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- PERSPECTIVE

ORIGIN OF LIFE

Life as We Don't Know It

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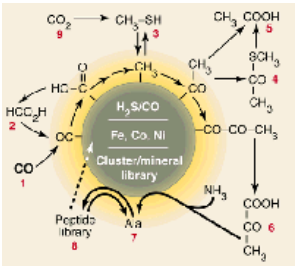
Theories of the origin of life on Earth fall into two general categories. Prebiotic broth theories postulate a protracted origin by the self-assembly of high-molecular weight structures, such as RNA, proteins, and vesicles, in a cold prebiotic broth of preaccumulated modules (1). More recently, theories based on a hydrothermal origin have gained ground. For example, the theory of a pressurized iron-sulfur world (2) suggests a fast origin by an autotrophic metabolism of low-molecular weight constituents, in an environment of iron sulfide and hot magmatic exhalations. Cody *et al.*'s (3) results on page 1337 of this issue provide key support for the latter theory and greatly strengthen the hope that it may one day be possible to understand and reconstruct the beginnings of life on Earth.

Pyruvic acid, CH_3–CO–COOH, is one of the most crucial constituents of extant intermediary metabolism. It occurs in numerous metabolic pathways, notably the reductive citric acid cycle and the pathways that produce amino acids and sugars. It has been suggested that pyruvic acid or its anion pyruvate formed primordially by double carbonylation (4). Cody *et al.* provide experimental support for this suggestion. They show that pyruvic acid forms from formic acid in the presence of nonylmercaptane and iron sulfide at 250°C and 200 MPa. Water is initially absent and forms only by the dehydration of the formic acid. This result poses fascinating thermodynamic and kinetic questions. Pyruvic acid is an extremely heat-sensitive compound that decomposes at its boiling point of 165°C. It appears paradoxical that at the very high temperature required for dehydration of formic acid, the relatively unstable pyruvic acid can form and exist at detectable concentrations. Moreover, it is astonishing that acetic acid is formed at a lower yield than pyruvic acid. The explanation may well lie in the very high pressure.

The work is particularly exciting because experience with organic synthesis in the high-pressure/high-temperature regime is very limited. The experiments require a combination of 200 MPa (corresponding to a rock depth of about 7 km or a 20-km water column) and 250°C, in addition to high CO pressure in the absence of water. It remains to be established whether such conditions are geophysically possible.

The new finding, if it holds, fills a critical gap in the experimental picture of the iron-sulfur world (see the figure). All individual reaction steps for a conversion of carbon monoxide **1** to peptides **8** have now been demonstrated: formation of methyl thioacetate **4** (4), of pyruvate **6** (1), of alanine **9** by reductive amination of pyruvate **6** (5), and of peptides **8** by activation of amino acids with CO/H_2S (6). The challenge will now be to overcome the discrepancies in the reaction conditions and to establish the right conditions for autocatalysis (reproduction) and evolution. This may involve a primitive version of the citrate cycle in which (methyl) thioacetate and pyruvate participate (3, 6, 7) and/or ligand (notably peptide) feed back to the catalytic metal center (3, 6).



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Reactions in the iron-sulfur world.

Reaction conditions are given in the table below. The dotted arrow represents ligand feedback.

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The reaction scheme in the figure is in substantial agreement with extant metabolism in terms of overall metabolic patterns, reaction pathways, and catalysts. The newly demonstrated formation of pyruvic acid by double carbonylation, however, has no analog in extant metabolism. It may have disappeared because of metabolic takeover, first by a reverse pyruvate-formate-lyase reaction and later, after the advent of thiamine pyrophosphate, by carboxylation with pyruvate oxidoreductase.

Cody *et al.*'s results support the view that the primordial organisms were autotrophs feeding on carbon monoxide. But more importantly, the reactions shown in the figure can still occur today because the required conditions are in general still available on Earth, albeit at a lesser frequency. They may thus be a source for geoorganics today; these geoorganics may serve as food for extant heterotrophs, and primitive microbes feeding on CO might still be tracked down in hot pressurized spaces previously inaccessible to exploration.

The reaction conditions chosen by Cody *et al.* are a compromise between the requirements of geochemical modeling and the requirements of the experimental technique. CO gas cannot be used at these very high pressures without extreme danger. Decomposition of formic acid was therefore used as a source for CO. This requires a temperature of 250°C and the absence of water. But in the real world, the temperature may well have been lower, as may have been the pressure. On early Earth, outgassing (the release of gases by volcanic activity) must have been massive and omnipresent, with a wide spectrum of physical conditions, which only

later became restricted to vents and volcanoes because of a thickening crust.

It is occasionally suggested that experiments within the iron-sulfur world theory demonstrate merely yet another source of organics for the prebiotic broth. This is a misconception. The new finding drives this point home. Pyruvate is too unstable to ever be considered as a slowly accumulating component in a prebiotic broth. The prebiotic broth theory and the iron-sulfur world theory are incompatible. The prebiotic broth experiments are parallel experiments that are producing a greater and greater medley of potential broth ingredients. Therefore, the maxim of the prebiotic broth theory is "order out of chaos." In contrast, the iron-sulfur world experiments are serial, aimed at long reaction cascades and catalytic feedback (metabolism) from the start. The maxim of the iron-sulfur world theory should therefore be "order out of order out of order."

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- PERSPECTIVE

NEUROBIOLOGY

Turning Attraction into Repulsion

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A series of molecular cues guide axons as they extend in the embryonic environment. In her Perspective, Pasquale discusses new findings that reveal how signals originating from adhesive contacts between axons and the cells they encounter can repel the axon, causing it to break the adhesive contacts and move away (Hattori *et al.* and Galko and Tessier-Lavigne).

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