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Ref. 80-7
June, 1980

National Science Foundation Grants:
OCE 76-23352
OCE 77-23978
OCE 78-26368
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ABSTRACT

Submarine hydrothermal systems provide all of the conditions necessary for the abiotic synthesis of organic compounds, polymers, and simple cell-like organisms. An analysis of the Archaean rock and fossil record shows that fossils of simple organisms are found in rocks deposited in hydrothermal environments. Biochemical experiments have shown that thermal energy is an efficient means for the abiotic synthesis of "protocell" structures. The continuous flow of circulating fluids in a hydrothermal system provides the thermal and chemical gradients which create the variation in conditions necessary for the successive reactions to take place. Other models for the origin of life fail to fulfill one or more of these requirements.

INTRODUCTION

Submarine hydrothermal vents recently discovered along mid-oceanic rift systems [1] provide all of the conditions necessary for the creation of life on Earth. We have found a parallel between the conditions in the vents and the conditions used by Sidney Fox and others [2,34] in the experimental abiotic synthesis of high molecular weight organic polymers and primitive organized structures (microspheres) [2] with many of the characteristics of living organisms.

It is apparent from an analysis of the events of early Earth history that hydrothermal activity connected with seafloor volcanism commenced simultaneously with the formation of the primeval oceans and that this followed soon after the final accretion of the Earth $\sim$3.9 billion years ago. In examining the earliest Precambrian rock and fossil record it is notable that organisms remarkably similar to the microspheres synthesized by Fox and his colleagues have been found in rock units which we and others have interpreted as spreading ridge, hydrothermal assemblages [3].

The convergence of data from many researchers in the fields of experimental biochemistry, micropaleontology, microbiology, planetology, and marine geology has convinced us that life almost certainly originated in submarine hydrothermal vents. In this paper, we synthesize the evidence from these fields to present a unified model for the origin of life on Earth. Our argument is presented in the following order:

(1) early Earth history and the origin of the atmosphere, oceans and hydrothermal systems;

(2) the history and results of abiotic synthesis experiments;

(3) a description of submarine hydrothermal processes and a discussion of the applicability of the Fox model to hydrothermal systems;

(4) the rock and fossil record of the earliest Precambrian;

(5) the evaluation of the hydrothermal vent hypothesis in comparison with other hypotheses for the origin of life;

(6) a description of the postulated first organisms.

In particular we will show that the hydrothermal circulation of fluids resulting from submarine volcanic activity created the necessary thermal and chemical gradients in which complex organic polymers and "protocells" could be formed.

Early Earth History

It is generally accepted that the Earth and the other terrestrial planets accreted roughly 4.5 BYBP. The processes of accretion, as discussed by Smith and others, [4] led to the differentiation of the core from the mantle. This stage of accretion and core-formation proceeded from roughly 4.6 to 4.2 BYBP.

Evidence from the Moon indicates that the inner Solar System was bombarded by large planetesimal objects (10-100 km in diameter) from 4.2 until about 3.9 BYBP [5]. The impact of as many as 10^3 to 10^4 of these objects onto the Earth would have significantly contributed to major volcanic activity as well as contributing significant mass to the early Earth. It is possible that as much as one-fifth of the Earth's mass was acquired in this period of giant impacting [6].

The enormous energies released through the processes of giant impacting and the decay of short-lived radionuclides would be sufficient to melt the surface of the planet, covering it with a hot silicate magma ($T>1600°C$) [4,7]. The magma would be convecting vigorously, degassing volatiles to form a primeval secondary atmosphere [4,7,8] and radiating heat into that atmosphere. The atmosphere itself would be convecting and radiating heat into space. Figure 1 presents a model of the heat transfer processes by which the Earth would cool.

As impacts diminished over time and as heat radiated into space, the surface of the planet eventually would have cooled sufficiently to permit thin crustal fragments to form. It has been postulated that this thin protocrust had an anorthositic composition [7]. Beneath this protocrust, the silicate melt would continue to convect and would also

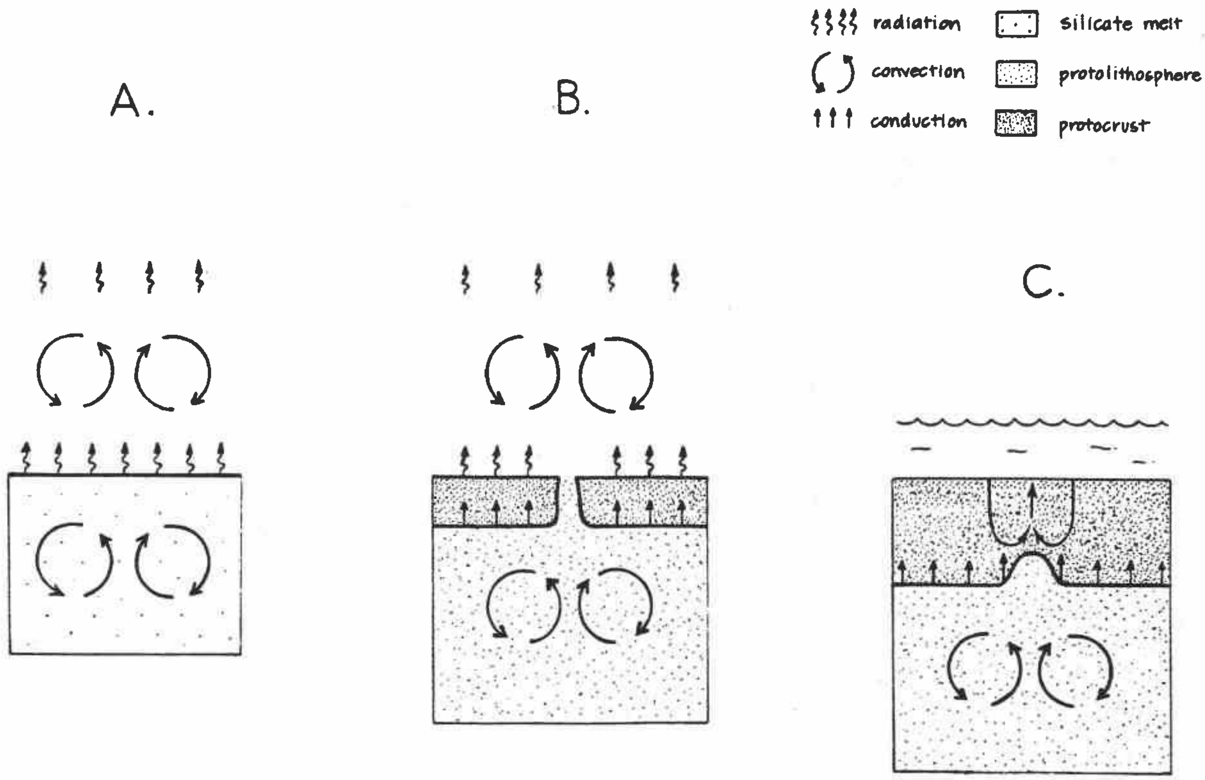

Figure 1. Heat transfer processes on the early Earth surface.

A. The molten surface formed by the secondary accretion process cools by radiation to the atmosphere which convects and radiates into space.

B. The formation of thin crustal plates, which transmit heat by conduction, provides an insulating shell which begins to lower atmospheric temperatures.

C. As the temperature at the surface cools below the liquidus of water it condenses from the atmosphere and the dominant heat transfer mechanism becomes convective circulation of water in the crust.

undergo fractionation through partial crystallization (see Figure 1). The accumulation of a less dense, fractionated melt beneath a dense anorthositic protocrust would cause the crust to be isostatically unstable; slabs of anorthosite would sink into the melt [9], exposing the less dense melt to more rapid cooling via radiation to the convecting atmosphere above. The crystallization of the less dense melt at the surface would lead to lateral inhomogeneities in the protocrust [7].

Ultimately, as cooling proceeded further, rafts of solid silicates would coalesce to form a continuous, though thin and brittle shell. Cooling through the shell of protocrust would proceed by conduction. Eventually, the surface of the planet would cool sufficiently to allow liquid water to condense from the atmosphere, and rains would begin on the primitive Earth.

The lateral inhomogeneities caused by the sequence of crystallization, the variations in density of the phases crystallized, and the processes of convective overturning would have a profound effect on the future evolution of the crust. The less dense, isostatically higher protocrust would form the primitive continental areas [7]. Ocean basins would form in areas of denser, isostatically lower protocrust [7]. The thin suboceanic crust would be subjected to both tidal and isostatic body forces and to surface drag from the underlying convective magma, leading it to fracture and forming primeval spreading centers [Figure 1c]. Once ocean waters came into contact with ocean basin volcanism, be it through individual volcanic centers or through rift zone processes, hydrothermal activity would commence.

It has been shown that hydrothermal convection at spreading centers is a highly efficient mechanism for the removal of heat from newly-formed crust [10]. It is reasonable to conclude that hydrothermal activity in ocean basins began with the formation of the oceans and at a time when significant quantities of volatiles were being degassed from the interior of the young planet. While it is not unreasonable to believe that the processes of plate tectonics began 3.8 billion years ago, the critical point which we wish to make is that the eruption of lava onto the seafloor and the hydrothermal cooling of that lava is the process required for our model of the origin of life.

Abiotic Synthesis Experiments

Early research on the origin of life was initially done by chemists and biochemists as experiments in closed laboratory systems. The typical procedure involved confining a mixture of gases believed to be present in a primeval Earth atmosphere in some sort of distilling container, providing an energy input, and analyzing the resulting reaction products. Calvin [11] gives a thorough discussion of the history and results of early experiments.

It has been argued by Miller, Holland, and others [11,12] that the early atmosphere which resulted from volcanic outgassing was a reducing one. The gases thought to have been present, and often observed as the products of volcanic outgassing, are H_2, H_2O, CO_2, H_2S, S, CH_4, NH_3, and/or SO_2 [12]. Various combinations of these gases have been used in abiotic synthesis experiments.

There were at least five different sources of energy available on a primitive Earth: high energy particles from radioactive decay, solar radiation (UV and visible), electrical discharges from the atmosphere, shock waves from planetesimal impacts, and thermal energy from volcanism. Many of the early experiments as well as many of the present day experiments use spark discharges in their closed systems. This was the procedure used by Miller and Urey in their pioneering experiments [11]. As recently as November, 1979, Yamagata, et al. reported on the phosphorylation of adenosine by electric discharges [13]. Other experiments have used UV radiation and electron bombardment (to simulate radioactive decay) as sources of energy input.

The pioneer in the use of thermal energy sources has been Sidney Fox, who, with his colleagues at the Institute of Molecular Evolution at the University of Miami, developed a model for the completion of the following sequence: primordial gases→amino acids→primitive protein→a primitive organized structure. Harada and Fox [14] compared the results they had obtained in thermal energy experiments using silica as a substrate with results obtained by Miller in spark discharge experiments. The thermal energy experiments produced a far greater variety of amino acids, as is shown in Table 1 taken from Harada and Fox [14].*

In a 1971 paper, Fox presented the thermal model on which he and his colleagues based their experiments. It consisted of "(a) heating the system above the boiling point of water; and (b) the intrusion of water." [2] He stated that the sequence required geologically anhydrizing temperatures (above 100°C) and sporadic rain or "other common geological events of water such as drought or recession of the seas." Using the model, Fox succeeded in creating a primitive organized structure which had many lifelike properties. Early experiments had shown how amino acids could readily be formed. In order to achieve polymerization of the amino acids into complex proteins, they heated the amino acids above the boiling point of water and created polymers with high molecular weights. When the polymers came into contact with liquid water, they spontaneously formed structurally organized units which had "a cellular type of ultrastructure, double layers, abilities to metabolize, to grow in size, to proliferate, to undergo selection, to bind polynucleotides, and to retain some macromolecules selectively." [2]*

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*See Appendix

TABLE 1. COMPOSITIONS OF AMINO ACIDS PRODUCED THERMALLY IN THE PRESENCE OF SILICA AND BY ELECTRIC DISCHARGE† (from Harada and Fox, 1965)

AMINO ACID	THERMAL SYNTHESIS			ELECTRIC DISCHARGE SYNTHESIS	
	Quartz sand (950°C) (%)	Silica gel (950°C) (%)	Silica gel (1050°C) (%)	Spark discharge‡ (%)	Silent discharge‡ (%)
Aspartic acid	3.4	2.5	15.2	0.3	0.1
Threonine	0.9	0.6	3.0	--	--
Serine	2.0	1.9	10.0	--	--
Glutamic acid	4.8	3.1	10.2	0.5	0.3
Proline	2.3	1.5	2.3	--	--
Glycine	60.3	68.8	24.4	50.8	41.4
Alanine	18.0	16.9	20.2	27.4	4.7
Valine	2.3	1.2	2.1	--	--
Alloisoleucine	0.3	0.3	1.4	--	--
Isoleucine	1.1	0.7	2.5	--	--
Leucine	2.4	1.5	4.6	--	--
Tyrosine	0.8	0.4	2.0	--	--
Phenylalanine	0.8	0.6	2.2	--	--
α-NH_2 butyric acid	0.6	--	--	4.0	0.6
β-Alanine	?§	?§	?§	12.1	2.3
Sarcosine	--	--	--	4.0	44.6
N-Methylalanine	--	--	--	0.8	6.5

† Basic amino acids are not listed in the table, because these amino acids have not been fully studied. Some analyses of the thermal products showed peaks corresponding to lysine (ornithine) and arginine.

‡ Recalculated from the results of Miller (1955).

§ β-Alanine peak obscured next to another unknown peak.

Modern Submarine Hydrothermal Systems

The quenching of newly injected crust on the sea floor by circulating seawater, which is clearly recorded in the earliest rocks (see below), continues in the same environment today. The first direct observations of these hydrothermal systems along mid-oceanic spreading centers was carried out in early 1977 along the Galapagos Rift [1] using the deep diving submersible, ALVIN. Research on the extensive set of data and samples collected on this expedition has allowed us to characterize the behavior of the interaction of seawater with newly erupted crust in great detail. More recent observations from the East Pacific Rise at 21°N provide additional significant information [15].

Sites of submarine volcanism bring together in a single system a unique combination of rocks, gases, heat, and water. The relevant features of a hydrothermal system are summarized in Figure 2. These sites of eruption occur where crustal plates are spreading apart, allowing magma to rise in the crust, approaching closer to the sea floor and producing a strong thermal gradient across the layer of previously erupted and cooled rock. This layer of rock has undergone thermal contraction and fracturing and is subject to tensional cracking resulting from the spreading of crustal plates. As a result, the crust is permeable and becomes saturated with seawater.

"Active" hydrothermal circulation is driven by the rapid transfer of heat from the magma to the water at the "cracking front" [16] (see Figure 2a). The water which saturates the cold, permeable rock "attacks" the magma body in a continuous cycle:

cooling→crack propagation→penetration→convection→cooling.

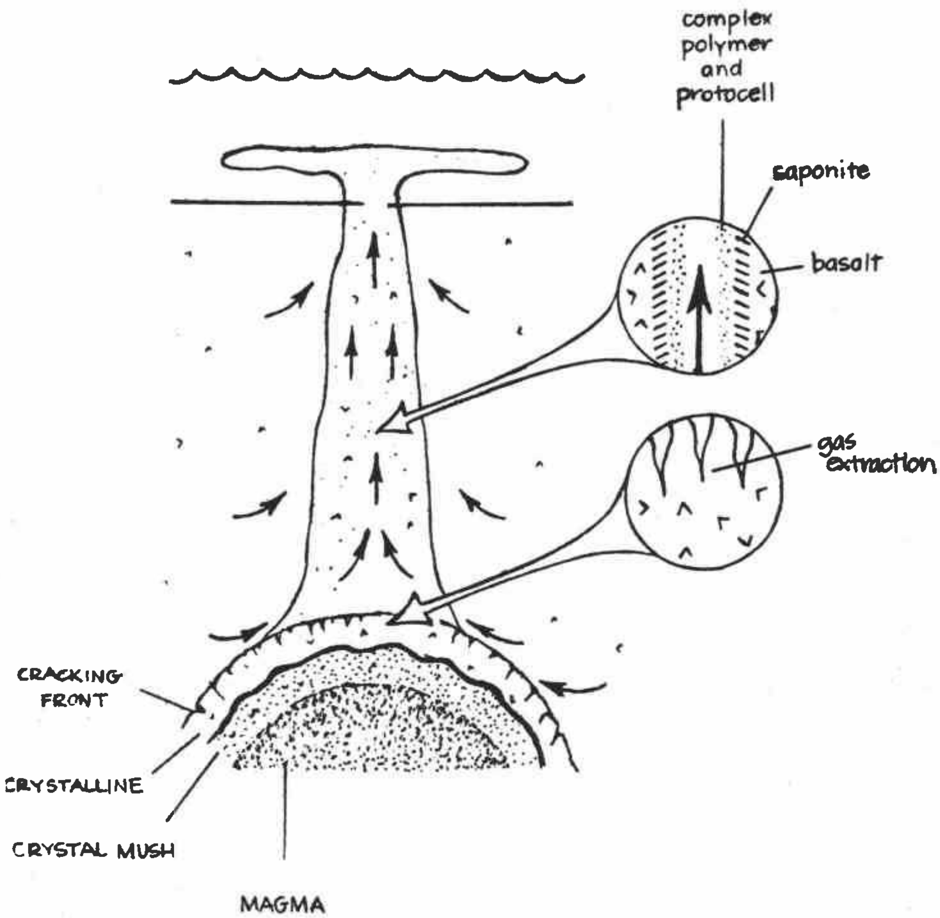

Figure 2a. Model seawater hydrothermal system.

Metal ions and gases are extracted from the rock at the cracking front where the fluid migrates toward the magma through fractures formed by thermal contraction. The hot, low density and low viscosity water rapidly convects upward, mixing with cooler water along the height of the column (arrows). This mixing of vent waters creates the thermal and chemical gradients which are necessary for abiotic synthesis (see text and Figure 2b). The waters emitted from the vents form a plume in the ambient bottom water. The plume carries suspended particles outward (such as seen in Figure 4) to be deposited around the vent.

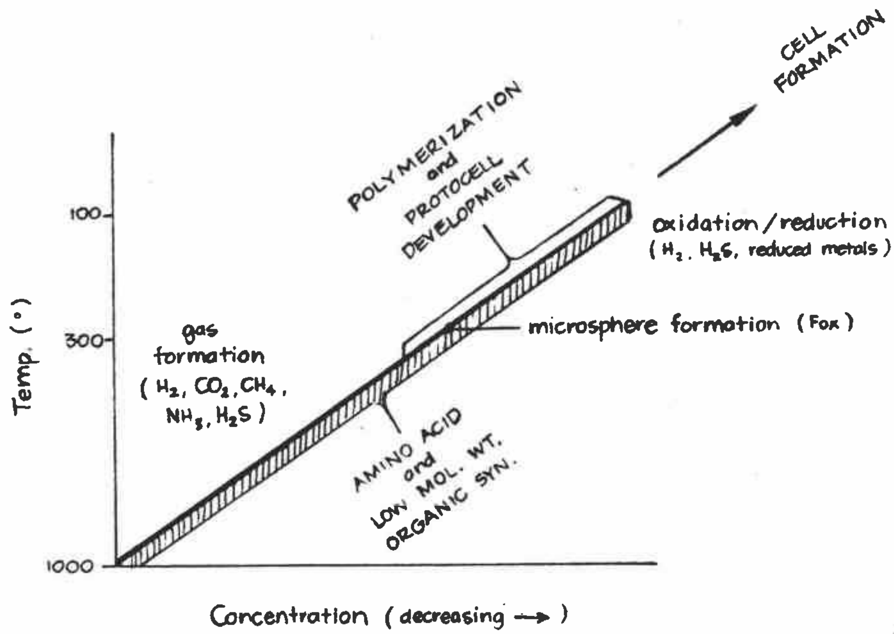

Figure 2b. The proposed sequence of chemical and biochemical events leading to the formation and development of Fox-like "protocells" would occur along the thermal and the chemical substrate concentration gradients which exist within the upwelling fluids of these hydrothermal vents. Amino acids and other reactive compounds such as thiocyanate and formaldehyde could be synthesized from the gases initially at high temperatures (800 to 1000°C) and then catalyzed by reactive compounds. At lower temperatures (80 to 100°C) additional amino acids and peptides could be synthesized. "Protocell" formation could occur at approximately 300°C (see Fox and text) or at lower temperatures depending on the pH and other chemical and physical conditions. These "protocells" would be deposited along with silica precipitated from the supersaturated hydrothermal fluids to form carbonaceous fossil-bearing cherts, such as found in the Isua rocks (Fig. 2a -- plume). Further development of the protocells could occur in the cooler reduced waters as a result of chemical oxidation/reduction reactions involving the reactive gases and reduced metals emanating from the vents.

Within the magma body, removal of heat from the upper surface leads to plating of the crystallizing mineral phases onto the roof of the magma chamber, forming a zone of crystalline mush which grades upward into solid rock. Heat from the magma is conducted across this interface up to the "cracking front" where it is extracted by circulating water.

This interface is the site where a significant fraction of the degassing of the Earth has occurred. Gordon and Lilley [17] have shown that significant quantities of CO_2, NH_3, and H_2 are present in the hydrothermal fluids at the Galapagos Rift. O'Neil [18] has analyzed the carbon isotope composition of CO_2 in these samples and found δC^{13} values of -5.1 to -5.3, establishing it as primordial carbon. Primordial He^3 is also present in the Galapagos fluids. The constancy of the He^3/heat ratio [19] in several individual vents suggests that the gases and heat are extracted from the rock in the same process [20]. In addition to these gases, the magma contains all of the naturally occurring elements. Most of them are extracted to some degree by the hydrothermal fluids and carried upward in solution.

As the magma crystallizes, the gases and the "incompatible" elements (those not entering the growing crystal lattices) are fractionated into the intercrystalline fluid, and when crystallization is complete, they occupy the intergranular spaces. As the rock cools below the solidus, the differential thermal contraction of the individual grains will tend to open an interconnecting network. As a fracture propagates into the vicinity, it introduces water into the network.

It is difficult to estimate the maximum temperatures this water can attain. The magma solidifies at $\sim$980°C. Lachenbruch [21] has suggested that such fractures, once intiated, could propagate past the solidus boundary into the area where residual fluids are not entirely crystallized. It is not unreasonable to believe that water could attain temperatures close to the solidus temperature of the magma. Evidence from the Galapagos Rift and the East Pacific Rise indicates that the water reaches temperatures greater than 350°C [15]. Water at these temperatures and sea floor depth has low density and viscosity [22], leading to very rapid convection and the ability to readily penetrate into the rocks. As the fluids rise, they enter an anastomosing and expanding set of fractures and fissures, all the while incorporating cooler water which is drawn into the rising plume from the adjacent cool, permeable rocks. Hot seawater interacts with the basalt, forming saponite, a magnesium-rich smectite clay which incorporates magnesium from the seawater. This process lowers the pH of the seawater by removing OH^- and lowers the Eh by oxidizing ferrous iron in the rocks through the reduction of $SO_4^=$ and/or oxygen from the dissociation of water. The fluids emerging out of the sea floor from vents on the Galapagos Rift and from most vents on the East Pacific Rise (21°N) had temperatures in the range of 10°-30°C [1] as a result of the mixing process. However, at some of the 21°N vents, water emerged at temperatures of ∿350°C. Presumably, there was a direct vertical channel to some depth in the rock. These high-temperature vents precipitate dissolved metals as sulfides, which form large pinnacles and cones [15]. These pinnacle and cone vents are also called chimneys.

It appears to us that submarine hydrothermal systems are ideal reactors for abiotic synthesis. The stages of the process which we are proposing are shown in Figure 2b. The raw materials could be extracted from the magma in the vicinity of the cracking front. Low molecular weight organic compounds could be synthesized at high temperatures and then rapidly moved upward along a gradient of continuously decreasing temperature and concentration. The exposed surface area in the fractures and interstices is coated with a clay mineral. Clays have been demonstrated to be effective catalysts for abiotic synthesis reactions [23].* Through clay catalysis the low molecular weight organic compounds could be polymerized into more complex compounds and plate out onto the walls of the fractures, forming protocells. The rising fluids would wash the protocells off the surfaces and transport them upward, depositing them in cooler environments in the vent system or on the adjacent sea floor. Continuous supply in a limited area could result in significant accumulations of these protocells.

Fox and his co-workers obtained their results years before the discovery of the hydrothermal vents and the associated animal communities of the Galapagos Rift Zone. It is apparent that hydrothermal vents provide the perfect geologic environment for a process very similar to the one described by Fox and his colleagues.

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*See Appendix

Evidence from the Oldest Rocks

The Onverwacht Series is a 3.5 billion year old [24] rock assemblage exposed in the Barberton Mountain Land of South Africa. The Onverwacht Series forms the base of the Swaziland System. Directly above the Onverwacht Series is the Fig Tree Series. A synthesis and review of the earlier field work in this area was presented by Anhaeusser, et al. in 1968 [24]. Recently, de Wit and Stern [3] have reinterpreted the entire Swaziland System in light of present-day knowledge of submarine hydrothermal processes.

Anhaeusser, et al. described a sequence of metamorphosed basic lavas with interlayered siliceous sediments, occasional thin carbonaceous chert horizons, and bands and lenses of serpentinized ultramafic rocks. de Wit and Stern noted the similarities in metamorphic textures and mineralogies to the sheeted dike complexes and flanking pillow lavas of Phanerozoic ophiolites. The extensive hydration of the Onverwacht minerals they feel is best explained by metamorphism at a spreading ridge. Evidence for this interpretation is also found in the presence of banded iron formation, rich metallogenic sediments, barites, carbonates, and cherts.

It is a very intriguing fact that a number of workers have isolated fossil forms from the Onverwacht Series. Engel, et al. [26] "isolated both siliceous and carbonaceous particles and *carbonaceous filaments*" from cherts, argillites and carbonate beds in the Onverwacht. They report in detail on samples from a chert zone near the base of the Threesprait Stage and a carbonaceous chert and argillite from the Hoogenoeg Stage. They state that the spheroidal fossil-like forms were more common in both of the carbonaceous chert zones than the filamentous forms. Furthermore, "the spheroids within the carbonaceous Onverwacht sediments not only have the morphologies of fossils, but also are intimately associated with the kerogen-bearing carbonaceous substances which appear to form parts of their walls and interiors. They are also closely associated with *kerogen-bearing, filamentous forms which have the appearance of microfossils*." [Italics ours]

Engel, et al. commented upon the difficulties of interpretation of the carbonaceous filaments, reporting that the filamentous layers in the carbonaceous argillic chert are of a diverse morphology and remarkably lifelike. They concluded that "many appear to be true fossils, although less well-preserved that those found in younger Precambrian sediments." There is a striking correlation between these fossil descriptions from 3.4 billion year old hydrothermal sediments and the appearance of complex filamentous organic structures in scanning electronmicrographs of chimney rock samples from 21°N. One of these micrographs is reproduced here as Figure 3.

Even more significant in terms of our model is a group of rocks which outcrop in southwest Greenland at the edge of the Greenland ice cap. This is the Isua supracrustal succession, which includes the oldest sedimentary rocks yet dated [27]. The stratigraphy and petrology of the Isua succession were described by Bridgwater, et al. [28]. The rocks are basic and ultrabasic greenschist, metamorphosed sediments and quartz feldspathic rocks with many similarities to younger greenschist belts. They have been metamorphosed to the amphibolite facies, and some units have retrogressed to the greenschist facies.

The rocks form a layered series. The present mineralogy represents the stable metamorphic assemblages appropriate for the bulk compositions of the rocks. Included in the succession is the Isua ironstone, a finely banded sequence of magnetite and quartz-rich layers. "These siliceous rocks are interpreted as chemical sediments. They are interlayered with chlorite-rich basic rocks interpreted as basic sills" [28]. We interpret this sequence of rocks as submarine lava flows and related hydrothermally-derived silica and iron oxides. The ironstones have been dated by Pb-Pb at 3.76 $\pm$ .07 BYBP [27].

Moorbath, et al. [29] describe the formational history of the Isua Series as follows: "The entire sequence of crustal formation in the Isua area evidenced by presently exposed rock types must have occurred within an interval of not more than ca. 200 m.y. prior to 3700 m.y. ago. This includes multi-stage fractionation of acid igneous rocks from the early mantle, their emplacement, deformation, and metamorphism, to form the presently exposed gneiss complex. It also includes the eruption of basic and acid volcanic rocks, erosion of pre-existing crustal rocks, and their deposition *in an aqueous environment as chemical and clastic sediments*, followed by low to medium grade metamorphism of this supracrustal series to form a typical greenstone belt assemblage." [Italics ours]

Though heavily metamorphosed and deformed, it is apparent to us that the Isua Series represents not just the oldest-known rock unit but the oldest-known hydrothermally-emplaced rock unit. The metamorphic mineral assemblages, the quartzo-feldspathic rocks, the magnetite-quartz ironstone unit, and the layered nature of all the sub-units of this series imply very strongly that the original rocks were ultrabasic intrusions, basic pillow-forming lavas, bedded cherts and banded iron formations such as are found throughout the Archaean, and that they are the products of submarine volcanism and its accompanying hydrothermal activity. In addition, in the cherty layers of the Isua metaquartzite, Pflug and Jaeschke-Boyer (1979) found microfossils bearing a striking resemblance to Fox's microspheres. They located cell-like inclusions in cherty-layers from the Isua Series and analyzed them utilizing Raman laser molecular microprobe. They described the inclusions as follows:

> The fossils occur as individual *unicells, filaments or cell colonies.* [Italics ours] Cells and cell families are usually surrounded by multilaminate sheaths which show a characteristic laminar structure. All specimens observed apparently belong to the same kind of organism named *Isua-sphaera*.
>
> The individual cells are more or less ellipsoid in shape. The mature cells range between 20 and 40 μm in diameter. The cell encloses a more or less globular hollow which is partly filled with organic matter. Apparently, this filling is a remnant of the former protoplasm which has been degraded during fossilisation. A vacuole is often contained in the cell lumen.
>
> Two different types of [Raman] spectre were obtained. One is typical of the brownish substance composing sheath, cell wall and cell filling.
>
> It can be concluded from the[se] analyses that *Isua-sphaera* consists of organic material which is partly present in a carbonised condition, partly in a high rank of coalification very similar to a final stage of graphitisation. This is in accordance with the metamorphic condition of the enclosing rock which seems to be in the range of an upper greenschist. [30]

The other type of spectra is typical of the cell vacuole. These spectra revealed the presence of esters and aliphatic hydrocarbons. They cite the characteristic budding behavior of the organisms and conclude that

Isuasphaera may represent a half-way line between a microsphere-like protobiont and subsequent evolution.*

We have just discussed two of the oldest known geologic terrains on Earth, and our discussion has shown that they are strikingly similar. Table 2 relates the similarities between the Isua Series and the Onverwacht Series to conditions found in the present day hydrothermal regimes of the Galapagos Rift Zone and the East Pacific Rise at 21°N. There are obvious parallels in every category.

Pflug and Jaeschke-Boyer said in the conclusion to their paper, "There is little doubt that *Isuasphaera* is an organism." However, they expressed the concern that the time span between the formation of the earth and the deposition of the Isua rocks, "roughly half a billion years ...appears too short for the evolution from a simple organic compound to an eukaryotic organism to have occurred." In reply to their concern, we will quote Sidney Fox's discussion of the problem:

> One way in which students of the total problem have dealt with the seemingly great complexity has been to postulate a long chemical evolution extending over, say, 25 million years. I will explain here why our experiments lead to the interpretation that the essential steps... could have occurred many times in a very short period, say 25 hr. [2]

To us, it no longer seems puzzling or inexplicable that the earliest known rock contains the fossils of primitive organisms. When one considers the results of abiotic synthesis experiments using thermal energy sources in the light of what we now know about hydrothermal systems in the ocean and the early history of the Earth, it seems natural and inevitable.

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*See Appendix

Figure 3. Scanning electronmicrograph showing thin unraveling strands of organic or possibly inorganic sheaths which were frequently found on hot chimney rock surfaces from 21°N. These structures resemble, to some extent, the fossil structures observed in ancient rocks (see text). Bar is 10 μm. The samples were fixed in sterile artificial seawater containing 2% gluteraldehyde within minutes after the Alvin surfaced. Sterile techniques were used with all specimens. The fixed samples were dried by the critical point method, then mounted on aluminum stubs and coated under a vacuum with a layer of gold 10-20 nm in thickness. The samples were viewed using an International Scientific Instruments Mini-SEM, Model MSM-2, Scanning Electron Microscope.

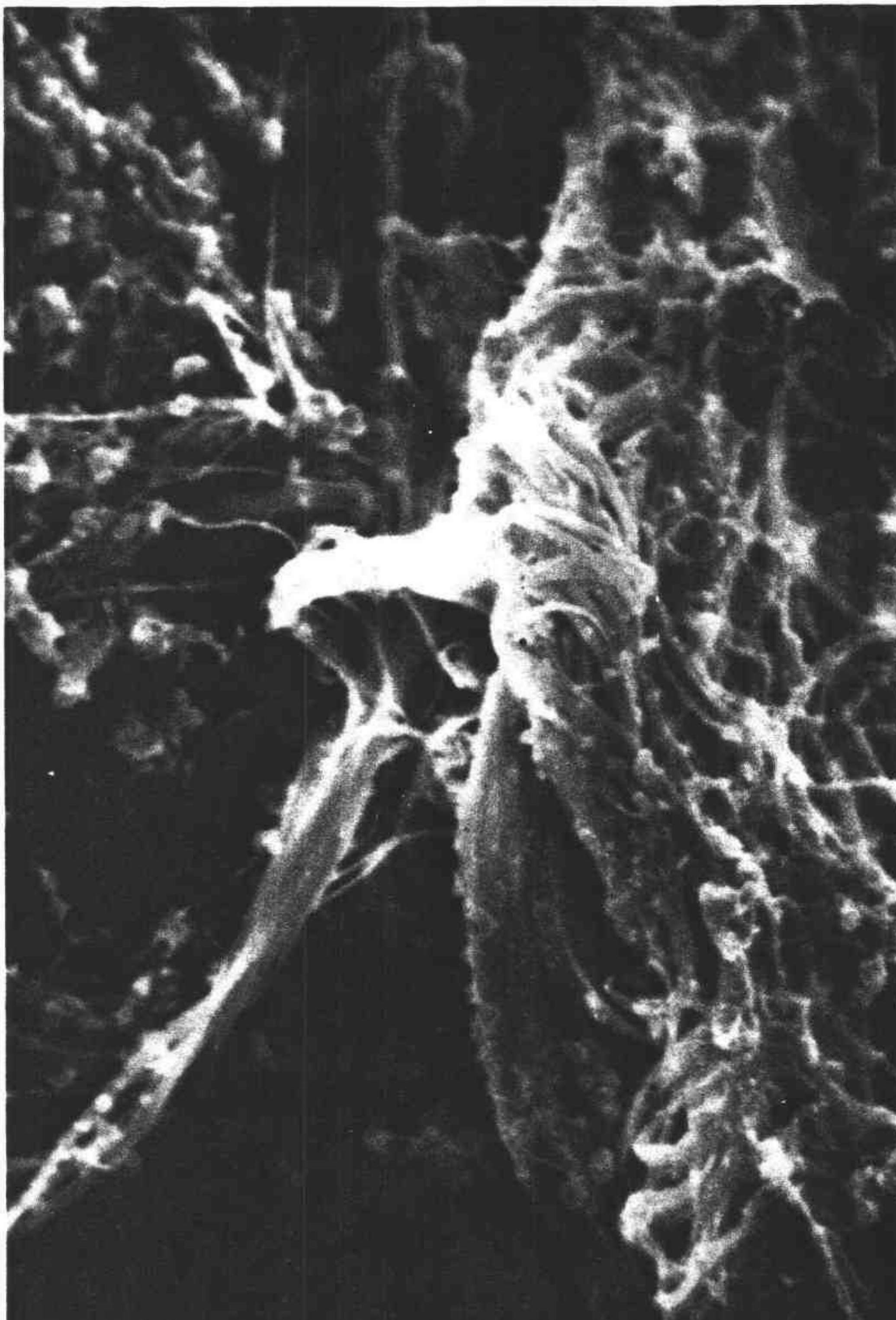

Figure 4. Scanning electronmicrograph showing the surfaces of inorganic crystals covered with deposits of organic debris and microorganisms which have been emitted from "black smoker" chimneys at 21°N and have settled on the surfaces of the chimneys and the surrounding rocks and animals. The samples were prepared as described in Figure 3. Photo magnified 400 x.

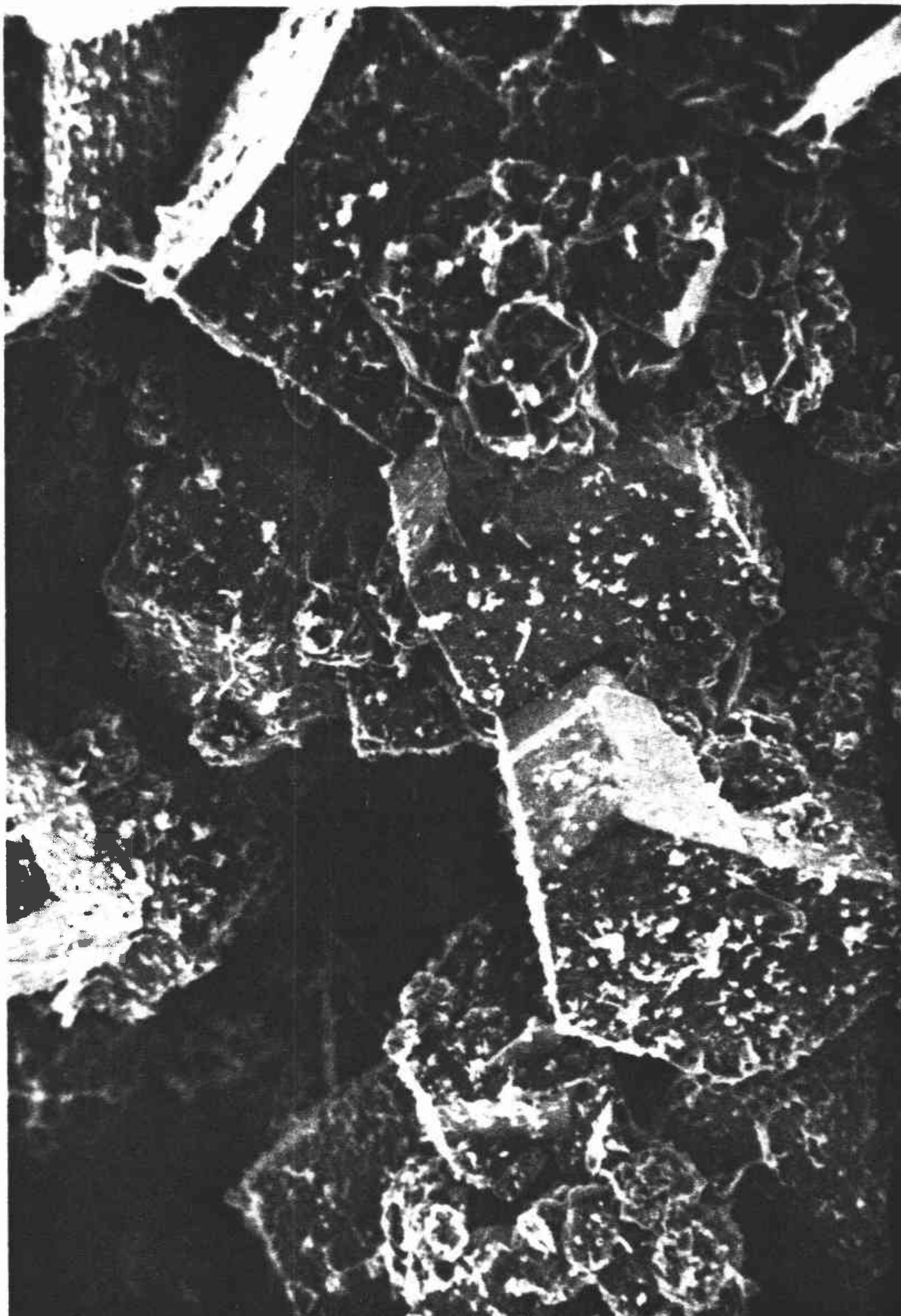

TABLE 2. A COMPARISON OF THE ISUA SERIES AND THE ONVERWACHT SERIES WITH PRESENT-DAY HYDROTHERMAL REGIMES

	ISUA	ONVERWACHT	GALAPAGOS/EAST PACIFIC RISE
Mineralogy	metamorphosed ultrabasic, basic, quartzose and iron-quartz rich	amphibolitized pillow basalts & ultrabasic layers, cherts	Low K tholeitic pillow basalts and subsurface dikes
Chemical sediments	chert, banded iron formation, carbonaceous quartzo-feldspathics, calc-silicates and carbonates	carbonaceous chert, calc-silicates and banded carbonates	radiolarian ooze, massive sulfides
Organisms	primitive cell-like microfossils	spheroidal cell-like microfossils	complex bacteriological and animal communities
Organic structures & compounds	filaments; aliphatic hydrocarbons	filaments; aromatic kerogen*	filaments; (Thiocyanate [38])

*The Fig Tree, stratigraphically above the Onverwacht, contains aliphatic kerogens (Engel, et al., 1968).

TABLE 3. COMPARISON OF THE PROPERTIES INHERENT IN THE VARIOUS HYPOTHESES FOR THE ORIGIN OF LIFE

HYPOTHESIS	EARLY ATMOSPHERE	ENVIRONMENT	ENERGY	KINDS OF GRADIENTS	EARLY MICROORGANISM	TIME TO EVOLVE ACTIVE "PROTOCELLS"
Oparin model	Reducing (gases)	"terrestrial" soup	UV, electric discharges	None	anaerobic heterotrophs	> 10^9 year
Panspermia	"	directed or non-directed cosmic source	Moot	T only*	anaerobic heterotrophs or phototrophs	preformed
Hydrothermal model	"	hydrothermal gradient in seawater	Heat	T, pH, chemical concentration	anaerobic chemoautotrophs	instantaneous reaction from polymers to "protocells"

*Irvine, W. M., S. B. Leschire and F. P. Schloerb, 1980, Thermal history, chemical composition and relationship of comets to the origin of life, NATURE 283: 748-749.

A Comparison of Hypotheses for the Origin of Life

Deep sea hydrothermal vents provide all of the conditions for the formation of both simple and complex reactive organic compounds, of the biochemically important polymers, and of Fox-like "protocells." The various reactive gases, H_2, CH_4, CO_2, NH_3, and H_2S, and the biochemically active metals, Fe, Mo, Cu, etc., which are considered to be necessary for the formation of important biochemical compounds, are continuously provided to the hydrothermal environment through outgassing and the interaction of vent waters with magma and newly-formed rock. All of the other currently-favored models for the origin of life lack one or many of the conditions necessary to make the transition from the synthesis of organic compounds to the formation of "protocell" structures. Table 3 offers a comparison of our model with the model of Oparin and the theory of panspermia. Many models, including Oparin's, picture an aquatic environment which contains high concentrations of organic compounds formed through the input of UV radiation or lightning discharges which somehow react to form larger molecules. Eventually these larger molecules are formed into "coacervates" or "protocells" that metaphysically acquire the capacity to "transport" organic compounds through a highly organized membrane and to carry on oxidation/reduction and synthetic reactions. These quasi-heterotrophs, as a result of continued exposure to ultraviolet light, develop other structures, including photon-absorbing porphyrins.

This "organic soup" hypothesis predicts that, before the formation of biochemically active "protocells," a protoenvironment would have to be formed which contained a high concentration of amino acids and other organic compounds. If such an environment had existed, very ancient sediments should contain detectable levels of these amino acids and organic compounds. This is not the case. Instead, in the oldest rocks known to exist, which formed shortly after the cessation of giant impacting [4,27], fossil structures have been found which strongly resemble the budding "protocells" described by Fox as resulting from experiments using thermal energy. There is no detectable sedimentary evidence for a pre-existing "organic soup". It is also important to point out that, in an aquatic environment, the concentrations of organic compounds would be quite dilute except at the site of synthesis, and heterotrophic organisms could not survive under these conditions. Consequently, the suggestion that the first protist was heterotrophic does not seem to be supportable.

Another problem with the "organic soup" model is that it is known that the conditions necessary for the formation of amino acids and of low molecular weight reactive organic compounds are different from the conditions required for the formation of macromolecules and "protocells." The "inorganic soup" hypothesis has all these processes taking place in the same vat under the same conditions. However, in our model for the origin of life in submarine hydrothermal systems, the rapid and continuous upward flow of fluids creates gradients of temperature, pH, and chemical concentration in which all of the synthetic reactions needed for the creation of life could take place.

Many biochemically active macromolecules contain various metals, particularly iron, molybdenum, manganese, copper, etc., as part of their structure. Molybdenum, for example, is important in various biochemical processes including the fixation of nitrogen and the reduction of nitrate.

It has been hypothesized that early in the evolution of cells or cell-like structures, metallo-proteins, including enzymes, were formed from simple polypeptides and that these early macromolecules were active, although inefficient when compared to present analogous compounds [31]. The importance of molybdenum in biochemical processes and the apparent scarcity of this metal in terrestrial environments has been used to support the hypothesis that the first microorganisms on earth originated from an extraterrestrial source where molybdenum was abundant [32]. However, the concentrations of molybdenum and other biochemically active metals are not limiting in the present ocean, and it is generally accepted that this was also the case in the early ocean [33]. Hydrothermal alteration of oceanic crust has been the primary source of these metals to both the ancient and the present oceans.

The synthesis of amino acids from gases by thermal energy has been repeatedly demonstrated in the laboratory (see Lemmon, 1970) [34]. The fact that very high temperatures (800° to 1000°C) are required to form amino acids has been a criticism of the hypothesis that life could have originated in high temperature environments. This is because the continued exposure of both low molecular weight organic compounds and polymers to high temperatures after formation leads to their rapid decomposition. It would take only minutes to denature complex protein structures at temperatures greater than 100°C. The temperature gradient of hydrothermal environments provides a natural solution to this problem.

It is also possible that many of the amino acids, particularly those with low molecular weights, are formed at much lower temperatures, given appropriate chemical conditions. It has been demonstrated, for example, that if, besides the presence of the usual gases, reactive compounds such as hydrogen cyanide and formaldehyde are available, amino acids can be synthesized at temperatures between 80° and 100°C [34]. Amino acids have also been synthesized from formaldehyde and hydroxylamine at 105°C in seawater [35]. This process was found to be greatly influenced by the concentration of molybdenum in artificial seawater. In addition, three different "protocell" structures were formed when amino acids were heated at 105°C in a modified seawater solution at pH 5.2 [36].

However, the synthesis of the high molecular weight amino acids, such as tyrosine and phenylalanine, from gases requires temperatures close to 1000°C. High temperatures (600°C to 950°C) are also required to form sulfur-containing amino acids from hydrogen sulfide [37], which is one of the most abundant gases formed in hydrothermal vents. Although oceanic hydrothermal waters have not been analyzed in order to detect hydrogen cyanide and formaldehyde, Dowler and Ingmanson [38] recently reported the discovery of thiocyanate in Red Sea brines.

In view of these experimental results, we believe that it is highly probable that amino acids are formed in hydrothermal environments over a wide temperature range, between roughly 100°C and 1000°C. These varying conditions exist in hydrothermal systems and the spatial distance of the hydrothermal temperature gradient is relatively short.

The formation of peptides and other organic polymers from low molecular weight intermediate compounds has also been shown to occur under varying conditions and at temperatures between roughly 150°C and 200°C. At temperatures lower than 100°C, the presence of polyphosphoric acid can initiate polymerization [14]. In the Fox recipe for the formation of "protocells", amino acids must be heated to 200°C or 300°C under dehydrating conditions [2]. However, in addition to the dehydration reaction described by Fox, it is possible to effectively remove water from amino acids and form peptides through the use of various reactive compounds, such as cyanamides and carbodiimides [39]. These compounds have been shown to initiate the condensation of amino acids to peptides and of certain purines and pyrimidines to nucleotides. The reactions proceed optimally under drying conditions at low pH and at temperatures between 60° and 100°C [40]. These conditions exist in the hydrothermal vents, including a low pH due to the presence of hydrogen sulfide. Although there are no published reports on the possible existence of condensing compounds in hydrothermal environments, the necessary reducing gases are present in the required concentrations in order for the synthesis of these compounds to readily take place.

The First Organisms

Earlier in this paper we discussed how "protocells" containing hydrothermally-derived organic compounds could have formed in very high numbers within the vents and would have been carried out into cooler ocean waters by circulating hydrothermal fluids. There is evidence that this process is still going on in the present vents since the most abundant groups of chemoautotrophic bacteria isolated from both the Galapagos and 21°N have a temperature range for growth from a minimum of 10° to 20°C to over 70°C (the higher minimum and maximum growth temperatures are from bacteria isolated from 21°N samples) [41]. Since the ambient water temperature around the vents is approximately 2°C these bacteria would be incapable of growth outside of the vents proper. The fact that a significant portion of the primary producers in these environments is found within the vents definitely underscores the efficiency of hydrothermal gradients in sustaining life.

In a hydrothermal system, it is highly probable that most of the amino acids and other organic compounds would be condensed or dehydrated into polymers and "protocells" shortly after their synthesis. Consequently, it does not seem likely that there would be an accumulation of soluble organics as in the Oparin model. Instead, *within* the newly-formed protocells, the synthesis of high molecular weight compounds would probably continue due to the inclusion of low molecular weight organic condensing compounds, reactive gases and reduced metals. Any high molecular weight substances internally synthesized would be unable to pass out of the early cells. This strongly implies that the first protocells and ultimately the first protists were *anaerobic chemoauto-*

trophs, organisms which could utilize the hydrothermally delivered gases such as CO_2, H_2, NH_3, H_2S (and perhaps sulfate or some other oxidized form of sulfur), and HCN with the reduced metals to carry out internal oxidation/reduction reactions. Eventually, biochemically active macromolecules and energy transforming compounds such as NAD and ATP would be formed. The use of inorganic gases and metals as energy sources in these early organisms would solve one of the major problems in current research into the origin of life: explaining the formation of the complex membrane required by heterotrophic organisms for the transport of organic compounds. It would also explain how these early heterotrophs would survive in an oceanic environment where organic compounds would be quite dilute and how they could perpetuate before evolving the complex macromolecules needed for division by binary fission. Fox [2] showed that "protocells" produced by heat were capable of budding and forming chains of cells, and Pflug and Jaeschke-Boyer [30] found fossil evidence of budding cells in the Isua rocks.

It is also quite conceivable that these early "protocells" were capable of reducing CO_2 and sulfate to methane and sulfides through the use of hydrogen. This implies that the earliest organisms were methanogens and related "archaeobacteria." The molecular analyses of tRNAs, rRNAs, and cell wall components of various species of methanogens not only indicate that they are distinctly separate from most of the other procaryotes but also that, as a nutritional group, they are markedly heterogeneous [42]. Balch, et al. [42] state that the apparent rate of change in the sequence of RNAs is more rapid in methanogens than in other groups of bacteria, such as the "eubacteria." Another possible interpretation of these data, in keeping with our hypothesis for the origin of life, is that many separate groups of methanogens, each developing somewhat different molecular structures and morphologies, evolved over some period of time and from different hydrothermal environments. It is conceivable that the most common present-day group of procaryotes, the "eubacteria," could have evolved from just one of the separate groups of methane producers.

CONCLUSION

In this paper we have drawn together data from diverse scientific disciplines to show that hydrothermal systems provide an ideal environment for the thermal abiotic synthesis of complex organic compounds and simple cell-like organisms. This hypothesis is compatible with the geology and paleontology of the Archaean and with the results of abiotic synthesis experiments. We are proposing that abiotic synthesis occurred as an integral part of the origin and evolution of the atmosphere, ocean, and crust.

We believe that the early Earth was a complex, evolving system. The system was probably driven then, as it is now, by mantle convection through the mechanism of plate tectonics. Then, as now, new crust formed along submarine rift systems and was cooled, degassed and metamorphosed through reaction with seawater. We believe that we have convincingly demonstrated that organisms were created as a result of this reaction.

One of the unavoidable conclusions to be drawn from our hypothesis is that the events leading to the formation of complex organic compounds and "protocell" structures are still occurring in present-day oceanic hydrothermal systems. However, the complex communities of bacteria in modern oceanic environments would outcompete and consume any abiotically synthesized protocells, preventing their evolution into more organized entities.

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APPENDIX TO "Submarine Hydrothermal Systems:

A Probable Site for the Origin of Life"

The foregoing paper describes a model for the abiotic synthesis of organic compounds and of "protocell"-like structures in submarine hydrothermal systems. Some objections have been raised with regard to three major issues:

1. Our use of and reference to the experimental work of Sidney Fox and his colleagues;
2. Our reference to a paper by Pflug and Jaeschke-Boyer describing simple cell-like structures in Isua rocks;
3. The unproven nature of many of the postulated reactions, especially our suggestion that formaldehyde, hydrogen cyanide, and/or other catalystic compounds may be present.

We do not feel that any of these objections seriously undermines our model, but we wish to discuss them in order to make our case and because we realize that many readers may have similar objections.

We have discussed Fox's model and the research results of Harada and Fox in our paper. An important point which we do make, but which seems to need reiterating, is that the model for abiotic synthesis using thermal energy and most of the research based upon that model were presented and evaluated <u>before</u> submarine hydrothermal systems and their complex biologic communities were known to exist, in fact before plate tectonics was taken seriously. Consequently, the early estimates of the amount of thermal energy due to volcanism available on a primitive earth are gross underestimates. Additionally, Fox's model assumed a continental --not an oceanic -- environment. Given the historical geology then generally accepted, the geologic environment he chose was plausible to some degree.

In the last six or seven years, the processes of hydrothermal metamorphism along spreading ridges have been intensely studied. The resulting revolution in ore geology is ongoing. What we are now proposing is an equivalent biological revolution. The abundance of thermal energy in submarine hydrothermal activity, the degassing of magma which occurs, and the natural pumping system which is found to occur in hydrothermal vents were previously unknown to the researchers and experimentalists seeking to understand the origin of life. We believe that the geologic environment which provides the closest analogue to the environment required for Fox's model of abiotic synthesis using thermal energy is a submarine hydrothermal system.

There are some obvious differences between the Fox model and ours. The most important difference is that the Fox model requires anhydrous conditions for the polymerization of amino acids. We propose that a similar result could be achieved with the catalytic action of clays [23] and the presence of cyanamides and carbodiimides [39]. It is true that these reactions and the presence of these compounds have not been demonstrated, but our model provides a theoretical framework within which experiments can be designed and carried out in order to test those proposals.

It is interesting to note that quartz has often been used as a substrate in thermal abiotic synthesis experiments [14]. This is not unreasonable if one is attempting to reproduce continental volcanic conditions. In our model we propose that the clays produced by the seawater alteration of basalt will act as substrates. We are intrigued by the possibility that clay substrates may be more hospitable to α-amino acids. Lawless and Boynton (Nature, 234, 405, 1973) reported on their attempt to synthesize amino acids using thermal energy. They used quartz as a substrate and obtained a larger proportion of β-amino acids relative to earlier experiments. We feel it would be most interesting to reproduce the experiment using clays.

We are aware that the work of Fox has been controversial, especially with regard to the significance of microspheres. We feel that our model provides an attractive and plausible environment in which thermal energy is abundant and in which a process analogous to the Fox model could reasonably occur.

One objection to the use of thermal energy in abiotic synthesis experiments is that the process has a "low yield" of amino acids (although there is some question whether it is truly "low yield"). The "organic soup" model, because it depends on achieving rather high concentrations of complex organic compounds, requires a "high yield" process of abiotic synthesis. However, the debate over "low yield" versus "high yield processes loses its meaning in the light of our model. The continuous process which we are proposing eliminates the need to build up high concentrations.

With regard to the second objection to our model, we have become aware that the work of Pflug and Jaeschke-Boyer is controversial. Their discoveries are a serious threat to ideas which have been long held dear by many persons. We do not wish to enter into a debate on the validity of their work. We do find it intriguing and interesting because our model explains their discoveries so readily. If a person's preconceptions are seriously threatened by the work of Pflug and Jaeschke-Boyer, then our model, which provides a plausible explanation, is an even greater threat. In the field of research on the origin of life, certain ideas have been dominant for so long that they have practically become dogma. It is not good science to prefer dogma to experimental evidence.

That leads us logically to the third objection, that the reactions we postulate have not been proven to occur in hydrothermal systems. Many, many times in science, a reaction is postulated based upon observed conditions. There is nothing unusual about postulating the occurrence of a reaction. We do not claim to have proof. We are proposing a hypothesis. It is foolish to dismiss a hypothesis because it is "unproven". Scientific hypotheses are never proven; they merely withstand attempts to disprove them. A valid scientific hypothesis is one which says so much about the world that it is almost certain to be disproven as our knowledge of nature progresses. A useful hypothesis is stated in such a way that we can readily define experiments or observations which have the potential of falsifying the hypothesis (cf. Karl Popper or Paul Feyerabend). We look at our model and see many instances where experiments can be designed in order to test it; therefore, we firmly believe our hypothesis is valid and useful.

On the other hand, we are aware that, in our enthusiasm for our ideas, we stated many of our speculations as fact. We fell out of the habit of scientific circumlocution and circumspection. We are preparing a revision of the manuscript which will correct these lapses. In the long term, time will tell whether our hypothesis survives the attempts made to disprove it. We have a great deal of confidence that it will.

ACKNOWLEDGEMENTS

Support for this research was provided by the people of the United States through the National Science Foundation: International Decade of Ocean Exploration Office Grant OCE 75-23352 and OCE 77-23978 (Bruce Malfait, Program Manager) and Oceanography Section Grant OCE 78-26368 and OCE 79-27283 (Neil Anderson, Program Manager; Robert Wall, Section Head), all made to Oregon State University.

Many thanks to G. Ross Heath for providing indispensable support. Charles Miller reviewed the manuscript and offered pertinent critical advice and encouragement. John Edmond, Erwin Suess, Lou Gordon, Marv Lilley, Jack Dymond, Roger Hart, Mitch Lyle, Chris Moser, Kathy Fischer, and many others provided important insight, new information, data, and support. SEH wishes to thank Dr. Raymond Sullivan of San Francisco State University for introducing the topic.

Special thanks to Al Soeldner, Laboratory Manager of the Electron-microscopy Lab, for valuable assistance in obtaining the electronmicro-graphs.

Elaine Benson, Susan Harmon, Nancy Kneisel, Regina Tison, and Pam Wegner have been our patient secretaries and typists. Dave Reinert made the photo prints. Peggy Lorence drafted the figures.

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