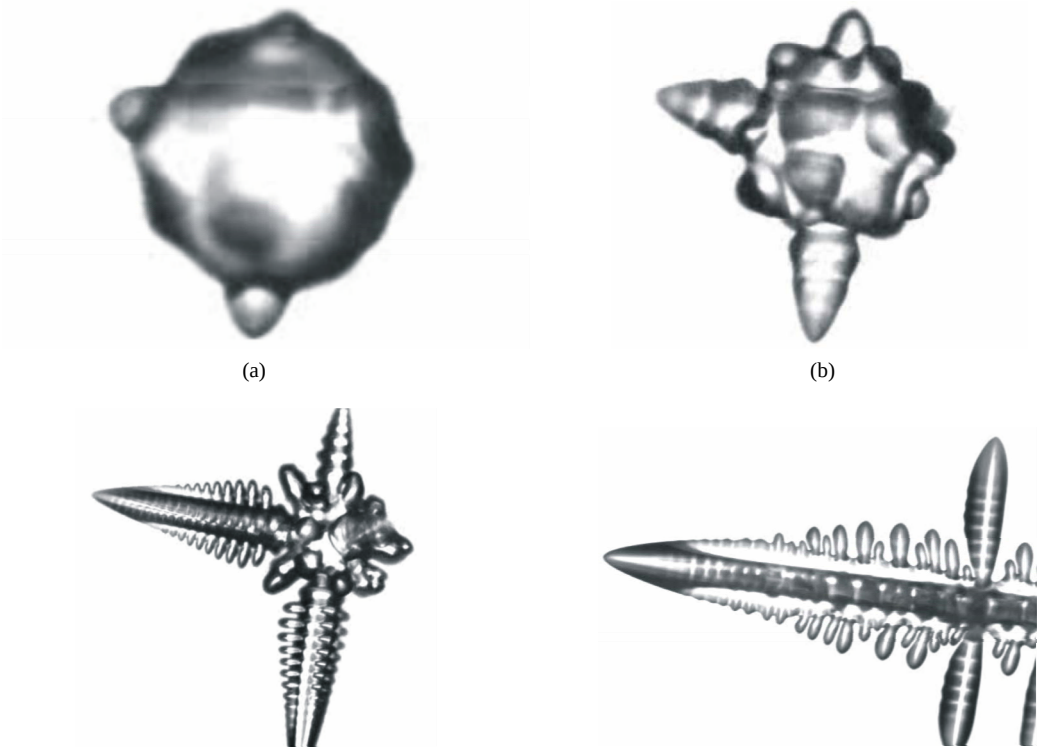

Figure 3. Time sequence of a growing crystal of ammonium chloride (a) initial fluctuations of the globular crystal; (b) formation of the primary stems; (c) formation of the side-branches; (d) competition between the side branches (A. Dougherty, http://ww2.lafayette.edu/~doughera/research/ crystal/index.html). The relative scale of the frames may be restored by comparing the central parts of (a), (b), (c) and the tip radii of (b), (c), (d).

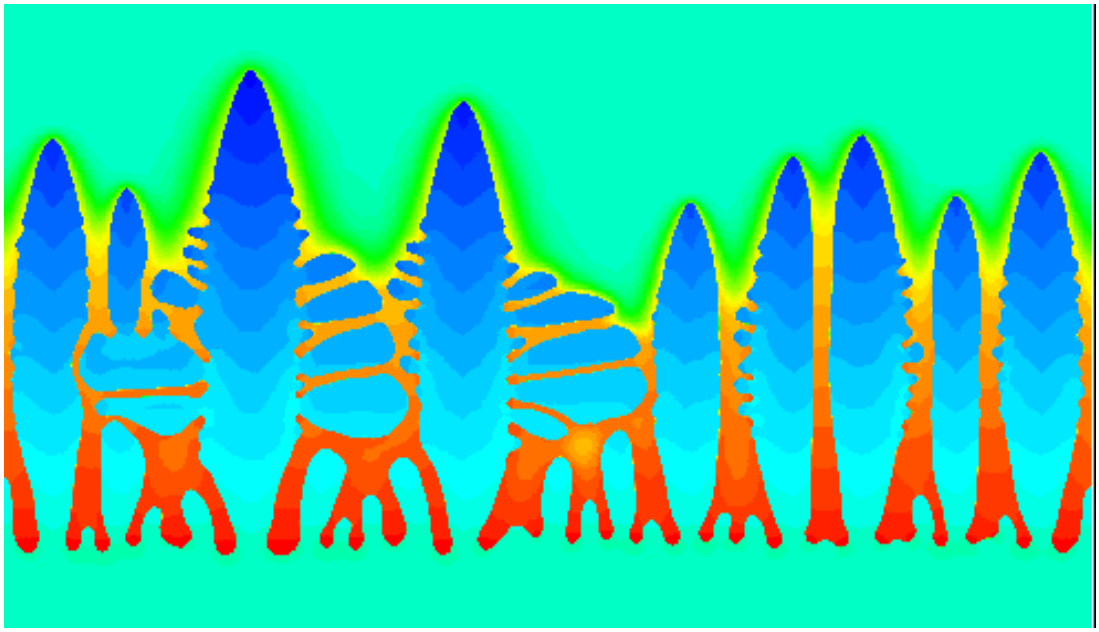

Figure 4. Numerical simulation of the dendritic growth in a binary alloy during directional solidification. Red, yellow, green, and lime colors represent diffusion field between growing crystals with the red corresponding to the highest concentration (lowest saturation) and lime—to the lowest concentration. (N. Provatas, J. Dantzig and N. Goldenfeld; reproduced with permission).

On the second level the structure is characterized by the periodicity of the side branches. The third level of complexity specifies positions of the active and passive branches in the system. The fourth level of dendritic complexity describes properties of the envelope of the dendritic structure—outline of the active branches.

Crystallization in supersaturated melts and solutions occurs naturally and does not need any enzymes other than inoculants of crystals (primary nucleation). If the inoculants are external particles or surfaces, the process is called heterogeneous nucleation [51, p. 93]. Dendrites are prone to fragmentation (**Figure 5**) that is, breaking off of small branches, which may be carried away from the parent structure by fluid flows into regions of greater supersaturation where they inoculate the solution (secondary nucleation) and start off another structure [51, p. 234]. Formation of dendrites in materials usually entails formation of 'grains' that is, individual crystallites formed by dendrites with different orientations obtained at nucleation (**Figure 6**). When the grains meet each other they form transition zones that is, grain boundaries, and the whole material obtains grain structure [56]. Grain boundaries are known to absorb trace components (atoms and molecules of different sizes), which, notwithstanding minute concentrations, significantly change properties of the entire material, e.g. turn it from ductile to brittle [57].

4. Justification

Firstly, all life functions are defined in terms that apply to living organisms only. To compare them to inorganic counterparts, the definitions must be, so to say, stripped off their 'life statuses' and considered just as natural processes. Otherwise we create an artificial divide between the organic and inorganic worlds, which may not allow us to reveal important relations between the two. Secondly, the life functions, as exemplified by the extant forms of life, are very sophisticated while the functions of a dendritic crystal that are discussed below are rudimentary. This, however, does not disqualify the latter from the status of predecessors of the former.

Growth of an organism is defined as irreversible increase in size and/or weight through synthesis of new material. For dendrites growth is a natural process that takes place under the appropriate conditions. Dendritic structures grow by way of rejecting latent energy and/or excess matter, a process which is greatly facilitated by fingered morphologies. *Metabolism* is a set of chemical reactions and transformations that require exchange of matter and energy between the growing system and the ambient environment, which serve the purpose of maintenance and propagation of the living system [58]. Dendritic metabolism is represented by rearrangement of the molecular species of the melt or solution, which makes maintenance and propagation of the crystal possible.

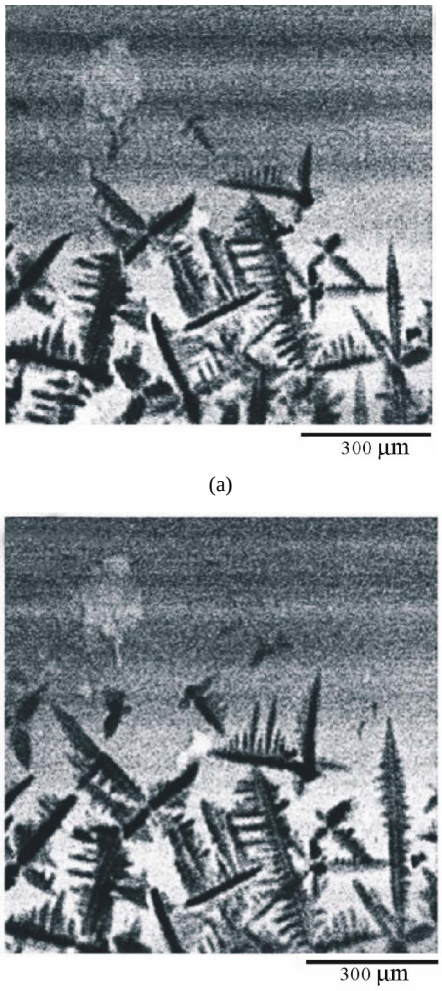

Figure 5. Fragmentation of dendritic crystals and their subsequent motion due to fluid flow.

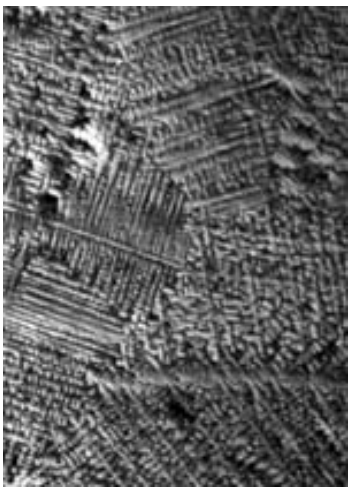

Figure 6. Granular structure of material after dendritic crystallization. Notice formation of grain boundaries between dendrites growing with different orientation.

Similar to the life metabolism, the dendritic one is accompanied by the release of latent heat and/or mass excess and their subsequent removal from the growing crystal. Unlike the life metabolism, the dendritic catabolism (breaking down of organic matter and harvesting energy) and anabolism (using energy to build components) are not separated in space and time. Notice that any process of crystal growth contains main traits of metabolism: steady flow of mass/energy to drive the machinery and a mechanism to use the free energy excess that comes with the flow for build-up of the new components. However, compared with the growth of a globular crystal, the dendritic metabolism has an additional, essential component. This is creation of the large amount of surface area, which is vital for dendrites because mass/energy exchange with ambience goes through the surface (**Figures 1**, **3** and **4**). Although free-energy reduction is the driving force of the growth (see **Figure 2**), additional surface area increases the free energy of the dendritic crystal compared to that of the globular one of the same volume. In other words, dendritic metabolism does not proceed completely 'downhill' (free energy decrease), it has an 'uphill' component (free energy excess) associated with the surface area creation. High density of interfaces in dendritic structures allows them to speed-up metabolism that is, remove heat and matter faster. The difference from a biological cell where this function is played by enzymes is that biological metabolism causes chemical changes, while dendritic metabolism causes phase changes.

Branching of dendrites may be considered as relic *division*. The most prominent property of the branching mechanism is its periodicity with the new branches being almost exact copies of the old ones. However, the new branches are more than just repetitions of the old ones because they carry information about the complexity of the whole structure; for instance, some of the branches are 'doomed' to stop growing very early, while others will grow up to large sizes (**Figures 1** and **3(d)**). The mechanism of periodic branching is similar to biological *replication*, which may be defined as "the ability to make copies of an information carrier" [59]. The difference is that the dendritic branching is an example of three-dimensional replication rather than one-dimensional replication of DNA. Contrary to amphiphilic vesicles that need external forcing for division [9,12], dendrites divide and replicate naturally because they 'do' this far from equilibrium.

Biological organisms contain *genetic information* which regulates their replication. Orgel [40] defined genetic in- formation as "the minimum number of instructions needed to specify the structure". Genetic information of a dendrite, according to Orgel's definition, is encrypted in its structure: it is contained in the special positions and sizes of the branches, same way as barcodes contain information about the product. Dendrites grow as self-similar, self-replicating structures with strict hierarchal order of branches, which is reminiscent of the order of generations in biological systems. Hierarchal structures of dendrites allow for the replication and propagation of genetic information from generation to generation.

Mutations in living organisms are defined as spontaneous changes of genetic information (DNA sequence). If the changes are 'found to be useful', they become permanently reflected in the reproductive process—*natural selection*. In abiotic systems, including dendrites, mutations correspond to thermal fluctuations and are similar to the genetic drifts. Dendritic fluctuations occur naturally because of the statistical nature of the systems; they appear in the form of small bumps, which compete for the fresh, unprocessed material in front of them (**Figure 3(a)**). A growing crystal produces more bumps than can survive to significant sizes. The bumps vary in the form and position; only those of them turn into needles which will make the nascent structure more efficient (**Figure 3(b)**). Later on, needles themselves will be covered by small bumps, future branches (**Figure 3(c)**), many of which will perish (passive branches) and only a few will survive (active branches) (**Figures 1** and **3(d)**). The survival of the dendritic branches is based on their geometrical positioning and timing of their appearance, which is the 'dendritic way' to pass genetic information to the future generations and, hence, make the mutation permanent. Although the selection principle for dendritic growth is still an active subject of research in the physics of pattern formation [49], it is absolutely clear that this principle is based on the stability of the growing structure. Thus dendritic structures demonstrate natural selection—differential reproduction—driven by stability and growth competition. This concludes the justification that dendritic structures possess *all* the essential characteristic functions of the living systems: growth, metabolism, division, replication, mutation and evolution in the form of natural selection.

Besides the basic functions of life, one can also see that dendritic structures possess built-in *homeostasis*. Indeed, if the ambient conditions change, e.g. temperature, pressure, or chemical potential, dendritic structures respond in many different ways in order to maintain the operating conditions. For example, if the temperature drops dendrites start growing faster, releasing more latent heat; this brings the surrounding temperature back to almost where it was before. Dendritic homeostasis does not come as a big surprise because, as known, homeostasis of biological organisms is an extension of the Le-Chatelier's principle of the abiotic world. However, there is another type of dendritic response to changing condi-

tions: dendrites make adjustments in the spacings of their primary and secondary branches [60,61]. At large supercoolings dendritic needles lose their branches and the crystal grows with spherulitic morphology. If the supercooling is great enough, the crystal may lose the needles all together and grow as a smooth entity [55,62]. One may consider these modifications as an example of dendritic *adaptation*. Thus, I have shown above that dendritic crystals possess all the basic biological functions.

In addition, one can notice that the organic world has 'intricate relations' with the dendritic morphologies. To begin with, many pure organic materials and their aqueous solutions undergo dendritic crystallization when they are cooled below their liquidus temperatures. Typical examples are ammonium chloride [63], pivalic acid [64], cyclohexanol [62], succinonitrile [54], cholesterol [65], and protein streptavidin [66-68]. Under 'lagoon-like' conditions aqueous solutions of potassium cyanide and ammonium hydroxide yield heterogeneous cyanide polymer particles [69]. When these particles were dissolved in dimethyl sulfoxide and allowed to dry on a microscope slide, they showed branched tubular morphologies reminiscent of snowflakes. Nucleotides and amino acids are known to crystallize with dendritic and spherulitic morphologies. Ramachandran and Natarajan [70] showed that L-tyrosine crystallizes in silica gel having spherulitic morphology with long needles. This is also true regarding the crystallization experiment of Shinitzky *et al.* [27] (D. Deamer, personal communication). Dendritic pattern of liquid-crystal growth in organic materials is a common place [71]. Spontaneous ordering of high concentrations of short strands of nucleic acids into a liquid crystalline phase displays dendritic structures [72]. Importantly that this process promotes selection and segregation of complementary sequences and ligation of neighboring strands by physical polymerization.

Furthermore, organic additives change crystallization pattern of many inorganic substances. Lopezcortes *et al.* [73] studied influence of halobacteria in the crystal formation of halite. Their analysis suggests that the proteinaceous constituents of extremely halophilic archaebacterial surface layers may determine the crystal form of halite and even "yielded dendritic crystals". Shibata *et al.* [74] studied effect of human blood additions on dendritic growth of cupric chloride crystals in aqueous solutions. Their evidence suggests that components of blood including amino acid, peptide and/or protein or some composition of them were chemisorbed on the dendrite surfaces. Eiden-Abmann *et al.* [75] studied the influence of amino acids on the formation and morphology of hydroxyapatite (calcium phosphate) in gelatin. They found that additions of amino acids (Asp, Glu, Ser, etc.) to the gelatin results in formation of spherulites consisting of many thin needles. Then, one can imagine how these materials grew from the prebiotic soup (heterogeneous mixture of organic compounds) once the temperature on early Earth was dropping below their liquidus temperatures.

Moreover, the dendritic morphology confers operational advantage to extant forms of life. Many plants have forms reminiscent of dendritic crystals [76] (although in crystals energy is received from outside while in plants—from inside [77]) and great similarities exist between the cellular morphology of plant tissue and structure of binary alloys undergone directional crystallization [78]. Animal bones have dendritic structure to allow for fluid flow through them [79]. Bacterial colonies, growing under conditions of starvation, form dendritic morphologies [80]. Also bacteria can trigger mineral formation under saturation conditions, but the reasons why bacteria favor or promote mineral nucleation are still unclear [81]. The nerve cells, neurons, have branching structures (also called dendrites due to their tree-like morphologies). Observations of neurons of different species suggest that neural branched geometry is certainly related, in part, to the expression of genetic factors, which are present during phylogenesis [82]. Even the process of transcription of DNA into RNA has dendritic morphology with DNA representing a primary stem and RNAs—sidebranches [83]. Curiously, the phylogenetic tree itself is morphologically very similar to crystalline dendrites, e.g. it has passive branches and active ones [84].

To summarize, on the one hand, dendritic crystals were present on the early Earth; on the other hand, dendritic morphologies are broadly utilized by extant forms of life. Hence, we may envision that the two are evolutionarily connected through a kind of 'branching gene'. Starting off with a dendritic-arms gene of the protobiont, which as we saw above can hold replicable information favoring its own propagation, it evolved into something like a clay gene [85] and a DNA gene at a later time.

The hypothesis of the dendritic nature of protobionts allows us to establish analogy between the existing components, functions, and other processes of biological organisms and their primordial counterparts. For example, crystallization is analogous to polymerization; nucleation—to heterogeneous catalysis; fragmentation of dendritic structures plus secondary nucleation of new structures is analogous to migration and 'gene flow' in biological systems. Grain structure of the material after crystallization is an analogue of cellular organization with the grain boundaries playing the role of cell membranes. The species segregated at the grain boundary are analogous to the membrane proteins, which are responsible for charge transfer through the membrane. According to my

hypothesis, the supersaturated solution is the forerunner of the food for modern organisms, while the unsaturated one—of the waste; crystal grains are prototypes of cells (preprocaryotes), and dendritic branches—of generations. Many authors noted profound similarity between the processes of crystal growth and enzymatic chemical reactions [8,85]; hence, mineral surfaces and inoculants are primordial enzymes and active sites. Diffusion of charge, heat, and solutes served as the transport system through premembranes and was part of the fossil metabolism. As known, individual cells have ability to sense chemical gradient and cell's development appears to be regulated by diffusible molecules—the process of chemo taxis. Hence, chemo- and thermo-taxes of microorganisms are rooted in the chemo- and thermo-taxes of dendrites (protobionts). A very high surface-to-volume ratio of dendritic structures was certainly favorable for catalysis of other biological reactions and transformations on their surfaces-prebiotic autocatalysis. Morphology of dendrites is their phenotype while complexity—the genotype. Genotype and phenotype of the primordial organism were not separated ('naked gene' of sorts, [46]), which is analogous to RNA world where one molecule (RNA) combined both types. These relations are reflected in the **Table 1** below.

5. Scenarios of the Origin of Biological Materials

If one accepts the geological data that support the fact that around the time of the origin of life early Earth was very near the freezing point of water [86-89], then the dendritic protobiont hypothesis may allow one to conjecture a scenario of thermo-chemical precipitation of the biologically important material. Monomeric components of the genetic apparatus precipitated in shallow water pools of dilute multi-component aqueous solutions of diverse organic molecules with the surfaces of rocks or

Table 1. Analogy between the extant components, functions, and other processes in biological organisms and their primordial counterparts.

Existing (biological)	Primordial (dendritic protobiont)
Components	
Organism	Dendrite
Properties (phenotype)	Morphology of the structure
Genetic information (genotype)	Complexity of the structure
Generations	Dendritic branches
Food	Supersaturated solution
Waste	Unsaturated solution
Active sites and enzymes	Mineral surfaces and inoculants
Cell	Crystal grain
Cell membrane	Grain boundary
Membrane protein	Species segregated at the grain boundary
Main Functions	
Growth	Growth
Metabolism	Rearrangement of the molecular species plus diffusion of excess heat and/or species
Division	Branching
Replication	Periodicity of branching
Mutation and genetic drift	Thermal fluctuations
Natural selection	Selection principle?
Other Functions and Processes	
Polymerization	Crystallization
Heterogeneous catalysis	Heterogeneous nucleation
Migration and gene flow	Fragmentation plus secondary nucleation
Homeostasis	LeChatelier's mechanism
Adaptation	Geometrical position and timing of branches

clays serving as catalysts of nucleation. The newly precipitated crystals grew by dendritic mechanism. When dendrites grow from aqueous solutions the supersaturation of the solution can be achieved through the processes of cooling and/or drying, both of which took place in pools of water on early Earth. Different substances have ability to precipitate from aqueous solutions using different mechanisms of growth. However, dendritic mechanism conferred an evolutionary advantage because, arguably, it is the fastest mechanism of growth, and the competition between a few substances that precipitated from aqueous solution by different mechanisms was won by dendrites—the quick had better chance to come alive on early Earth. "Selection pressure favors any chemical system that can process matter more rapidly and make more of its kind." [58] Due to highly cooperative nature of crystallization, dendritic structures created high concentrations of biomonomers at one place—the concentration gap problem. The genetic materials of increased sophistication appeared through the successive and overlapping stages of material coevolution where the dendritic protobionts were on the lower steps of the case and the organic biomaterials on the upper ones, "a genetic staircase"-type scenario [18]. Branching morphologies, once started as a physico-chemical process, 'entered' the genome on the later stages of evolution.

This scenario may apply to amino acids whose crystallization is known to purify the material from water [27] and make the reaction of polymerization more probable. High cooperativity of crystallization could have been the reason for the appearance of biochirality because dendritic crystallization is also known to enhance the SCSB mechanism through fragmentation [11,22,23]. Periodic temperature variations, e.g. due to circadian cycles, provided the source of free energy and caused periodic freezing and thawing of dendritic structures with the reaction of polymerization taking place in the molten state. New cycles of crystallization led to formation of more and more organized matter with clearly living functions. In a way, these cycles were the first example of evolution by way of extinction and speciation [90,91]. A set of experiments may be suggested to test this scenario. For example, one may subject a dilute solution of amino acids to periodic temperature variations around the freezing point and watch for the formation and growth of peptide chains in the solution.

One may envision other scenarios that also allow transferring dendritic functions onto the organic world; for instance, formation and growth of inorganic crystals, e.g. calcite ($CaCO_3$) [92], in the prebiotic conditions and subsequent adsorption of organic substances on their surfaces [25,26]. This mechanism, however, has been considered in the literature and will not be elaborated here any further. Notice that all scenarios indicate that biochirality appeared together with the first signs of life and cellular organization.

6. Discussion

Based on the fact that growth of dendritic crystals of inorganic or simple organic molecules possesses *all* basic functions of life and may contain 'genetic' information stored in their branches, I presented a hypothesis of the dendritic nature of a protobiont. According to this hypothesis the protobionts formed through a physico-chemical process known as dendritic crystallization. The branching dendritic crystalline structures helped build living systems by lending them functions so that organic chemical evolution is just one natural consequence of the evolution of matter in the universe. A self-replicating biological system with adaptation emerged from simple molecules using a completely abiotic mechanism of nonequilibrium phase transitions. Dendritic structures assisted the emergence of the genetic apparatus, which otherwise would have been thermally improbable. This mechanism could act simultaneously or intermittently at different places on the early Earth and created similar structures everywhere. Hence, to explain the similarities in the living systems there is no need to invoke the concept of LUCA because, according to the hypothesis, they arise as a result of thermodynamic necessity. The dendritic protobiont hypothesis supports the assumption of a 'second genesis of life' [30,93] and helps explain why "life established itself on Earth fairly quickly once conditions permitted" [33]. The full and complete biological functionality was already lurking 'deeply in the inorganic world', waiting to be revealed and utilized. Although the dendritic crystals were the living organism in the primordial world, they should not be considered contemporary living systems because they are not made of the right material, macromolecules. One should not forget also that the primordial conditions were completely different from the present ones.

Although there are other abiotic systems that possess some of the biological functions, significance of the dendritic crystal growth mechanism is in that it possesses all of the basic life functionality. Obviously, the dendritic scenario does not necessarily need carbon-based solutes in water-based solutions; it can work with e.g. silicon-based solutes and/or hydrogen sulfide-based solvents. This may have an important implication for extraterrestrial origin-of-life scenarios.

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