

Letter to the Editor

## Borophosphates and silicophosphates as plausible contributors to the emergence of life



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### ABSTRACT

Scientific explanations for the origin of life are incomplete and may differ on some issues. Here, we argue that some prebiological steps have occurred in environments with borophosphates and/or silicophosphates in the form of hydrogels, on the basis of their chemical groups and structural properties. These could have decreased the diffusion rate of some prebiotic molecules, stabilized molecules with vicinal *cis*-diol groups, reduced the hydrolytic activity of water and inserted catalytic metal ions into their networks. Additionally, these hydrogels could have acted as reaction media, supplied a phosphate source for phosphorylations and produced crystals that may have permitted enantiomeric enrichment of prebiotic molecules, thus providing conditions for the emergence of protocells.

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### 1. Introduction

The biological nature of both nucleic acids and proteins gives little information about which one was crucial for the origin of life because each needs the other for the *modus operandi* of the current cells. Thus, nucleic acids store genetic information for the protein bioproduction, and the biosynthesis of nucleic acids as well as their stability depend of proteins as enzymes.

The discovery of RNA's (ribonucleic acid) biocatalytic ability as ribozyme led to the RNA world hypothesis as an explanation for the origin of life [1]. However, abiogenic syntheses of both RNA and its monomers (ribonucleotides) were not yet accomplished, although some ribonucleosides precursors have been synthesized under postulated prebiological conditions [2].

The physical and chemical characteristics (including abundance and availability of the starting materials) of the sites where primordial reactions took place is limited; thus, a wide range of options can be considered for the prebiotic syntheses, such as media to carry out reactions [2] and hydrothermal environments that are suitable for availability of reactants and range of temperatures [3].

On the other hand, explanations on the emergence of the homochirality and the prebiological phosphorylations have discrepancies within the prebiotic chemistry research community. Now we propose hydrogels (and their derived chiral minerals) of

borophosphates and/or silicophosphates and ribose (or related compounds) as prebiological drivers for those prebiotic processes.

### 2. Borophosphates and silicophosphates

Several minerals with borate, silicate or phosphate are known [4], and some borophosphates [5] and silicophosphates [6–10] have been characterized. Provided that plate tectonics was active in the Hadean and Archean, relatively high concentrations of borate are likely to have been available in the ocean system of the early Earth [11].

On the other hand, non-glass borophosphates and silicophosphates containing relatively abundant chemical elements have been obtained by different synthetic procedures, including from gels [12–19], with their crystallization being determined by reagents' ratio [12,15].

The diversity of borophosphates and silicophosphates supports the possibility that they could have occurred under prebiotic environments and perhaps participating in prebiological reactions, including as hydrogels.

### 3. Hydrogels

Gels are three-dimensional structures that can be obtained from different solvents. They display a wide variety of characteristics, such as properties of both solids and liquids, and have a large number of applications [20].

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In particular, hydrogels are three-dimensional cross-linked networks dispersed in aqueous media [21]. Their properties are dependent on diverse structural arrangements of water [20].

Natural hydrogels can be formed in environments containing rocks and water, and they can be produced from montmorillonite (a smectite [22]), hydrotalcite [23] and other inorganic materials [24]. On the other hand, inorganic hydrogels derived from both silicate and sulfuric acid were identified [25]. Other inorganic hydrogels, by addition/replacement of borate and/or phosphoric acid, can probably be obtained through similar procedures. Note that some borophosphates and silicophosphates have been achieved from gels [12,13,15–18]. Interaction between hydrogels and minerals could have happened during the prebiological evolution.

Ribose is the component that determines the chiral properties of RNA, as well as the phosphorylated module of this nucleic acid. These could have constrained its involvement to the prebiotic chemistry.

#### 4. Ribose

Ribose is widely distributed in all organisms, as a basis of numerous biomolecules, indicating its remote origin. Its abiotic synthesis by the formose cycle has been known for over one and a half century [26]. However, a low yield is obtained; only long-term accumulation, by increasing its stability, can overcome this constraint [2]. This has led to the proposition that other sugars have preceded ribose in the early stages of life, what is not supported by experimental evidence [27]. Surprisingly, ribose has advantages over other pentoses and also hexoses as components of polynucleotides. This is because it has a significantly lower number of inadequate chemical interactions than other sugars [28], which suggests its prebiotic selection.

Ribose occurs in living organisms with a specific conformer and also a particular enantiomer. Some ions can induce additional stability and specific conformations at this pentose [29,30].

#### 5. Stabilization and induced conformations of ribose

Borate may be critical to the formose reaction. The ribose-borate bound species protect some hydroxyl groups of ribose from side reactions, as well as favor its furanose isomers, see Fig. 1 (pyranose is the predominant in aqueous solutions), i.e. the single conformer of the pentose derivatives in all organisms [31–34] induced by the electrostatic field of borate [35]. Borate stability of this pentose is more efficient than to other aldopentoses [36]. Furthermore, this ribose stabilization is within a reasonable range of temperatures, as well as in the presence of the most abundant alkaline and alkaline-earth cations. These are feasible for borate, but not to other abundant anions in the current oceans [34,37]. Moreover, boron is essential for several organisms, usually associated with sugars [38]. All of these conditions seem to point to its prebiological chemistry contribution related to ribose.

Silicon is much more abundant than boron in the Earth's crust and is essential for several living organisms [38]. Silicate also promotes monosaccharide synthesis by the formose reaction, as well as the ribose stabilization, see Fig. 2 [39–42]. Silicon, as silicate, can also have been a prebiological chemical element.

Other metal ions or complexes have been applied to improve the ribose synthesis, such as  $Pb^{2+}$  [43],  $MoO_4^{2-}$  [44] and Zn-pyrolone [45]. However, lead is not a biological metal, and both molybdenum, as molybdate, and zinc availabilities were limited on the early Earth [38], indicating their absence or negligible involvement in the prebiological chemistry.

Other ribose-related compounds could have been prebiotic molecules, such as its derivative with cyanamide, proposed as

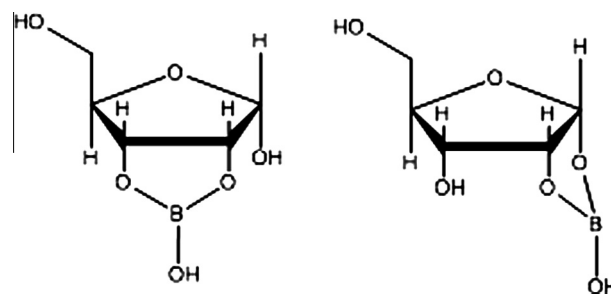


Fig. 1. Examples of ribose-borate bound species.

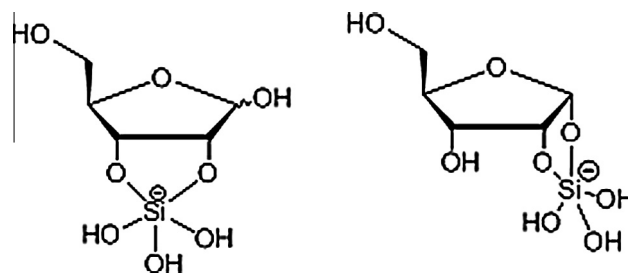


Fig. 2. Examples of ribose-silicate bound species.

starting material to prepare ribonucleoside precursors [46]. Its ribose product is a very stable bicyclic compound with the pentose framework as furanosyl [47], i.e. similar to current ribose-related biomolecules. Hence, interactions between ribose and hydrogels mentioned in this article can be found in other ribose derivatives.

If hydrogels from borate and/or silicate and phosphate have oxygen-boron bonds without all of the oxygen atoms linked to the phosphorus atoms, as some borophosphates [16,19], hence oxygen-free atoms can interact with ribose (or other molecules with vicinal *cis*-diol groups) similarly to the inorganic borate, thus increasing its stability and inducing it as furanose, see Fig. 3 and compare with Fig. 1.

#### 6. Homochirality in life and its emergence

Homochirality in both amino acids and sugars is prevalent in all terrestrial life [48,49] and it has been proposed that its origin is connected with the emergence of life [50].

Current hypotheses about the advent of the homochirality are typically related to amino acids, such as induced by polarized radiation [51] or by parity violating weak interaction [52]. Revision papers based on physical and chemical criteria [48], chiral symmetry-breaking processes using mathematical models [53] and from extraterrestrial origin [54] have been published.

Chirality in synthesis takes place by participation of chiral molecules usually as catalysts, solvents or reagents. Indeed, in their absence, an equal amount of enantiomers, i.e., a racemic mixture, are attained. However, chirality can also be associated with the crystal structure or space group of minerals and may be independent of their molecular composition [55]; this occurs more commonly than is generally recognized [56].

Racemic mixtures in aqueous boiling solutions (i.e. related to hydrothermal conditions) can be induced toward a single chiral solid phase, as was achieved from sodium chlorate racemic mixture [57], despite this salt must have been absent in the reducing environment of the early Earth.

On the other hand, alterations in composition or temperature of the hydrogels can lead to phase transitions with crystalline

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