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Astrobiology in Europe, 20 years of expectations

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Review Article

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Abstract

During 20 years, the European astrobiologists collaborated within EANA, the European Astrobiology Network Association, to help European researchers developing astrobiology programmes to share their knowledge, to foster their cooperation, to attract young scientists to this quickly evolving interactive field of research, and to explain astrobiology to the public at large. The experiment of Stanley Miller in 1953 launched the ambitious hope that chemists would be able to shed light on the origins of life by recreating a simple life form in a test tube. However, the dream has not yet been accomplished, despite the great volume of effort and innovation put forward by the scientific community.

Quite different scenarios and routes, often in competition, have been followed and tested in the laboratory to explain the origin of life, thus opening many questions:

- Organic life versus mineral life?
- Home-made organics versus extra-terrestrial delivery?
- Organics from atmospheric CO_2 or CH_4 or from hydrothermal systems?
- Warm little pond or submarine vent environment?
- Primeval soup or metabolism first for the inception of life?
- Bulk chemistry or chemistry on the rocks?
- Unique event or plurality?
- Reasoned chemistry or messy alchemy-type approach?
- Chance mechanism versus determinate mechanism for the origin of homochirality?

Life emerged on Earth when parts of chemical automata self-assembled to generate automata capable of both self-reproduction and evolution. Sometimes, a minor error during this process generated more efficient automata, which became the dominant entities. Like Luigi Pirandello in his '*Six Characters in Search of an Author*', chemists are in search of the ingredients (the actors), the environment (the scene) and the most plausible scenarios for a start of life on Earth, about 4 billion years ago.

Six characters in search of a scenario

1. The first chemical automata most likely emerged in liquid water. According to its molecular weight, water should be a gas under standard terrestrial conditions. Its liquid state is due to its ability to form hydrogen bonds, which also makes water a good solvent. Water is essential in the production of clays and other weathering products of minerals in rock, which would have provided a continual supply of nutrients for life (Westall and Brack, 2018). The earliest environments in which chemical automata developed were locations in which wet–dry cycling conditions prevailed, thereby helping to drive the types of prebiotic chemical reactions required for the emergence of life as we know it (Forsythe *et al.*, 2015; Becker *et al.*, 2019; Campbell *et al.*, 2019; Damer and Deamer, 2020).
2. It is generally believed that the essential components of primitive chemical automata were organic molecules, i.e., molecules containing carbon and hydrogen atoms associated with oxygen, nitrogen and sulphur atoms, as known in present-day life. With the exception of the small amounts of organic molecules formed in the primitive atmosphere, the majority of carbon molecules on the early Earth were either delivered by carbonaceous chondrites (Pizzarello and Shock, 2010), micrometeorites (Maurette, 2006; Rojas *et al.*, 2021), and cometary material (Altwegg *et al.*, 2016), or formed in the subsurface by processes catalysed by aqueous reactions on the surfaces of subsurface rocks (Martin *et al.*, 2008; Westall *et al.*, 2018).
3. Rock and minerals constituting the earliest environments would have played an essential role in the processes that led to the emergence of life (Hazen and Sverjensky, 2010).

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[1] André Brack co-founded EANA with Beda Hoffmann, Gerda Horneck and David Wynn-Williams (Brack *et al.*, 2001). The network was formalized in Spring 2001 during the First European Workshop on Exo/Astobiology held in Frascati, Italy.

Pumice is an example of a rock type that has been promoted as a unique substrate with remarkable potential to support the origin of life (Brasier *et al.*, 2011). Clays also offer advantageous features as key minerals in origin of life scenarios due to their (1) molecular order and repeating topological arrangement with the ability to serve as polymerization templates, (2) large adsorption capacity with the ability to concentrate organic chemicals, and (3) shielding capacity to protect templated organic molecules against sunlight and other types of incoming radiation (Ertem, 2021). The confinement of several types of organic molecules essential for life in silica gel were likely also necessary for the earliest life (Gorrell *et al.*, 2017; Dass *et al.*, 2018).

4. The environmental 'stage'.

Any plausible model for the origin of life must take into account the geological complexity and diversity of the primitive Earth. Liquid water was present at Earth's surface due to the size of the planet, its distance from the Sun, and the greenhouse atmosphere maintained by crustal recycling as a result of plate tectonics and volcanism that continuously released water and key atmospheric gases important for life, such as CO_2 trapped in subducted carbonates. Other environmental conditions on the ancient Earth were very different from those of today: anoxygenic (Schopf *et al.*, 2017), a warm to hot and acidic ocean; a high flux of ultraviolet (UV) radiation (Ranjan and Sasselov, 2017); and a great amount of volcanic and hydrothermal activity (Nisbet and Sleep, 2001; Westall *et al.*, 2018). Accordingly, hydrothermal environments at Earth's surface have gained renewed interest as possible cradles for the origin of life (Sojo *et al.*, 2016; Branscomb and Russell, 2018; Westall *et al.*, 2018; Deamer *et al.*, 2019; Damer and Deamer, 2020; White *et al.*, 2020).

5. Far from equilibrium wet–dry cycling reactants

One can envision a promising area for future research that consists of an open system in which far-from-equilibrium wet–dry cycling of organic reactions occurs repeatedly and iteratively at mineral surfaces under hydrothermal-like conditions. Additional detailed reviews about the origins of cellular life can be found in the literature (Schrum *et al.*, 2010; Lambert *et al.*, 2012; Ruiz-Mirazo *et al.*, 2014; Camprubí *et al.*, 2019).

6. Systems chemistry, the reduction of a problem to a set of essential characteristics, was first used in 2005 by von Kiedrowski (Kindermann *et al.*, 2005; Stankiewicz and Eckardt, 2006) when describing the kinetic and computational analysis of a nearly exponential organic replicator. As a relevant example of the system chemistry approach, Sutherland's team showed that precursors of ribonucleotides, amino acids and lipids can all be derived by the reductive homologation of hydrogen cyanide and some of its derivatives, and thus that all the cellular subsystems could have arisen simultaneously through common chemistry (Patel *et al.*, 2015). The essence of systems chemistry has also been recently expanded (Strazewski, 2019).

Two main possible strategies

Two distinct operational approaches, autotrophy and heterotrophy, are promoted as enabling the earliest life, their differences depending upon the role played by CO_2. In the 'metabolism-first approach,' the proponents of autotrophic life call for the direct formation of simple molecules from CO_2 to rapidly generate life (Wächtershäuser, 2007). Energy sources with the capacity to reduce CO_2 were likely provided by the oxidative formation of pyrite from iron sulphide and hydrogen sulphide, which would have given rise to a two-dimensional 'surface metabolism'. A laboratory test of this scenario in an oceanic setting, as proposed by Michael Russell and colleagues (Martin *et al.*, 2008), simulated the prevailing conditions in alkaline hydrothermal vents and generated low yields of simple organics (Herschy *et al.*, 2014). Hence, a challenge remains for the proponents of a metabolism first approach, namely, to produce large enough precursor prebiotic molecules to support the emergence of life as we know it.

In the second hypothesis, the primeval soup scenario (also known as 'replication first'), complex organic molecules accumulated in a warm little pond, à la Darwin. Laboratory efforts to generate a primitive living cell-like system with hydrothermal conditions, wet–dry cycling, or minerals continue to the present, with further endeavours required to focus on constraining the boundary conditions to produce key molecules, such as protein enzymes and ribonucleic acid (RNA), and to compartmentalize them (Monnard and Walde, 2015).

For compartmentalization, amphiphilic compounds can spontaneously assemble into membranous vesicles in hydrothermal fluids (Milshteyn *et al.*, 2018; Damer and Deamer, 2020). Experiments that demonstrate how different prebiotically available building blocks can become the precursors of vesicle-forming phospholipids were reviewed by Fiore and Strazewski (2016). For example, mixtures of C10–C15 single-chain amphiphiles form vesicles in aqueous solutions at temperatures of ~70 °C in the presence of isoprenoids and under strongly alkaline conditions (Jordan *et al.*, 2019). Vesicles functionalized with RNA and peptides would have provided an interesting step towards the formation of early protocells (Izgu *et al.*, 2016).

As for prebiotic peptides, considerations include the role of mineral surface chemistry in controlling their origin (Erastova *et al.*, 2017) and formation on oxide surfaces (Lambert *et al.*, 2013; Kitadai *et al.*, 2017) and on other minerals (Kitadai *et al.*, 2021). The self-assembly of longer prebiotic polypeptides produced by the condensation of non-activated amino acids on oxide surfaces has also been reported (Martra *et al.*, 2014). Environmentally driven wet–dry cycles would have favoured ester-mediated amide bond formation (Forsythe *et al.*, 2015), and short peptides were likely to have played a role in the steps that led to the formation of a protocell (Fishkis, 2007), as reviewed in Frenkel-Pinter *et al.* (2020).

RNA played probably a starring role in life's emergence (Budin and Szostak, 2010; Bernhardt, 2012; Higgs and Lehman, 2015; Benner *et al.*, 2020), and significant progress in producing it abiotically in a 'one-pot synthesis' has been made. As already mentioned, Sutherland's team has simultaneously produced the precursors of nucleic acid amino acids and lipids, starting with hydrogen cyanide, hydrogen sulphide, and UV light (Patel *et al.*, 2015). The synthesis of the pyrimidine nucleosides driven solely by wet–dry cycles has also been reported (Becker *et al.*, 2019). In addition, 5′-mono- and diphosphates can form selectively in one-pot reactions in the presence of phosphate-containing minerals. Diamidophosphate efficiently phosphorylates a wide variety of potential building blocks, nucleosides/nucleotides, amino acids, and lipid precursors under aqueous conditions. Significantly, higher-order structures, oligonucleotides, peptides, and liposomes, are formed under the same phosphorylation reaction conditions (Gibard *et al.*, 2018).

Conclusion

Despite encouraging results, it still seems difficult to consider that life started with true RNA molecules, long RNA strands being not simple enough and yet too difficult to build under prebiotic conditions. As a prediction, a promising avenue will consist of open wet–dry cycling organic reactions running far from equilibrium, at mineral surfaces and under hydrothermal-like conditions. Let brave chemists face the challenge. To young astrobiologists, I absolutely agree with James Fraser Stoddart when he recommended '*Whatever you do, tackle a 'big problem' in chemistry. Although the road you will travel along will be quite unpredictable, it will reveal an endless supply of surprises and the experience will be a rewarding one*' (Stoddart, 2012).

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