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THE ORIGIN OF BIOLOGICAL MACROMOLECULES ON THE EARTH
- THE HYPOTHESIS OF INORGANIC TEMPLATE

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16. Abstract Vol. 21, 1976. pp. 112-119 The author reviews studies about the origin of life. He then discusses the nonrandom organization of organelles from a structural and functional point of view. After postulating that the origin of biomacromolecules was not a random event, the paper develops the hypothesis that polypeptides and polynucleotides were formed on an inorganic template. Only "information-containing" structures can pass natural selection and develop through evolution.			
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THE ORIGIN OF BIOLOGICAL MACROMOLECULES ON THE EARTH
- THE HYPOTHESIS OF INORGANIC TEMPLATE

Tzyy-Shen Lu*

The question of how life originated in the early history of the earth has been given increasing attention from scientists. It also has aroused a great controversy between Materialists and Idealists in philosophy. All the Idealists who favor exploiting labors believe that God created the first life. Although the ancient Materialists accepted the concept that life originated from the prebiotic world, they could not find any scientific evidence for this hypothesis. About a hundred years ago, Engels pointed out that life must originate from chemical reactions. This postulation seems to be the only right answer to the origin of life. /112**

Engels has also indicated that life is a product of nature. Protein is a natural biological compound formed under optimal conditions, and also a product of chemical reactions. The above two statements are compatible with each other. Lenin once pointed out that the earth was formed long before organic compounds were synthesized. According to Materialism, the emergence of life is merely an important step in the development of the earth. In order to understand the origin of life, we should start with prebiotic chemistry, as indicated by Lenin.

Many extensive studies have been done about the origin of life during the past fifty years. The subjects which have been studied are: 1) formation of the building blocks of biological macromolecules - amino acid and nucleic acid. 2) origin of biological macromolecules on the earth. 3) the formation of life from biological macromolecules.

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** Numbers in margin indicate pagination in original foreign text.

Several studies suggest that: the primitive atmosphere was very different from the present atmosphere. It contained little or no oxygen, no ozone, and was essentially a reducing medium. Under these conditions, the extremely intense radiation, together with lightning, thunder, and earthquakes can induce some chemical reactions. Many researchers successfully synthesized amino acid and nucleic acid from simple chemical compounds under a simulated prebiotic environment. These studies indicated that the building blocks of macromolecules might be formed long before the emergence of life.

The purpose of this study is to explore the origin of biological macromolecules.

How could amino acid and nucleic acid organize in a sequential order to form biological polymers? And further - to form protocells? There are two hypothesis dealing with these questions: Coacervates' theory of Oparin and the protenoid microsphere theory of Fox. About fifty years ago, Oparin postulated that the coacervates were capable of interacting with the surrounding medium by osmosis, selectively absorbing amino acid and nucleic acid which could form polymers within /113 the colloids. This function resembles the metabolism of present cells. The framed polymers can assemble together to form protocells through long-term evolution. Fox proposed that polymers of amino acids could form proteinoïd microspheres. On this basis, the interaction between different biopolymers can lead to the formation of protocells. Many studies have been done about these two hypotheses. Considerable supporting evidence was obtained, but the weakness of both hypothesis has also been found; both of them believed that the prebiotic genesis of monomer and polymer, and the assembly of polymers occurred randomly. Their basic mistake was randomness in their concepts, which led them in a wrong direction.

The modern cell originates from a primitive biological substance; therefore, there must be a close relationship between these two structures. In order to explore the origin of life, it seems to be necessary to understand the characteristics of present cells. Many studies of molecular biology have suggested that the organization of cells follows the sequence: nonrandom polymerization; nonrandom organization of organelles or cells from biopolymers; functional organization.

(a) Nonrandom Polymerization

Protein is a biopolymer, consisting of 20 amino acids, which are arranged in a unique sequence. Within one species, there is only one (or a few) amino acid sequence for one specific protein, no matter how big the protein is. Occasionally, a defect in the polymerization (possibly one amino acid substituted by another amino acid) will cause an illness known as molecular disease. Nevertheless, the new sequence may also imply a big step in evolution. Differences in the composition of related homologous hormones in distinct species will point out the closeness of the relationship between species; the more different the sequences are, the less related the species are. This distinct difference will only appear at certain positions in the sequence. Several distinct insulins have been shown chemically in Table 1 and Figure 1. It has been suggested that all insulins may have originated from one proinsulin chain, which breaks up to form active insulin with the deletion of the C-peptide chain. Active insulin consists of two peptide chains: an A-chain with 21 amino acids and one disulfide linkage; B-chain with 30 amino acids. These two are joined by two interchain disulfide linkages. Among the 23 species examined in our study, amino acid substitutions have been recorded at 29 of the 51 positions in the insulin molecule [1].

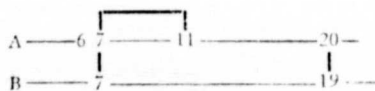


Figure 1: The structure of the insulin molecule.
A, B; A- and B- polypeptide chains
Wide black lines indicate the disulfide linkages.

The relationship between chemical composition and species evolution has mostly been done on cytochrome C and hemoglobin. These studies suggested that the functional protein allowed limited substitutions of amino acid, but not at the functional positions in the protein. This indicates that there is a certain order in the structure.

The sequence of nucleotides in nucleic acid has also been determined. All the studies have suggested that there is a correlation between the closeness of the relationship and chemical structure of the nucleic acid. All the homologous nucleic acids contain the same functional

Table 1: The distinct difference in amino acid sequence of insulin.

Species	Sequence no.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21
Human, pig, dog, rabbit, sperm whale		Gly	Ile	Val	Glu	Gln	Cys	Cys	Thr	Ser	Ile	Cys	Ser	Leu	Tyr	Gln	Leu	Glu	Asn	Tyr	Cys	Asn
Beef									Ala	Val												
Sheep, goat, horse									Ala	Gly	Val											
Elephant										Gly	Val											
Whale									Ala	Thr												
Peking Duck									Glu	Asn	Pro											
Chicken, turkey									His	Asn	Thr											
Mouse (I, II)																						
Guinea Pig									Ala	Gly	Thr		Thr	Arg	His							
Beaver Mouse									Asn				Arg	Asn								
Cod									His	Arg	Pro		Asp	Ile	Phe	Asn						
Angler Fish									His	Arg	Pro		Asp	Ile	Phe	Asn						
Toad fish I									His	Arg	Pro		Asp	Ile	Phe	Asn						
Toad fish II									His	Lys	Pro		Asp	Lys	Phe	Asn					Ser	
Tuna I, Bonito I									His	Lys	Pro		Asp	Ile	Phe							
Tuna II									His	Arg	Pro			Ile	Phe							
Bonito II									His	Arg	Pro			Ile	Phe							
Round-mouth fish									His	Lys	Pro			Ile		Asp						Gln

Table 1: (continued)

Species	Sequence no.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21
Man, elephant	B1	Phe	Val	Asn	Gln	His	Leu	Cys	Gly	Ser	His	Leu	Val	Glu	Ala	Leu	Tyr	Leu	Val	Cys	Gly	Glu
Beef, pig, bird, goat, sheep, dog, whale			Ala	Ala																		
Peking duck			Ala	Ala																		
Turkey, chicken			Ala	Ala																		
Rabbit					Lys																	
Mice (I)					Lys																	
Mice (II)											Asn				Thr						Gln	Asp
Guinea Pig											Gln				Thr						Arg	His
Beaver mice				Tyr		Ser																Asp
Cod																						Asp
Angler Fish																						Asp
Toad Fish																						Asp
Tuna I																						Asp
Bonito I																						Asp
Round-Mouth fish																						Val

Table 1 (continued)

Species	Sequence no.	22	<u>23</u>	<u>24</u>	<u>25</u>	<u>26</u>	27	28	29	<u>30</u>
Man, elephant		Arg	Gly	Phe	Phe	Tyr	Thr	Pro	Lys	Thr
Beef, pig, bird, goat, sheep, dog, whale										Ala
Peking Duck							Ser			
Turkey, chicken							Ser			Ala
Rabbit										Ser
Mice (I)										Ser
(II)									Met	Ser
Guinea Pig		Asp				Ile			Asp	
Beaver mice		(-	-	undefined	-	-)		
Cod						Asn				-
Angler Fish						Asn				-
Toad fish						Asn	Ser			-
Tuna I						Asn				-
Bonito I						Asn				-
Round-Mouth fish		Asp				Thr	Lys, Met			

Amino acid sequences in 23 vertebrate insulins are shown in this table. Numbers indicate the positions in the sequence. The underlined number represents no substitution of amino acid on this position. The blanks in sequences indicate that this insulin possesses the same amino acid as human insulin at this particular position. The "-" sign stands for no amino acid at this position.

structure. Obviously, the synthesis of nucleic acid is also a non-random process.

The tertiary structure of biopolymers contributes greatly to its unique function in organisms. Denaturation, the destruction of the tertiary structure, can cause a defect in protein function. Studies of the mechanism of denaturation have suggested that the primary structure of protein possesses inherent organizational characteristics which would have aided the formation of specific higher-level structures under optimal conditions. The unique function of protein appears at the formation of the higher-level structure. Similar evidence was found in nucleic acid. Therefore, we can conclude that the higher-level structure of the bio-macromolecule is not a product of a random process, its organization exists in its primary structure.

(b) Nonrandom Organization of Organelles from Macromolecules.

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a. Structural point of view.

The cell is the building block of the organism. Each cell consists of organelles which are formed by the nonrandom arrangement of biomacromolecules. Some cells, like muscle cells shown in Figure 2 and virus cells in Figure 3, reveal a rigid cell structure pattern. The cell membrane does not show a rigid structure pattern, but it still contains the orderly arrangement of biopolymers. Recent studies on nucleoprotein have suggested that it is composed of histones which are formed by many amino acids surrounding the nucleic acid. The nucleoprotein can be formed from its components under certain conditions.

b. Functional Point of View.

The unique function of a biological system is based on the non-random arrangement of biomacromolecules, and metabolism is just a series of biochemical reactions controlled by enzyme systems. For example, muscle contraction is attributed to the specific arrangement of filaments in the myofibril; protein synthesis is a coordination of functions of RNA and enzyme systems.

The studies on different organelles (or cells) have suggested that the orderly structure and unique function are not a result of a random occurrence, but a nonrandom process.

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Figure 2: Electron micrograph of human muscle tissue.



Bundles of myofibrils arranged in order like bamboo grooves. There are several bands of different sizes traversing each myofibril. They are the I-band consisting of Z-lines, and the A-band consisting of H-lines and M-lines.

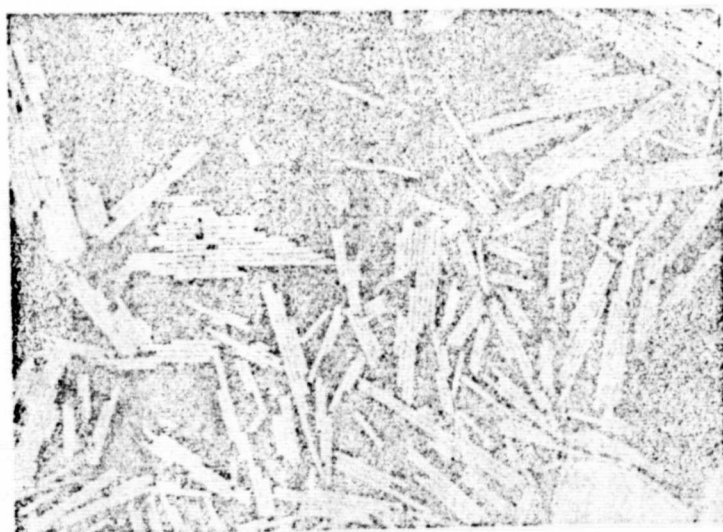
In the A-band, many filaments line up along the longitudinal direction of the myofibril, which are known as thick filaments. The thin filaments can be vaguely seen between thick filaments. Both filaments are composed of specific proteins. The contraction and relaxation of muscle are mainly attributed to the relative movements of these two filaments.

The muscle in this figure is under contraction. The I-band will be under relaxation longer.

SR: membrane.

M, G: other organelles.

Figure 3: The electron micrograph of tomato virus (magnif., 35000 X)



The rods shown in this picture, which are of different length but the same radius, are tomato virus molecules. A black line can be vaguely seen at the center of the rod, which indicates that the composition of the central part is different from that of the other parts. Stripes can be seen vaguely on the rod, which suggests the orderly arrangement of biomacromolecules in the virus.

Contemporary biological studies suggest that the orderly arrangement of biomacromolecules contributes greatly to the functions of the organism.

As mentioned earlier, both Oparin and Fox believed that the emergence of life occurred by chance. Many western scientists are still under their influence. Eigen, a German scientist has pointed out that the origin of life must be random, and the self-assembly associated with it also occurred by chance. Therefore, the question of how life can reach orderliness from randomness has always been the biggest problem for these hypotheses. Many scientists, including Fox, have tried to find an interpretation for this question. Fox assumed that the unique sequence of the peptide chain was due to the selective interaction between amino acids, but he failed to verify his own hypothesis through his studies. Sorm, a Czech, also failed to find any evidence. Orgel et. al., [3], tried to tackle the question with nucleic acids. He suggested that nonrandom nucleic acids came before nonrandom proteins. This suggestion certainly has its own merit; however, one will have to answer the question of how nonrandom nucleic acids occurred.

Since it is difficult to find an interpretation from randomness to orderliness, why cannot we assume that the origination of the biomacromolecule was not a random event and it was produced from the prebiotic world?

In the evaluation of coincidence and certainty, Engels has pointed out that metaphysics leads to a controversy between coincidence and certainty. He has also indicated: coincidence and certainty are highly compatible. An "occasional" incidence might be controlled by "certain" inner factors; the problem is just to find out these factors. The emergence of primitive life is a result of natural selection from the repeated appearance of coincidences, which is an inevitable product from the earth's development.

Chairman Mao has pointed out: The basic reason for material to develop is the contradiction existing in itself, not outside the substance. This contradiction exists in every substance, which can cause the change and development. Since life is an inevitable pro-

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duct of the earth's development, one then should try to understand the appearance of biological substances from the material evolution on earth.

There are five forms of evolution: mechanical, physical, chemical, biological, and social changes. Engels has pointed out that the consistency of all changes in nature is no more a philosophical topic, but a scientific fact. He also indicated that one form of change must be developed from the former one. Since the biological changes follow the prebiotic changes, the former must derive from the latter, and the origination of biological substances must exist in the prebiotic world. As mentioned earlier, the present synthesis of biomacromolecules is nonrandom, so we can simply speculate. The original synthesis of macromolecules was not random; it existed in the prebiological world. In other words, a prebiotic template which led to reproduction of the original biomacromolecules existed in the prebiological world.

This hypothesis can be approached from two directions:

1. The amount of necessary material for original polymers.
2. The possible amount of material can be provided by inorganic templates.

For the first part of the hypothesis, we find some evidence in the protein: 1) for some simple proteins, its function is mainly /117 attributed to the primary structure, not secondary or tertiary. 2) only a few proteins, insulin, chymotrypsin, thrombin, lysozyme, Stuart-Prower factor, fibrinogen, and γ -globulin, contain two peptide chains with disulfide linkages in the structure. The former five were first formed as one chain molecule, then cleaved into two chains by an enzyme. These complicated proteins, except the first two, can all be found in blood which is in a later stage of evolution. Therefore we can assume that the original proteins contain only one simple chain. 3) nowadays, we can find 20 amino acids in protein, but Jukes suggested that the original proteins were composed of only 10 amino acids. Extensive studies are needed on Jukes [4] hypothesis; nevertheless, it seems acceptable. From the above observations, we can assume that the original proteins are very simple; therefore, the inorganic material for its formation will not be too complicated.

The same hypothesis can apply to nucleic acid; primitive nucleic acids were all one chain biopolymers. There were no classifications of RNA and DNA. Moreover, some rare bases of contemporary nucleic acids are formed by methylation after the formation of nucleic acid. This may imply that these rare bases do not exist in the primitive nucleic acids; they are products of later evolution. Under these circumstances, we can also hypothesize that the inorganic material for the formation of those primitive nucleic acids might not be too complicated.

In summary, if original nonrandom biopolymers were formed from inorganic templates, the necessary amount of material would not be as large as expected. Only a few more complicated materials are needed to serve this purpose.

Next is the question of how much information the prebiotic world can provide. Even the prebiotic world was not completely chaotic; it contained some orderly-structured inorganic crystals, including optically asymmetrical compounds. Crystals of simple compounds have orderly structures, but they are too simple to contain a great amount of material. Therefore, the inorganic template would most probably exist in a large, complicated crystal. Bernal [5] mentioned vaguely the concept of "inorganic template" in his publication in 1951, but disclaimed his own idea sixteen years later. Cairn-Smith [6] studied in detail some crystal properties of mica including the lattice structure, dislocation and defects, and suggested that those properties made mica capable of serving as an inorganic template in the original biopolymer synthesis.

The surface of the earth is mostly covered by clay. Its main constituent is silicate, which is very complicated both in terms of chemical structure and crystal structure. We try to use Montmorillonite as an example to study the possibility of inorganic layers, Si-O-Si and Al-O-Al layers. The distance between layers can change due to the adsorption of water or other molecules. Many elements, Al, Zn, Fe, Mn and Mg, can replace Si in the Si-O-Si layer to form negatively-charged structures. Al in the Al-O-Al layer can also be replaced by Mg. Montmorillonite can adsorb many organic compounds including polar compounds such as alcohols, aldehydes and amines, and less polar aromatic compounds. The adsorbed molecules will either

form a homogeneous or heterogeneous layer within the crystal lattice. It has been suggested that the activated amino acyl compound is adsorbed by Montmorillonite; then the amino acid will move into the interlayer space. The amino group of amino acid will attack the negatively-charged portion of the Al-O-Al layer (e.g., O atom in Al-O-Al). We can also assume that the side chains of amino acid will attack the opposite charged portion of the crystal layers. The less polar side-chain, aromatic group and hydrophobic lipids, will interact with the less polar portion of the layer. The difference in polarity of the side-chains will decide the location of interaction between crystal and adsorbed molecules. This unique arrangement can be used as the inorganic material for primitive protein synthesis. Moreover, the substitution of Al by other elements in the Al-O-Al layer can cause differences in the interlayer space. Therefore, the location of interaction between crystal and adsorbed molecules can also be attributed to the individual amino acid. From the above observations, Montmorillonite seems to be a possible inorganic template for original biopolymer synthesis. Of course, there are other properties needed for a compound to become an inorganic template.

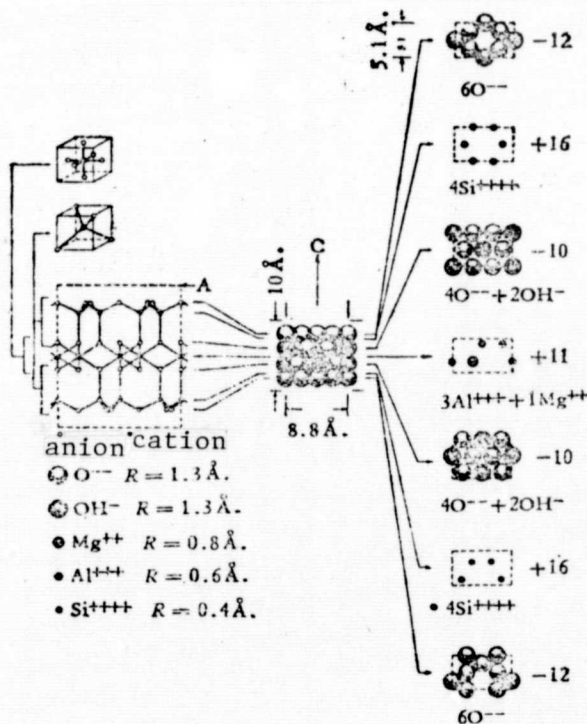


Figure 4: The structure of Montmorillonite.

The structural diagram on the left shows the layer structure of Montmorillonite. The top and bottom layers are Si-O-Si layers; the one inbetween is Al-O-Al layer. The layer existing between layers of Montmorillonite is the adsorbed molecules.

The middle diagram is an accumulation picture of atoms represented by balls which are made in proportion to their radii.

The diagram on the right shows the arrangement of atoms in different layers along the C-axis of atoms in each layer. From the arrangement of Si and Al, we can understand the possibility of replacement of these atoms by other atoms.

Many studies have been done on the inorganic template. Paecht-Horowitz et al., [7] reported that polymerization of alanine and adenosine was enhanced 30-40 times by the introduction of Montmorillonite into the aqueous amino acid solution. Degens and Metheja [8] also reported that the addition of Montmorillonite into a dry amino acid mixture or an 80°C aqueous amino acid solution can enhance the polymerization. Nevertheless, the alkaline amino acid cannot be found in the polymer, because of its strong affinity to the lattice of Montmorillonite. The same catalytic effect of kaolinite was observed. Orgel also carried out similar studies. The above-mentioned studies only show the catalytic effect of silicate, but - from the "inorganic template" point of view - whether these inorganic compounds really provide information for original protein synthesis needs further verification. However, these studies did show that those inorganic compounds possessed all the properties of being a template.

There are many inorganic compounds other than Montmorillonite which can serve as an inorganic template. For example, silica gel and Al_2O_3 can separate amino acid, which indicates their potentials as inorganic template. Many studies have indicated that many rocks contain different kinds of amino acid, even the meteor contains a certain kind of amino acid. The adsorption of amino acid by rock seems to be very common. The hypothesis mentioned above can also apply to nucleic acid. Since there are only a few varieties of bases in nucleic acid, the inorganic template for its polymerization must be less complicated than that for original protein synthesis.

Primitive biopolymers are different from the present biopolymers. /119 Their primary structures are much simpler. There is a long-term evolution involved from the appearance of high molecular weight biopolymers to "life". But original biopolymers have new properties that simple compounds do not have. Therefore, evolving from simple compounds to primitive biopolymers, is a change in properties.

How did the primitive biopolymer evolve to life?

Recent studies in molecular biology have suggested that many organelles can be formed under optimal conditions. For example, the flagellum of bacteria can be broken down into protein molecules and then reassembled to form another flagellum. The formation of TMV (Tobacco Mosaic Virus) and nucleoprotein from their constituents

has also been observed. Although the complete formation of mitochondria from its separate constituents has not been successful, the active oxidative phosphorylation fragments were reformed from the protein constituents. Unfortunately, the formation of the cell membrane is not successful, but one would assume that it is only a problem of finding the right conditions. It has been commonly accepted that the formation mechanism is similar to that of combination of molecules. Of course, the mechanisms and conditions of cell formation need more extensive studies. The formation of some organelles requires factors outside the organelles, but there must be an inner factor which can trigger the formation. This inner factor plays a more important role in the organization of protocells.

On the basis of the above hypothesis, we can assume that primitive proteins and nucleic acids once synthesized can form more complicated organelles and then a primitive organism. There might be several proteins or nucleic acids formed on one inorganic template, which could self-assemble to form a system that has the potential to develop a biological function. The polypeptides and polynucleotides formed on the inorganic template are reproducible, but only part of those reproducible structures possess the potential of developing a biological function. Darwin proposed the theory of "natural selection, or survival of the fittest". Natural selection plays an important role in evolution from biomacromolecules to the primitive organism. The reproduced macromolecules selected by nature contain certain "information" in their orderly structure. The rest of the macromolecules selected against nature would eventually disappear. This touches the relativity of ordered structures. "Ordered" is relative. Only a portion of the ordered structure compound from an inorganic template can pass the selection and evolve into complex systems and eventually, life. To illustrate further, we will call these nature-selected, orderly structured compounds as "information-containing" substances. It can be seen clearly that the "information-containing" structure in biopolymers is different from the "ordered" structure in an inorganic template. The former comes from the latter, yet higher than the latter. This is consistent with the "development" philosophy in Materialism. As for the "ordered" structure of inorganic material in the template, it is beyond the scope of this paper.

The hypothesis discussed in this paper is that polypeptides and polynucleotides were formed on the inorganic template. Only "information-containing" structures can pass natural selection, and develop into complex systems through long-term evolution.

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