Pyrite and the origin of life

SIR—R. J. P. Williams in his recent News and Views article[1] raises among other problems the following: "... we must search for the functional advantage for the material [such as FeS_2] in the biological niche in which it is synthesized". A detailed hypothetical answer to this problem, as far as pyrite is concerned, is central to the recent theories of the origin of life due to G. Wächtershäuser. The answer is formulated in the title of the first of his papers on the origin of life: "Pyrite Formation, the First Energy Source for Life: a Hypothesis"[2].

In this paper, Wächtershäuser notes that certain anaerobic bacteria combine archaic features with a chemoautotrophic metabolism. Starting from this observation, he recommends "the re-examination of the long neglected view that the origin of life and the origin of [chemo-]autotrophic metabolisms coincide" — in contrast with the widely adopted heterotrophic soup theory, and also with the view that the original metabolism is photoautotrophic.

Wächtershäuser suggests that the neglect of the chemoautotrophic option results from the problem of finding a likely energy source. He proposes the anaerobic biomineralization of pyrite as the first energy source for life, calculating the yield of free energy from this process. (More recently, upon a suggestion by Ian R. Kaplan, he has also calculated the still more highly exergonic formation of pyrite when Fe_3S_4 intervenes.)

This hypothesis suggested further hypotheses of great explanatory and predictive power covering precellular life and the formation of cells. These hypotheses enabled Wächtershäuser to predict that the production of pyrite via carbon fixation (in competition with hydrogen formation) by some of the extant archaic bacteria is highly probable[2,6]. Possible corroborations of this prediction seem to be offered by the two recent letters to *Nature*[3,4] discussed by Williams[1].

Another prediction of central importance to Wächtershäuser's theory is the evolution of precellular forms of life (surface metabolists); that is, of monomolecular layers of autocatalytic processes of carbon fixation, bound, *in statu nascendi*, to positively charged mineral surfaces such as pyrite crystals that are biomineralized by the surface metabolists themselves[2,5,6]. This prediction shows a striking similarity with Williams's suggestions that the "surfaces of iron sulphide minerals could also have been catalysts [for the metabolism]".

Indeed, Wächtershäuser points out that his surface metabolists act as catalysts for the biomineralization of pyrite, whereas the pyrite surfaces act as catalysts for the autocatalytic surface metabolism[5,6]. Another of his predictions (also possibly supported by Farina *et al.*[3] and Mann *et al.*[4]) implies that the surfaces of biogenic pyrite are binding, *in statu nascendi*, membranes of lipids and of anionic peptides[5], a hypothesis which could explain the otherwise still unexplained asymmetry in the structure of the membranes.

These hints may, I hope, draw attention to a new theory — to my knowledge, the first radical alternative to Haldane's and Oparin's soup theory. A workable alternative seems to me urgently needed, especially in view of the low temperature of the soup required by the most prominent representatives of the theory[7]. This temperature requirement seems to me contradicted by the logic of the geophysical situation as well as by the empirical findings of Woese: both suggest a hot origin of life (with which Wächtershäuser's theory is compatible). Incidentally, Woese's hot origin hypothesis appears to render sterile all those speculations about hot-temperature sterilization, due perhaps to the impact of meteorites.

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1. Williams, J.R.P. *Nature* **343**, 213–214 (1990).
2. Wächtershäuser, G. *System. appl. Microbiol.* **10**, 207–210 (1988).
3. Farina, M. *et al. Nature* **343**, 256–258 (1990).
4. Mann, S. *et al. Nature* **343**, 258–261 (1990).
5. Wächtershäuser, G. *Microb. Rev.* **52**, 452–484 (1988).
6. Wächtershäuser, G. *Proc. natn. Acad. Sci. U.S.A.* **87**, 200–204 (1990).
7. Miller, S.L. & Bada, J.L. *Nature* **334**, 609–611 (1988).

SIR—Wächtershäuser[1,2] has described how the exergonic formation of pyrite (FeS_2) could have been the electron-donating energy source for an autotrophic origin of life. Early Archaean submarine hot springs of moderate (100–200 °C) temperature may have issued through fine iron monosulphide tubes comprising natural chemical gardens — catalytic flow reactors in which the first metabolists could have been cultured[3]. Hall *et al.*[4] have also argued for an ancient origin of ferredoxins, the iron-sulphur-proteins that occur in all types of organisms, which are involved in a wide range of biochemical reactions as agents for electron transfer, but which originally catalysed anaerobic fermentation[4].

We suggest one more important role for iron sulphide, that of nucleating the membrane of the earliest cell walls. We have demonstrated that iron monosulphide gels can form macroscopic spherical shells 1 to 20 mm across[3]. On a microscopic scale, spherical aggregates of pyrite about 5 μm in diameter are found within fossil hydrothermal chimneys. These framboids appear to have grown inorganically from a spherical shell of iron sulphide gel. The pyrite crystallites comprising the interior of these framboids are about 0.1 μm across, and so are similar in size to those found in the magnetotactic bacteria.

In a hydrothermal chimney, the iron sulphide shells would initially have consisted of periodic arrays of iron monosulphide crystallites, which would have grown by the diffusion of iron from an aqueous phase into polysulphide droplets. These shells would have provided a site for concentration of polar organic species, by adsorption onto the iron sulphide surfaces.

Once a critical concentration of polar organics had been adsorbed, further growth of the iron sulphides would have been inhibited. Then the organics could become inherently ordered to a liquid crystalline structure. The periodic arrays of the iron sulphide crystallites may have assisted this ordering, developing an organic layer with varied degrees of structure similar to those reported by McConnell *et al.*[5] in phospholipid membranes. A further concentration of organics onto the first adsorbed layer could follow, the whole comprising a cell membrane precursor.

Similarities between the iron sulphide bearing bacteria and the earliest stages of framboidal iron sulphides give further support for the view that inorganic and organic evolution to the earliest life forms relied on a redox reaction involving carbon fixation concomitant with the oxidation of iron monosulphide to disulphide.

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1. Wächtershäuser, G. *System. appl. Microbiol.* **10**, 207–210 (1988).
2. Wächtershäuser, G. *Microbiol. Rev.* **52**, 452–484 (1988).
3. Russell, M.J., Hall, A.J. & Turner, D. *Terra Nova* **1**, 238–241 (1989).
4. Hall, D.O., Cammack, R. & Rao, K.K. *Nature* **233**, 136–138 (1971).
5. McConnell, H.M., Tamm, L.K. & Weis, R.M. *Proc. natn. Acad. Sci. U.S.A.* **81**, 3249–3253 (1984).

Geomorphology and mantle plumes

SIR—The recent article by Cox[1] is valuable in highlighting the potential significance of geomorphic data in assessing models of continental rifting and associated surface uplift. In his analysis of the geomorphic evidence for the plume-related surface uplift model proposed by White and McKenzie[2], there are, however, important elements that permit well supported alternative interpretations.