Original synthesis Article

Man creation had began since the creation of the first biological material very likely in Clay

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Abstract – Among conclusions deduced from my recent deep study of holy Qur'an verses related to the topic of man creation, the man creation had begun a very long period of time before his emergence. It had begun with the creation of his earthy and clayey nature in clay. Very likely "his earthy and clayey nature" means the basic structures of his bio-molecules that, common to all living beings, are formed from elements (ions) present in soil and clay. Thus the objective of the creation of living beings is to reach that of humans. In the present paper I will show that this conclusion is not in discordance with science. In fact, I present a synthesis of dispersed published data and new complementary insights for showing how montmorillonite clay could be the cradle of the early life evolution. I propose that the birth of life would occur within Earth's surfaces on saturated clay bedrock, and in montmorillonite crystals. Considering the RNA like as the key component of primordial life by exhibiting both catalytic and genetic activities, I present hypothetical steps showing how the formation of precursors of this RNA like would be accomplished in montmorillonite. The distinguished structure of the later could help not only the formation of RNA like oligomers parallel to the surfaces sheets in the interlayer spaces, but also the fixing of some formed trimers to the edges of sheets of adjacent layers perpendicularly to their surfaces given that the length of the trimer is equal to the interlayer space during the wetted state. The encapsulation of each fixed trimer with an attracted oligomer in a fatty acid vesicle would be the start point of the formation of a pre codon-anticodon complex that could lead, since the beginning, to the emergence of a primitive genetic code.

Keywords – Man creation, Early life evolution, Montmorillonite clay, RNA oligomerization, Codon-anticodon complex, Genetic code.

Introduction

The holy Qur'an verses were verbally revealed by God "Allah" to the Prophet Muhammad (salla Allahou'alaihi wa sallam) through the angel Gabriel (Jibril) gradually over a period of approximately 23 years from 609 to 632 CE. The ancient explanations of verses related to the topic of man creation are very superficial and not accurate even at the language level and cannot be accepted particularly if we analyzed deeply all separate verses on this topic together. Thus on the basis of these ancient explanations it seems some discordance between scientific and religious thoughts. But in 2006, after a deep study, I have demonstrated that there is any contradiction between rigorous scientific results on man creation and corresponding holy Qur'an verses (Chaabani 2006). Among conclusions that I have deduced from eloquent terms presented in distinct abridgement of some Qur'an verses, the man creation had begun a very long period of time before his emergence. It had begun with the creation of his earthy and clayey nature in clay. Very likely "his earthy and clayey nature" means the basic structures of his bio-molecules that, common to all living beings, are formed from elements (ions) present in soil and clay. These explanations show that the objective of the creation of living beings is to reach that of humans. In addition, some other verses represent signs of what we designate currently the general or the central idea of biological evolution which is limited to the fact that all living beings on Earth share a common ancestor which had evolved from simple to complex forms. This general concept of evolution was clearly presented in many ancient writings of some Islamic Renaissance scholars in the wider Middle East particularly such as the last one "Ibn Khaldun" who had seen life at 1332-1406 (for review see Chaabani 2011, 2014). In fact this general idea of biological evolution is strongly supported by modern scientific data, but the different mechanisms of this evolution proposed firstly in a detailed theory by "Charles Darwin" in 1859, then by other evolutionists, cannot be proved or denied by the science. In this paper I will present a synthesis scenario, composed by scientific data previously published and new insights, showing that the clay could be the cradle of the creation of the first biological material that after about 3.5 milliards of years of evolution had led to the emergence of modern man.

Current state of scientific knowledge on the early life evolution

It is generally thought that the emergence of life on the Earth had begun with the appearance of specific conditions when liquid hydrocarbons and water could meet each other and interact during very long period until syntheses of bio-molecules became possible. These syntheses started with simple molecules such as amino acids or nucleotides had mainly resulted in the formation of macromolecules such as primordial informational polymers and those making up the compartmentalized structures. Then a correspondence between these macromolecules and structures was developed within a complex system, which possesses a mysterious synchronization leading to the emergence of the first pre-cell. However to go into details and explanations about how each of these general major stages had been occurred, numerous hypotheses, theoretical models and experimental studies were carried out, but until now major problems are far from being resolved. In this paper we will limit to two questions: where early life would be created and how the first genetic material would be formed?

As answer to the first question scientists have presented different hypotheses about the place where the origin of life occurred. According to them life originated at the ocean's edge, under frozen ocean, at deep-sea vents, deep in the earth's crust, or in clay. No one can confirm or deny each of these hypotheses. However in the present paper I will explain why I support the last one and why I prefer add the term "on" for speaking about the fact that early life might be created on and in clay.

As answer to the second question innumerable studies were carried out particularly those focused on the formation of the first potential genetic polymer. The stage of formation of primordial polymers of nucleic acids is linked to that of proteins in a controversy between nucleic acids-first and proteins-first scenarios, given that the replication of nucleic acids was dependent on protein enzymes and the synthesis of protein enzymes was dependent on nucleic acids. However, a primordial informational polymer of nucleic acids would be the earlier because (1) it could direct the synthesis of its complement from mononucleotides or short oligonucleotides, while no equivalent mechanism was known for the replication of a polypeptide (Woese 1967; Crick 1968; Orgel 1968), and (2) it was revealed for several small RNA that they are able to catalyze certain reactions of protein matrix synthesis and because of their enzymatic activity they are called ribozymes (Cech 1986). Hence, a primodial RNA like would be the most probable potential genetic polymer formed since the beginning of the early life evolution well before the formation of proteins and a DNA like evolved from it. Thus, an "RNA-World" hypothesis was advanced and developed (e. g., Gilbert 1986; Joyce 2002; Orgel 2004). Several more recent discoveries lend further support to the RNA world hypothesis by showing that RNA possesses a remarkable diversity of structural and metabolic functions. For example, I can quote the case of riboswitches recognized as important elements in the control of gene expression (Nahvi *et al.* 2002; Serganov and Nudler 2013), and the case of microRNAs (miRNAs), approximately 21-nucleotide-long, which are non-coding RNAs and considered as key post-transcriptional regulators of gene expression (e. g., Lagos-Quintana *et al.* 2004).

In the modern world, the polymerization of RNA monomers carries out from nucleoside 5' triphosphate monomers in the presence of DNA template and specific protein enzymes. But under prebiotic conditions and according to RNA-World hypothesis the first RNA polymers had been formed in absence of template and protein enzymes. To identify the pathways for this prebiotic formation many authors have hypothesized the involvement of mineral surfaces and showed that montmorilonite clay can catalyse the oligomerization of this RNA-World (Ferris *et al.* 1996; Huang and Ferris 2006; Joshi *et al.* 2009) and can help supplying a protocell-like compartment such as fatty acid vesicle or clay hydrogel environment (Hanczyc *et al.* 2003, 2007; Yang *et al.* 2013). In continuity with these findings, I will present in this paper complementary proposals showing how distinguished structure and properties of montmontrillonite clay would favorite the emergence of a preliminary background permitting RNAs like to function by triplet since the beginning, and therefore leading to an early development of a primitive genetic code.

Early Life might be created on Clay Bedrock

By dating the rocks in the ever-changing crust, as well as neighbors such as the moon and visiting meteorites, scientists supposed that Earth was formed about 4.54 billion years ago from the swirling dust and gas remnants of an old star's supernova explosion (Brent Dalrymple 2001). Soon after this formation, it is thought that warm oceans gradually formed, from steam escaping from the crust and from volcanic activity and icy meteorites and a primitive crust solidified and cooled to the point where liquid water could condense. Water began pooling into the first lakes, seas, and oceans. The interaction of water, heat, and rock set off the early life evolution that had lasted a very long time until the emergence of simple beings. In fact, the water is indispensable to any aspect of life from its first emergence until now because, among other things, in water many chemicals dissolve easily and therefore could be mixed together and reacted, because liquid water is the right temperature for chemical reactions to happen, and also because many chemicals have parts which are attracted to water and parts which are repelled by it. Hence syntheses of the first bio-molecules need very long period of a continual presence of water even in very slight amounts.

As I will explain in the following the oceans conditions do not agree with the scenario that I will propose and, consequently, I consider that early life evolution would be happened rather within Earth's surfaces and more precisely on saturated bedrock made up by layers of soil rich in clay. In fact, in presence of water the clay, particularly montmorillonite, can form a so-called "filter cake" and enable soil layers to become quite impermeable. This impermeable bedrock in harmony with particular environmental conditions can keep at least a minimum amount of water during periods when no new water arrives at the level of this bedrock.

RNA, the Key Component of Primordial Life, might be formed in Clay

There are three main groups of clay minerals: Kaolinite, Illite and Smectite. Clay rocks may contain a mixture of these minerals. The most common Smectite is Montmorillinite which is the main constituent of bentonite, derived by weathering of volcanic ash, and it is very likely to have been present on the Early Earth known by high levels of volcanic activity. Clay minerals have layer structures, each particle layer being comprised of two basic individual sheets. One, called a tetrahedral, is dominated by a plane of silicon atoms surrounded by oxygen atoms. The other is an aluminum/magnesium sheet known as octahedral sheet because each aluminum/magnesium atom and associated oxygens and hydroxyls comprise an eight sided building block or octahedron. Based on the number and arrangement of tetrahedral and octahedral sheets contained in the crystal units or layers, silicate clays are classified into two different groups: the 1:1 type minerals (one tetrahedral to one octahedral sheet) and the 2:1 type minerals (White 1987).

Montmorillonite belongs to the 2:1 type: each layer is made up of an octahedral sheet sandwiched between two tetrahedral sheets with shared apical oxygen atoms (Fig. 1). With no defects or substitutions, its chemical formula would be Al₄Si₈O₂₀(OH)₄. But usually there are other elements substituted for silicon and aluminium depending on their abundance when the montmorillonite was formed. The usual replacements elements are Fe^{2+} , Fe^{3+} , and Mg^{2+} in place of Al^{3+} in the octahedral layers and Al^{3+} for Si⁴⁺ in the tetrahedral layers (for review see White 1987; Liu and Zhang 2014). These substitutions, often non-equivalent (e.g. Mg for Al or Al for Si), lead to a net negative surface charge which can interact strongly by electrostatic forces with charge balancing cations. Owing to this peculiar structure, montmorillonite clay has several physicochemical properties such as a large surface area, high adsorption capacity, swelling and ion exchange. Many experimental studies were carried out for showing its key role in the early life evolution particularly during the formation of a pre-RNA World. In the following, I will try to put together some published results and conclusions with new complementary theoretical propositions in a synthesis scenario that I can present in three principal successive steps.

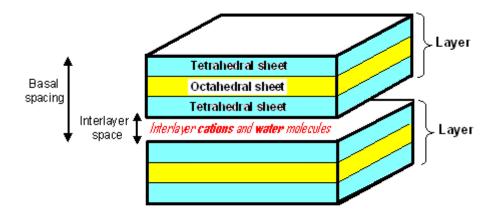


Fig. 1. Schematic illustration of montmorillionite clay mineral structure (2:1 type).

First Step: Oligomerization of RNA Nucleotides in Clay Interlayer Space

Montmrillionite clay can catalyze the formation of RNA oligomers in aqueous solution by joining activated mono RNA nucleotides to join together to form longer chains (Huang and Ferris 2006). The extent of catalysis depends on the magnitude of the negative charge on the montmorillonite lattice and the number of cations associated with it. Accordingly, in suitable experimental conditions, it is possible to obtain oligomers of 40 to 50-mers up to the length of small ribozymes (Joshi *et al.* 2009). In addition the sequence of RNA nucleotides so formed is not the results of a random synthetic process: (1) pyrimidine nucleotides elongate at a significantly slower rate than purine nucleotides, and (2) in presence of a mixture of right-handed (D) and left-handed (L) ribose, the formed dimmers are predominantly D-D or L-L, as opposed to mixed D-L (Joshi *et al.* 2000; Ferris 2005). In parallel to these experimental demonstrations on the possibility of the oligomreization of RNA nucleotides in montmorillonite clay, researchers are accomplished additional experiences aiming to find some details relating to the mechanism of this oligomerization.

As noted above the layer surfaces of montmorillonite are charged negatively. When wetted, the level of abundance of these charges and the amount of water and cations present in the environment permit a variable space between layers occupied by entering water and cations; while when dried, these interlayer spaces are reduced and occupied by partially hydrated cations which hold the layers together. Thus montmorillonite is able to expand and contract its structures while maintaining its crystallographic integrity. Thanks to this ability, RNA nucleotide oligomerization would occur mainly in the interlayer space parallel to surface sheets of montmorillonite (Fig. 2). The following explanations deduced from experimental results come to support this consideration:

- The expanded interlayer space is wide enough to accommodate polymers (Mazo *et al.* 2008).

- The negative charges of the layer surfaces of montmorillonite, in addition to their strong interaction by electrostatic forces with charge balancing cations (mainly Na^+ and Ca^{+2}) existing in the interlayer space, they bind with positively charged organics, while neutral and negatively charged organics may also be absorbed. In the case of nucleotides they may bind by van der Waals forces between the silicate layer of the

montmorillonite and the purine and pyrimidine bases of the nucleotides, when the pH is near 7 (Kawamura and Ferris 1999; Ertem and Ferris 2000).

- The activated monomers react to form oligomers when they are intercalated between the layers of the montmorillonite. Namely, the interlayer provides an environment where the activated monomers are proximate and can readily react to form phosphodiester bonds (Joshi *et al.* 2009).

- X-ray diffraction studies revealed changes in layer separations of montmorillonite as reaction occurs (Aldersly *et al.* 2011).

Accordingly, for a successful development of oligomerization process of RNA nucleotides in the interlayer space of montmorillionite clay, I propose that the interlayer space would be moderately expanded, namely in presence of relatively low amounts of water and cations (see Fig. 2). My proposal agrees with experimental results presented in the Joshi et al. paper (2009), and from which I could deduce the general following explanation: When cations (such as Na⁺) and water are quite present in the environment and enter in high amounts in the interlayer regions by the attraction of the high negative charge on montmorillionite surfaces, they prevent the activated monomer from entering and consequently block the oligomerization. Thus, among other conditions, the catalystic role of montmorillonite could be possible when montmorillionite has not a high negative charge on its layer surfaces or it has a high negative charge but, in its environment, cations and water were present in relatively low amounts. I opt for the second eventuality that does not agree with the ocean conditions but it could fit in with the continent prebiotic environment during the Archaean era (~ 3.5 billion years ago) at a more or less dry period but with a continual presence of a relatively low amount of water maintained, as I have already proposed, by a saturated clay bedrock.

In fact during the Archaean era the initial salinity of the oceans was slightly higher to the modern value (1.5 to 2 times) (Knauth 1998, 2005), the ancient continent would contain a non high amount of cations (slightly higher than that of the present continent) and variable amounts of water under continental alternating dry and wet periods that, as I will explain, suitable for the formation of RNA oligomers and their evolution. In addition, the relatively high level of temperature varying between 55 °C to 85 °C (Knauth 2005) or far cooler \leq 40 °C (Hren et al. 2009) during the Archaean era would not affect the interlayer space where oligomerization process occurs because the basal spacing of montmorillionite remains practically unchanged in the 20 - 100 °C temperature (Zhou *et al.* 1997).

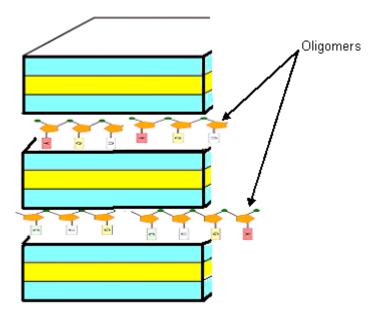


Fig. 2. Shematic representation of the formation of RNA oligomers in the interlayer space in presence of relatively low amount of cations and water molecules (first step)

The variable interlayer space is estimated indirectly through approximated values of the basal spacing which is the distance between similar faces of adjacent layers (in other words it is the thickness of each layer plus the interlayer space, see Fig. 1). The basal spacing could vary from about 0.98 to 1.8 + nm (e. g., White 1987) even to 2 nm (Brady and Weil 1996; Capkova *et al.* 1998). Exceptionally the basal spacing could continue to expanding more than 2 nm until the separation of layers. From these approximate estimations it would be reasonable to consider that layer thickness could be of about 0.98 nm while the interlayer space could reach 1.02 nm during the wetted state for given a basal spacing of 2 nm (Fig. 2). This situation concerns the second step that I can present as follows.

Second Step: Fixation of Formed Trimers Perpendicularly to Surface Sheets:

As noted above the formation of innumerable RNA oligomers occurred in interlayer space parallel to surface sheets of montmorillionite during a relatively dry period on a saturated clay bedrock, mainly in continual presence of a low amount of water. When the climate changes in rainy period, water with cations diffuse within the interlayer space until the space reaches about 1.02 nm (corresponding basal spacing of 2 nm) (Fig. 3). Consequently, RNA oligomers come off the layer surfaces under the pressure of diffused water and cations for leaving the interlayer space by the other side. During this exit some trimers, being in position perpendicularly to the surfaces sheets of adjacent layers, come to touch by their two extremities the break edge of these surfaces because the length of the trimers is equal to the interlayer space of 1.02 nm: if we consider that the distance between two neighboring bases in RNA is approximately the same of that in one strand of DNA double helice of 0.34 nm, the length of an RNA trimers is about 0.34 nm x 3 = 1.02 nm. As the edges of the surface sheets carry also charged sites (Sposito 1984), they could bind with each of the two extremities of the trimer (Fig. 3).

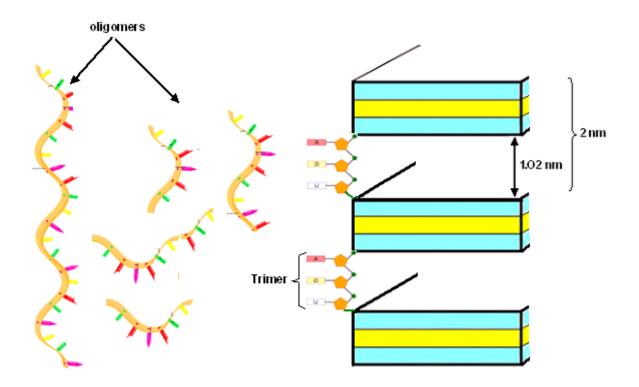


Fig. 3. Second step: Exit of formed oligomers from interlayer space and fixation of some trimers in position perpendicularly to the surfaces sheets of adjacent layers.

Thirst Step: Emergence of a Pre Codon - Anticodon Complex

According to the present scenario, the formed RNA oligomers are protect by the montmontmorillonite layers, but after their exit from the interlayer space they need a protection from some severe prebiotic conditions such as the relatively high temperature and the high amounts of UV and gamma ray radiations. For such protection, it was experimentally proved that montmorillionite clay can also catalysed the formation of closed vesicles from micelles composed of simple aliphatic carboxylic acids already present in the prebiotic environment and that particles of the clay and / or RNA oligomers could become encapsulated within these vesicles (Hanczyc et al. 2003, 2007). Thus, I propose that just at their exit from the interlayer space, in particular prebiotic conditions, RNA oligomers could be immediately encapsulated within micelles of fatty acid vesicles which provide protection and compartmented environments for further biochemical reactions (Fig. 4). Once formed, such vesicles can grow by incorporating fatty acid supplied as micelles and can divide without dilution of their contents by extrusion through small pores (Hanczyc et al. 2003). In addition these cell-like vesicles composed of fatty acids show enough permeability to nucleotides to allow nucleic acid elongation inside them (Mansy et al. 2008). They are also extremely thermostable and retain internal RNA and DNA oligonucleotides at temperatures ranging from 0°C to 100°C (Mansy and Szostak 2008).

As show in the Figure 4, each fixed trimer would be encapsulation with the nearer attracted oligomer in a vesicle where a pre codon-anticodon could emerged: the accompanying oligomer would be evolved in transfer RNA (tRNA) like by binding one of its complementary sequence (anticodon) to the fixed trimer considered as a codon and by the fulfillment of other events such as the appearance of hairpin and the addition of new nucleotides that could enter through the membrane of such vesicle that I designate codon-anticodon vesicle. However, each free oligomer would be encapsulated sole or with others in a vesicle where they could be elongated by binding together and / or by adding new penetrated nucleotides for working out possible ribozymes.

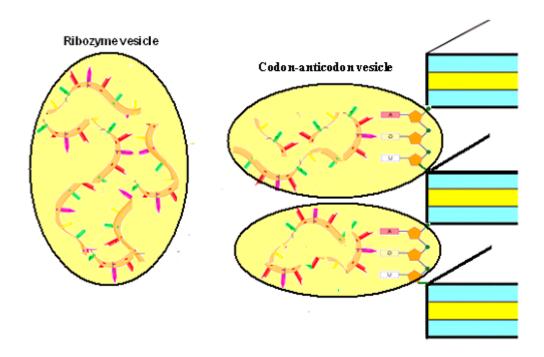


Fig. 4. Thirst step: emergence of pre codon-anticodon complexes in protective vesicles

After this wet period and at the beginning of a relatively less humid or dry period, the amount of water begin again to be low and consequently the montmorillonite interlayer space decrease until reaching the state of a possible resumption of the formation of new RNA oligomers. However during the decrease of interlayer space, under the movement of the break edges of successive layers each codon-anticodon vesicle comes off the edges of clay layers and finds itself, out the crystal clay, on contact with the vesicle above and that below. Hence, these vesicles would merge for permitting the connection between nucleotides of successive triplets. Consequently a long chain of codon-anticodon complexes could appear within a long unique vesicle. This chain would represent the first particular operational RNA already linked to the corresponding tRNAs like. The possible early emergence of a tRNA like is also advanced otherwise by some authors (e. g., Lacey and Staves 1990) who suggested that the tRNAs would be monophyletic having a single ancestor that gave origin to the diversity known today. In addition, a so early emergence of aminoacyl-tRNA synthetase enzymes was also suggested by other authors (Caetano-Anolles *et al.* 2013) who showed that the origin of the genetic code is tightly coupled to the history of aminoacyl-tRNA synthetase and their interactions with tRNA and the early emergence of the 'operational' RNA code. In the same way it was noted that the evolutionary pattern of diversification of tRNA may indicate how the chemical relationship between these molecules and aminoacyl-tRNA synthesases could have co-evolved to give origin to the system of correlation between anticodons and amino acids (Farias *et al.* 2014).

Finally I can note that my proposition on the possibility of the emergence of a background of a triplet code since the beginning agrees with the early timing estimated to the development of the genetic code. In fact, as the latter has remained invariant in all organisms on earth since the emergence of the first beings, it was demonstrated, from a statistical analysis of tRNA, that this universal genetic code cannot be older than 3.8 (\pm 0.6) billion years and thus is not older than, but almost as older, our planet (Eigen *et al.* 1989). This approximate estimation of the genetic code age permits to think to the possibility of an extraterrestrial origin of this genetic code that could be formed in riboorganisms which achieved a primitive form of life long before the Earth was formed (José et al. 2010). Any one can deny the possible extraterrestrial origin of some organic molecules such as some amino acids and purines already found in meteorites, but for the genetic code is difficult to opt for as much as no convincing sign was found. On the other hand I think that this approximate age of genetic code of about 3.8 billion years is not in discordance with the possibility of its emergence on Earth. In fact the latter, formed at about 4.54 billion years ago, has a habitable environment from about 4.3 billion years ago and the age of the ancient discovered rock including simple beings is about 3.5 billion years ago (Hofmann et al. 1999). Hence, the early life evolution surely started well before this date, and the date of about 3.8 billion years ago could coincide with that of the first stage of early life evolution because this evolution had lasted a hung tract of time for leading to the formation of first cell and because rocks of such great antiquity are rare, and possible rocks with earlier fossils have not yet been

discovered. However, the age of genetic code so estimated agrees with my proposition that the background of genetic code by triplet was formed since the beginning.

Conclusion

Several problems related to the origin of life are not yet resolved particularly the fact that we cannot provide plausible conditions and mechanisms for the pre-biotic formation of the different elements involved in the early life evolution. However some observations such as those of the involvement of clay minerals in the polymerization of bio-molecules on early Earth would be promising for a better research progress. These observations agree with the conclusion, deduced from Qur'an that the man creation had begun a very long period of time before his emergence and it had begun with the creation of his earthy and clayey nature in clay. In this paper I have presented a hypothetical synthesis scenario in which dispersed published data were brought together with new complementary insights for showing how montmorillonite clay could be the cradle of the creation of the first biological material that after about 3.5 milliards years of evolution had led to the emergence of modern man. First, opting for an early life evolution within Earth's surfaces, I have proposed that the continual presence of a minimum amount of water indispensable for chemical reactions could be possible if early life has originated on saturated clay bedrock. Second, I consider that the formation of the key component of primordial life, RNA like, would be accomplished in montmorillonite clay. In fact the interlayer space of the latter represents the ideal place for the oligomerization of RNA nucleotides by favoring their binding as they found in its 2-dimensional crowded parallel to the layer sheets and by favoring the protection of formed RNA oligomers from severe prebiotic environment conditions. In addition, in suitable conditions the formed RNA oligomers leave the interlayer space and during their exit some trimers, changing their position perpendicularly to surfaces sheets, bind each of their two extremities with the edges of sheets given that the length of the trimer is equal to the interlayer space during the wetted state. The possible encapsulation of each fixed trimer with the nearer attracted oligomer in a vesicle would represent the start point of the emergence of a preliminary background of a triplet code. This agrees with the early timing estimated to the development of the genetic code. The present hypothetic synthesis scenario would open up new tracks for future experimental research works that could check at least some of the present new proposals.

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