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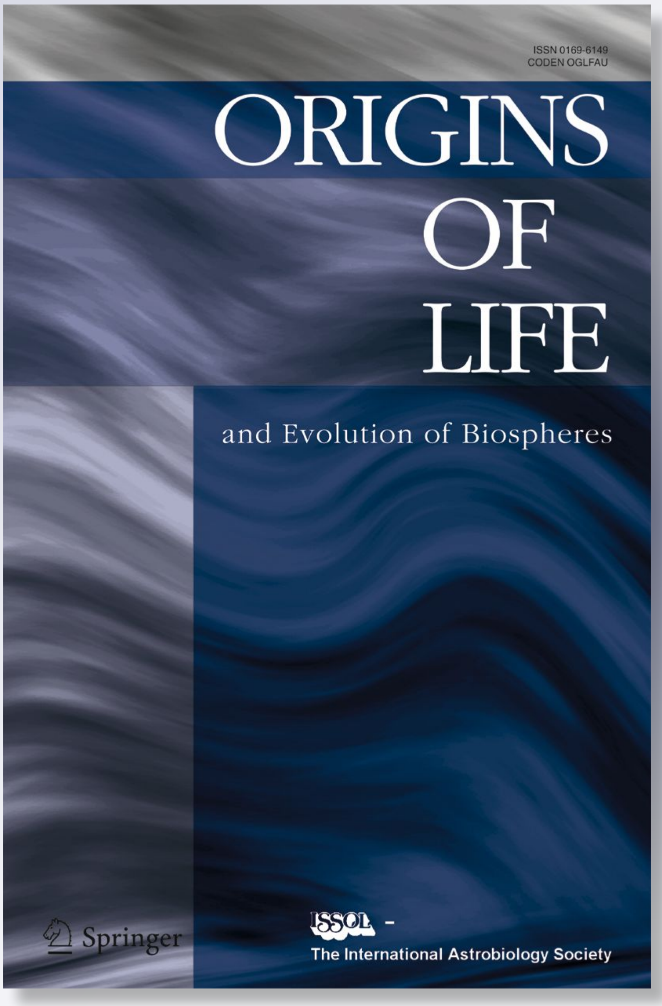

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PREBIOTIC CHEMISTRY

Hypothesis: Origin of Life in Deep-Reaching Tectonic Faults

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Abstract The worldwide discussion on the origin of life encounters difficulties when it comes to estimate the conditions of the early earth and to define plausible environments for the development of the first complex organic molecules. Until now, the role of the earth's crust has been more or less ignored. In our opinion, deep-reaching open, interconnected tectonic fault systems may provide possible reaction habitats ranging from nano- to centimetre and even larger dimensions for the formation of prebiotic molecules. In addition to the presence of all necessary raw materials including phosphate, as well as variable pressure and temperature conditions, we suggest that supercritical CO_2 as a nonpolar solvent could have played an important role. A hypothetical model for the origin of life is proposed which will be used to design crucial experiments for the model's verification. Because all proposed processes could still occur in tectonic faults at the present time, it may be possible to detect and analyse the formation of prebiotic molecules in order to assess the validity of the proposed hypothesis.

Keywords Origin · Life · Strike-slip faults · $scCO_2$ · Prebiotic molecules

Introduction

A large variety of proposals and models for the origin of life have been proposed (Schopf 2002; Davies 2003; Bada 2004; Botta 2004; Luisi 2006; Rauchfuß 2008). Newer investigations

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indicate the existence of submarine hydrothermal vents, which, compared to black smokers, have the advantage of lower temperatures (Lost City Hydrothermal Field) (Martin and Russel 2007; Martin et al. 2008). In addition, recent studies support the view that a group of universal proteins that was active in relatively acidic conditions was also present in hot environments (Perez-Jimenez et al. 2011). Based on these findings, we present a model for the origin of life in open tectonic fault systems (mainly strike-slip faults) serving as possible geochemically reactive habitats within the earliest structures of the ancient continental crust.

Up to the present day, strike-slip faults may provide reactive habitats with dimensions ranging from nano- to centimetres and above which amount to several cubic kilometres in sum. They are filled mainly with supercritical and subcritical high-salinity waters and supercritical and subcritical gases. Here, all the elements necessary for the development of prebiotic molecules exist in variable concentrations and quantities. Furthermore, they also possess periodically changing pressure and temperature conditions, varying pH values, metallic surfaces, clay minerals and a large number of catalysts. While cosmic and UV radiation play no role, nuclear radiation may affect the chemical evolution of the molecules inside the crust. Carbon dioxide (CO_2) is of crucial importance. It can be present in an almost pure form as a supercritical medium ($scCO_2$) at a crustal depth of less than 1 km (Kaszuba et al. 2006). On the surface, gases with high CO_2 concentrations are found in the mofettes of many volcanic regions (Vogtland, Czech-German border zone (Koch et al. 2008) or Yellowstone) on continents. Extrapolation of data on CO_2 flux at Yellowstone, for instance, imply that this region is one of the earth's most productive CO_2 sources, releasing 45,000 tons per day (Werner and Brantley 2003). For the early earth, a large degree of CO_2 degassing as a consequence of rapid accretion has been postulated (Walker 1985).

Inside strike-slip faults, a two-phase system formed by supercritical CO_2 in liquid water could provide a possible environment for different condensation and polymerisation reactions. As an example, an alkene dissolved in $scCO_2$ could react with CO_2 and NH_3 (via an electrophilic attack of CO_2 towards the π-bond assisted by protonation and followed by a reaction of the resulting carbenium ion with NH_3). This reaction leads to the formation of a beta amino acid, a compound which will be less soluble in $scCO_2$ and therefore is likely to be accumulated in an adjacent aqueous phase. Inside the earliest cratons, extreme earth tides may have played an important role for cyclic variations within the fluid-water interface and for the development of gradients. Based on these conditions, evolutionary steps in which prebiotic molecules were condensed to long-chained molecules could have taken place in the early continental crust.

The Continental Crust of the Early Earth

At a certain time on the early earth, cratons existed that were composed of more SiO_2-rich rocks compared to the komatiitic-basaltic crust. Good examples for this are the 4 Ga Acasta Gneisses from the Slave Province in NW Canada, the oldest known rocks on Earth (Bowring and Williams 1999). The oldest minerals of up to 4.4 Ga are detrital zircons from the Jack Hills area of Western Australia (Wilde et al. 2001; Harrison et al. 2008). They contain quartz, feldspar, muscovite and monazite inclusions that are typical for the continental crust. These geochemically evolved rocks have been present for at least 4.4 Ga and therefore indicate crust formation.

During the formation of cratons, vertical strike-slip faults must have been formed simultaneously due to variable stress conditions. At these faults, jerky fissures or creeping movements took place under a critical rise of tension. These processes, still occurring up to this day, produced open channels reaching deep into the mantle and permitting a variety of gases to ascend at the faces of the blocks. The gases could have been present more or less as pure phases (mainly CO_2) or as mixtures of CO_2, N_2, water vapour, SO_2/H_2S, hydrogen, CO, NH_3 and trace gases. Different acids (sulphuric acid, hydrochloric acid, hydrofluoric acid) ensured a low to very low pH value. Solutions covered a wide temperature and pressure range, spanning from subcritical to supercritical conditions. The low pH values caused some rock-forming minerals to be decomposed or transferred into aluminium-rich secondary minerals, thus providing reactive surfaces. Inside the vertical fault zones, hydrothermal activity led to a high degree of mineralisation by quartz and ores, mostly at lower depths. These are sulphidic Pb-Cu-Zn ores comparable with younger formations. In addition, all other metals - in reduced state - could have been part of this process together with sulphur, phosphate anions, halides and radioactive elements. In strike-slip faults, material flows are substantially lower than in black smokers. Strike-slip faults also show significantly lower temperatures in the upper crustal zones. Basaltic dykes and grabbroidic intrusions are parts of all proto-continents. The phosphate that is bound in apatite can occur in up to 0.5 wt%; the alkali-rich substitutes contain P_2O_5 in concentrations of between 1 and 2 wt% (Bergmann 1987). Apatite dissolves in acidic water and is an important source of phosphate for prebiotic chemistry. In fumaroles emerging through apatite-bearing basalts, pyrophosphate and tripolyphosphate are apparent (Yamagata et al. 1991).

Characteristics of Strike-Slip Faults

Inside strike-slip faults, material flows from the depths of the earth to its surface depend on temperature gradients and ascending gases and continuously decreasing pressure. A sudden pressure decrease occurs when tectonic activity opens up new faults. As a result, areas of vacuum that are very rapidly filled by fluids or gases could have occurred for a short time. This rapid expansion process is expected to promote condensation and poly-condensation reactions by rapidly removing water from the original chemical equilibrium. In the high-pressure situation, all reactants are highly concentrated. Equilibria with intermediate products such as oligomers are quickly formed. The expansion process leads to the evaporation of water (which is thus removed from the equilibria), while the non-volatile reaction products remain in the liquid or solid state. On account of the minimal water concentration at this stage, hydrolytic reactions are blocked. This process is somewhat comparable to the technical procedure of spray drying and could potentially support condensation reactions which otherwise are difficult to imagine under ambient conditions.

Strike-slip faults that are open over extended periods are in most cases covered by quartz dykes. If quartz is squeezed by crustal movement, piezoelectric currents are triggered. The combination of the arising areas of vacuum which are almost immediately filling with gas together with strong local electrical currents make gas discharge scenarios appear plausible. This is reminiscent of the Miller-Urey synthesis conducted in 1953 (Miller 1953). Furthermore, earth tides which had been many times stronger than today may have affected processes in fault systems by inducing periodic level changes. Groundwater fluctuations dependent on earth tides can still be detected in wells today (Kümpel 1997).

Proven Conditions for the Formation of Prebiotic Molecules

In our opinion, the depiction of the tectonic environment indicates that the basic conditions for the formation of prebiotic molecules in terms of the starting substances, physicochemical conditions, periodic change and spatial compartimentation could have been met inside the tectonically sheared earth's crust.

Conditions for the formation of prebiotic molecules verified in laboratory experiments could well be transferred to real conditions inside the earth's crust. Fujioka et al. (2009) showed the potential for amino acid synthesis as well as oligomerisation reactions based on a nitrogen source and keto acids in $scCO_2$. Additional examples are given by Rauchfuß (2008), but there have been many difficulties in transferring laboratory results to previous models on the origin of life. Fundamental problems exist regarding the concentrations of the reactants as well as concerning the availability of catalysts. The availability of Pb^{2+} ions is of particular importance as had been shown in the discussion on the formation of ribose (Zubay 1998). Furthermore, boric acid influences the stability of ribose (Ricardo et al. 2004). Strike-slip faults could provide both, high lead availability leading to the formation of lead ore dykes, as well as the occurrence of boric acid or other boron minerals. Chemical solution of boron-bearing minerals within fault zones indicates considerable concentrations of boron compounds. Boron-bearing minerals are closely linked to highly differentiated granitic melts, as the oldest zircons indicate for parts of the crust (see above). Granitic melts might have played an important role in the early stage of the formation of the earth's crust as can be seen on the example of Iceland. Here, after formation of this island, rhyolitic/granitic melts were formed in a purely basaltic environment within a few million years (Oskarsson et al. 1982).

However, in all previous studies the fundamental question remained. How have larger organic molecules, e.g. the products of the polycondensation of α-amino acids or monosaccharides, evolved in an aqueous solution? Matthews' (2000) brief explanation of this as yet unanswered question referred to the absence of the prerequisites - an anhydrous state, high temperatures and an acidic environment – on the Earth's surface.

Our model is intended to propose a potential alternative environment for these reactions.

The Model

Overall View

Based on present knowledge, one can assume that supercritical CO_2, ascending from the upper mantle, was present in the upper crust of the early earth below a depth of approximately 1000 m (critical point of pure CO_2: 74 bar; 31°C). Above this level, CO_2 is expected to be in a subcritical state.

Supercritical CO_2 is a non-polar solvent widely used in "green chemistry" (Anastas and Kirchhoff 2002). Supercritical CO_2 is a suitable solvent for non-polar reactants and allows for chemical reactions which normally occur in the absence of water. Under the influence of periodically changing conditions, possible polar reaction products could be transferred into a neighbouring aqueous environment.

In the following, we present a hypothetical profile of the crust which is the basis for the discussed model (Figs. 1 and 2). The crust is approximately 20 km thick. The temperature gradient is assumed to be 40°C/km for cooler areas, but locally it could have been twice as high. This hypothetical crust profile could have developed after formation of the first proto-continents.

The profile can be divided into three sections showing different phase states of the media which could have been present in the faults. At its lowest level, below 7 to 9 km, H_2O (depending on the salt content) and CO_2 (depending on the purity of the gas) are supercritical (scH_2O, $scCO_2$). Above this level, H_2O is transferred to the subcritical state (labelled as

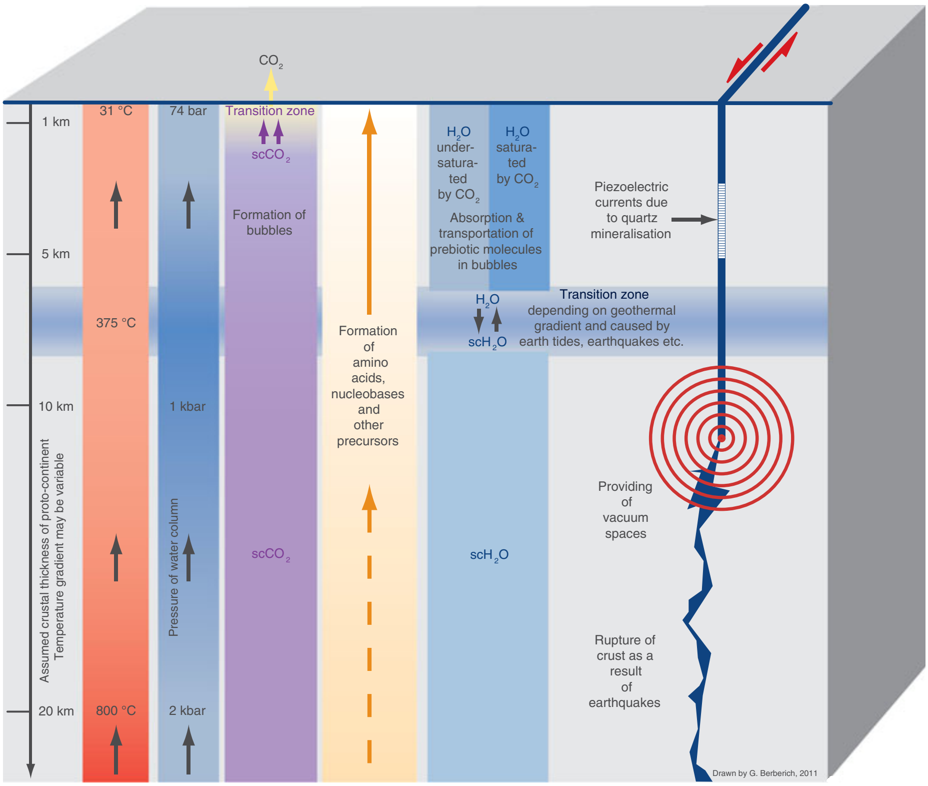

Fig. 1 Parameters of a hypothetical crust profile with stability fields of H_2O and CO_2 in different phase states and possible areas for the formation of prebiotic molecules.

"H_2O" in Fig. 1), while CO_2 remains supercritical just below the 1 km level. Above the 1 km level up to the surface, both substances coexist in a subcritical phase.

Changing material flows and different supplies of single components may cause variations in the ratio of CO_2 and H_2O. It is assumed that, depending on temperature and the availability of necessary chemical components, prebiotic molecules are formed at different levels in the crust. Essential catalysts and siliceous and metallic surfaces (pyrite) are assumed to be present in any variations. All additional tectonic activity inside the fault zones would promote the conditions for $scCO_2$ to ascend from the mantle into the crust. $ScCO_2$ could extract non-polar prebiotic molecules from subcritical H_2O and transport them to the upper levels.

scH_2O/H_2O Transition Zone

In the scH_2O/H_2O transition zone, we assume that the formation of $scCO_2$ bubbles takes place (Oparin et al. 2005). Alternating supply of ascending carbon dioxide and strong earth tides stimulate mixing in this transition zone. At this position, we believe that two possibilities are given for the further development of the bubbles: while rising to the surface, the bubbles can either merge into a CO_2-undersaturated or into a CO_2-saturated aqueous phase.

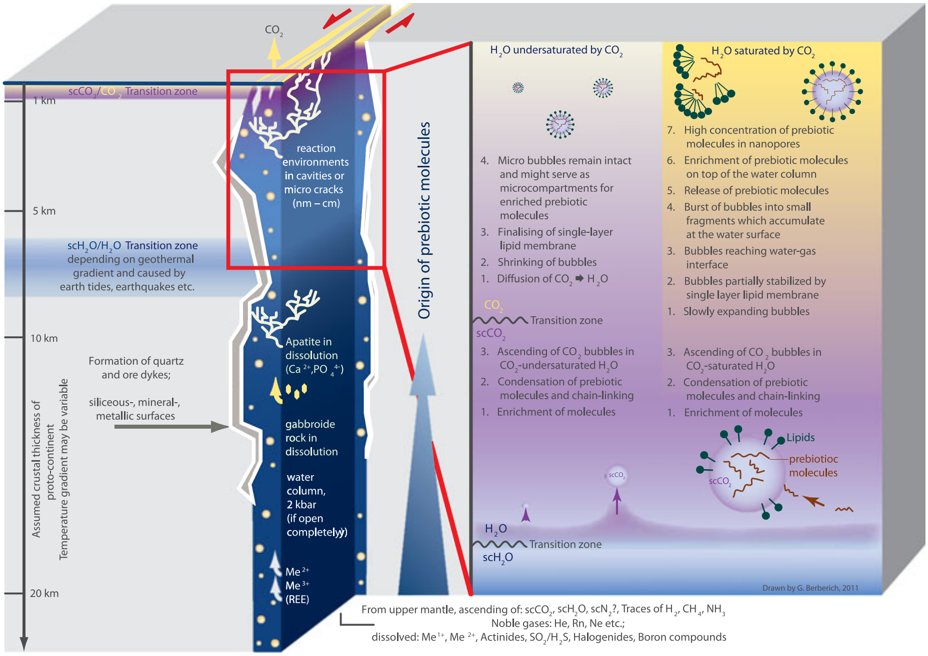

Fig. 2 Detailed section of the crust profile relating to the postulated development of processes leading to the enrichment of prebiotic molecules

If $scCO_2$ bubbles enter CO_2-undersaturated water, CO_2 will diffuse out of the bubbles while they ascend, causing them to shrink gradually. If the bubbles rise in a CO_2-saturated water column, their diameter will be enlarged continuously with the decreasing pressure on entering the subcritical zone.

Depending on the starting situation, each bubble of $scCO_2$ may contain a wide variety of nonpolar molecules. Presumably, the bubbles are also enriched by the uptake of further molecules of low or moderate polarity from the subcritical water column. Inside the bubbles, an accumulation of amphiphilic organic components (e.g. lipids) at the phase boundary with the subcritical water is possible. Lipids arranged with their hydrophilic ends along the outer wall of the bubbles could form a closed single-layer membrane as the bubbles shrink on account of the CO_2 diffusion into the surrounding water. At this stage, long-lasting compartments may form which could be regarded as preliminary cell structures. Inside these compartments, all hydrophobic and therefore less water-insoluble components are collected and concentrated. In this process, potential reaction chambers are created in which organic chemical reactions usually occurring in non-polar solvents are possible, including even the condensation of molecules forming longer chains. Condensation reactions leading to long-chained molecules within the $scCO_2$ bubbles may depend on the variation of concentration, temperature and pressure connected to the process of ascending. If complete dissolution of CO_2 in water occurs, the molecules trapped within the lipid membranes form an initial highly concentrated assembly of prebiotic substances.

When the bubbles reach the top of the CO_2-saturated water column, they collapse and thereby cause the contents of the bubble to be sprayed over the surface of the water and the surrounding area. The substances originally dissolved in or associated to the bubbles, e.g. long-chained molecules formed by polycondensation or single layer membrane fragments, now accumulate at the water surface. They thus could be available for reactions in the upper levels of fault systems that provide low-temperature reaction chambers. Here, the fragments could further be concentrated in small capillary spaces and enclosed by lipids.

Top of the Water Column-Cyclicality

Under the conditions of the early earth, tidal fluctuations of the water level caused the liquid content of the column to undergo a vertical motion of several metres. Hence, one can expect very efficient mixing of all the constituents near the rock surface of the fault. This also applies to the interface between water and the gas atmosphere at the top of the column. A seam area would have formed induced by the alternating dry and wet periods connected to the tidal fluctuations. In addition, CO_2 geyser effects may take place, leading to an ejection of liquid and gaseous material to the surface.

We believe that, within the proposed scenario, all conditions for selective evolution of the first biomolecules and even of cells are met. The further evolution of prebotic processes could have been induced by the cyclicality which was supplied by strong tidal fluctuations.

Consequences

In the presented model, we assume that a continuous production of prebiotic molecules has been taking place on a large scale. From the numerous molecules which may have been formed, the most stable compounds could have been selected over geological periods, leading to the development of a still endless variety of proto-cells. Following chemical evolution, biological evolution began with the formation of self-replicating molecules together with cell structures.

We also propose that the described conditions still exist today within some parts of the continental crust. Hence we conclude that all described processes should occur in a similar way in tectonic faults up to this day. In this case, the only reason why those processes are no longer easily detected on the earth's surface should be the activity of the biosphere: since present biological activities either disintegrate or reproduce the most indicative organic compounds, it is very difficult to find a proof on the surface. Nevertheless, inside tectonic faults it should be possible to detect and analyse the formation of prebiotic molecules in order to verify the validity of the proposed hypothesis.

A clear indication of the geological provenance of the corresponding organic substances would be provided if potentially chiral molecules can be shown to be present in racemic mixtures, making them distinguishable from similar molecules of biological origin. Furthermore, their isotopic composition can help to rule out a possible biological origin. In addition to deep drillings using specific drilling fluids, the analysis of fluid inclusions of quartz dykes in former deep crustal regions are another possible approach. Single steps of the proposed formation of biomolecules can also be reproduced and analysed under $scCO_2$ conditions in high-pressure installations. In any event, detecting traces of advanced prebiotic organic chemistry would be one of the most thrilling scientific discoveries in the search for the origin of biological life.

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