

# Morphology: An ambiguous indicator of biogenicity

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*From a bird's-eye view, the General Cemetery looks like a laid down tree, enormous, with a thick and short trunk, made by the starting core of graves, from which four powerful arms starts, born consecutively, adjoining in born, but that later, by successive bifurcation, spread until disappearing from view, forming ..... a leafy crown, where the life and the death are indistinguishable (Saramago, 2000).*

## Abstract

This paper deals with the difficulty of decoding the origins of natural structures through the study of their morphological features. We focus on the case of primitive life detection, where it is clear that the principles of comparative anatomy cannot be applied. A range of inorganic processes are described, that result in morphologies emulating biological shapes. We focus on geochemically plausible processes, in particular, the formation of inorganic biomorphs in alkaline silica-rich environments, which are described in detail.

Whether unearthing dinosaur bones or ammonite shells buried inside ancient rocks, modern scientists harbour no doubts that they are observing the remains of organisms which were alive sometime in the past. It is considered self-evident that these structures are biogenic and no other explanation can be scientifically supported to explain their formation. However, sure as we are about the significance of these remnants, it is currently forgotten that it is not shape alone that informs us of their biogenicity.

Not much more than a century and a half ago, the origin of fossils was a source of confusion to even the most erudite scientists. The correct explanation for the genesis of these forms can be traced back to Greek and Roman times and to many Renaissance thinkers. Nevertheless, the genesis of these strange stones, whose shapes bore clear similarities to living organisms, was a major controversy in eighteenth and even nineteenth century Europe, comparable (and certainly related) to that between catastrophist and uniformitarian theories, then advanced to account for the history of the Earth. Scientists supporting the biogenic origin of fossils were subjected to ridicule; elaborate hoaxes were perpetrated to discredit amateur collectors, such as the celebrated “Lügensteine” of Würzburg – hand-carved life-like shapes in sedimentary rocks – that brought Beringer undone in the eighteenth century (Jahn and Woolf, 1963). The controversy was settled definitively by Baron de Cuvier, who introduced the brilliant concept of comparative anatomy (Tassy, 1991). Rather than appealing to their morphology in isolation, Cuvier demonstrated that fossils were indeed remnants of living organisms because fragmentary components could be reconstructed as elements of a jigsaw puzzle to make a unitary form. Cuvier’s *coup de grace* proved that fossils were not purely inorganic sports of a mischievous Mother Nature. Comparative anatomy made it possible to distinguish between true fossils and the pieces of any fictional bestiary. Subsequently, the development of a science of paleontology led to a definitive construction of geological time and fossils became a central tool for reconstruction of the history of life on Earth.

Until the 1960’s, (Tyler and Barghoorn, 1954), (Barghoorn and Tyler, 1965) identifiable fossils were restricted to the last six hundred million years before the present (Schopf, 1983). The entire fossil record at that time, from the late-Proterozoic Ediacara explosion to the evidence of human remains in the Pleistocene, consisted of remnants of organisms displaying compartmentalization and differentiated functions. Even the tiniest remnants, microfossils carefully studied by petroleum geologists, were shelled organisms with morphologically sophisticated carcasses (mainly made of silica or calcium carbonate), surely derived biologically. Fossil detection continued without a hiccup, and the induced sense of certainty allowed the concept of morphology to creep in – once again – as a tool for detection of biogenicity. Complacency bred an ignorance of history, and Cuvier was all but forgotten. Indeed, morphological evidence moved to centre stage. The great concept underlying morphology, symmetry, began to be used to divide the world into two large groups: the realm of the inanimate and the realm of the living; the cold, faceted world of minerals and the uncertain world of sensuous curves of the living (Weyl, 1952). Though it is rarely explicitly announced, there is no doubt that contemporary scientists concerned with origins of life remain inculcated with the false notion that the living and inorganic realms can be distinguished on the basis of morphology.

That division lies at the heart of the controversy of morphological measures for detection of primitive life, a problem crucial to extraterrestrial life detection and to the timing of the origins of life on the Earth (McKay et al., 1996a), (Cady, 1998), (McKay et al., 1996b), (Buick, 1990), (Knoll, 1998). When looking for primitive life, the principles of comparative anatomy *cannot* be used because – by definition – we are looking for remnants of simple, unshelled and uncompartimentalized structures. These forms are sufficiently uncomplicated to be confused with tiny mineral precipitates. The literature launched by the study of the Martian meteorite ALH84001 is replete with assumption that the shapes of life forms and of minerals are distinct. For example, the NASA team – in support of their claim of nanofossils in the Martian meteorite ALH84001 – write, “*Some of the features in ALH84001 (e.g. filaments) are common biogenic markers on Earth. We conclude that the evidence for fossilized microbes and their products is not conclusive, but cannot be readily explained by nonbiologic (sic) processes, and therefore should not be ignored* (Gibson). In part, for the reasons presented in this paper, that assumption is unfounded. Our own objections are echoed by others, and today there is extreme scepticism of the claim for Martian life on the basis of the ALH84001 analysis (for an overview, cf. Treiman).

Crystals are today considered to be inanimate aggregates, though it is arguable that they do satisfy some of the essential characteristics of life, provided “life” is not defined (circularly) in terms of its chemistry (Cairns-Smith, 1982). Indeed, the father of the concept of morphology, Goethe, considered inorganic crystals to be alive while they grew, and distinguished between “organic life” and “inorganic life”. That attitude is dismissed today, largely on the basis of the lack of (structural or information entropic content) complexity in simple, model crystals compared with biological organisms. The faceted forms of simple crystals are readily explained by simple structural principles of atomic packing (Amoros, 1978). Certainly, the classical domain of mineral morphology, for instance the view offered by *Dana’s System of Mineralogy* (Dana and Dana, 1962), comprises faceted single crystals, twins and fluid inclusions (negative crystals). However, the forms of inorganic crystals extend beyond that inventory of Platonic forms and planar polyhedra. To that catalogue, we must add, at the very minimum, ordered fractals (crystalline dendrites), spherulites and occasional random fractal dendrites, formed when the rate of aggregation controls the pattern formation (Hopkinson et al., 1998). Already, the distinction between biological and mineral form is blurred. Thus, the fern-like forms of “sick” sodium chloride crystals were advanced as evidence for life in crystals by the (otherwise) respected biologist, Ernst Haeckel, in 1917 (Haeckel, 1917). Haeckel, then the leading morphologist of simpler organisms, was also struck by similarities in microscopic form between liquid crystals and animate matter. The discovery of liquid crystals, thirty years earlier, by Reinitzer and Lehmann (Lehmann, 1889), posed a real riddle to morphologists, not dissimilar to that of the fossils-hunters a century earlier. In the optical microscope, liquid crystals were observed to spontaneously form radially symmetric “cells”, that budded and grew. Twisted, worm-like “myelin figures”, that bore astonishing resemblance to the optical micrographs of human nerve myelin, were also observed to grow *in vitro* (Haeckel, 1917). These phenomena led many scientists to the conclusion that liquid crystals were synonymous with life.

Today, liquid crystals are considered no more alive than conventional solid crystals, though it is becoming clear that the liquid crystalline state is often synonymous with biological self-organisation. The compartmentalisation capacity of

cell-membrane lipids, essential to living organisms, is the same feature that leads to the liquid crystallinity of these lipids: their dual hydrophobic-hydrophilic *surfactant* nature. Under appropriate conditions, many abiotic surfactants, for instance fatty acids and soaps (Deamer, 1997) arrange themselves in two-dimensional membrane-like structures (Luisi et al., 1999), including the microspheres of prebiological coacervates (Fox and Dose, 1977), (Orlovskij et al., 1982), (Hargreaves et al., 1971). These shapes can exhibit phase transitions and instabilities, from spherical and ellipsoidal shapes to more complicated patterns as discocytes, stomatocytes, echinocytes and necklaces of small vesicles. All these forms are due to locally lamellar self-organisation. Other local organisations are possible, including rods and three-dimensional “sponges”, dependent on the lipid-water solutions conditions (Hyde et al., 1997). These self-assemblies share a common structural feature with biogenic matter: they are smoothly curved, rather than Euclidean and faceted. At the molecular scale (around 10Å), they exhibit orientational ordering of the constituent *surfactant* molecules, with no translational order. Liquid crystals are, however, sometimes translationally ordered at the mesoscopic (supermolecular, *ca.* 100Å scale). The complex interplay between translational and orientational order at distinct length scales is a hallmark of biotic structures, yet (*organic*) liquid crystals demonstrate that this microstructural feature is not confined to the biological realm. The novel assemblies we describe below share this multi-scale orientational *vs.* translational ordering, hitherto assumed to be a signature of organic (and often biological) assemblies.

A further twist in the story of animate *vs.* inanimate form arose with Haeckel's and Lehmann's contemporary, Pieter Harting, who saw for the first time the extraordinary wealth of structural and material variety possible in composite materials. Indeed, Haeckel distinguished “biocrystals” from solid and liquid crystals by their form “produced by the united working of the inorganic mineral substance and the organic plasma” (Haeckel, 1917, loc. cit. p.9). Harting, who deserves to be considered the founder of the current synthetic biomorphic materials design push (exemplified by the abundance of articles in *Nature* and *Science* (see, for example, Mann and Ozin, 1996)) investigated the precipitation process of simple inorganic salts, grown in the presence of organic “impurities” (Ozin and Olivier, 1995). His interest was the mechanism of biomineral formation in animal skeletons, which were known to be chemically complex inorganic-organic composites. Harting's precipitates displayed remarkable diversity, and many of the features of biological morphology were reproduced, including calcitic spheres, septate “worms”, banded lamellae, dumbbells and semi-transparent gelatinous membranes (Harting, 1872). His work is significant, in that he showed that biomimetic forms could be precipitated *in vitro* in inorganic-organic *composite* materials. Nevertheless, his organic components were universally of biological origin, such as gelatin, albumen, blood, bile and chopped oyster meat. Harting's remarkable *biomorphs* were indeed inanimate, but their formation involved true biochemical species. The rediscovery of the amazing and yet not fully understood phenomenon of chemical gardens (Clunies Ross, 1910), introduced at the beginning of the 20th century, opened another route in the search for biomimetic structures. The work on the formation of these osmotic-driven inorganic tubules and related structures resulted in a strongly mechanistic and Lamarckian approach to the origin of life, as illustrated by the work of Leduc (Leduc, 1911) and the “*synthetic biology*” school in Europe, and the work of Herrera (Herrera, 1910) on *plasmogeny* in America, although both authors also thoroughly explored the use of biochemicals (Herrera, 1942). The development of biochemistry and the foundation of cell theory, closed forever that mechanistic approach

to understanding the nature of life. However, that imaginative work yielded a huge assortment of biomimetic structures, driven by novel physical phenomena. The morphogenesis of inorganic materials was extended far beyond the classical mineralogical and materials science view.

From a current astrobiological perspective, a century later, the possibility of inorganic structures mirroring so closely biological morphologies and textures is indeed impressive and surprising, but there is no evidence of any relationship between the processes controlling these structures and life itself.

Most of these structures are certainly formed by physical and chemical instabilities and coupling of reaction-transport non-linear processes and their study is a challenge for today's materials scientists. A limited repertoire of physico-chemical processes lead to these morphologies. Among them, Liesegang-type patterns driven by coupling between precipitation and diffusive mass transport (Henisch, 1988), chaotic mixing of fluids (Ottino et al., 1988), (Ottino, 1989), salt efflorescences, and the well-known manganese and iron oxide dendrites formed by decoration of Saffman-Taylor interfaces (Saffman and Taylor, 1958) strongly emulating the forms of fossil plants (Ball, 1999), (García-Ruiz et al., 1994). A most beautiful description of this last fractal structure, very appropriate in the context of this comment, is the General Cemetery imagined by Saramago, a random structure cited in the foreword. The spectrum of materials produced by these phenomena may be vast and in many cases totally irrelevant to primitive life detection. Therefore, it is of primary importance to demonstrate the formation of non-crystallographic structures under physico-chemical conditions characteristic of geological environments of the primitive Earth.

In what follows, we focus on a specific type of biomimetic structure which we are currently studying. These materials, that form spontaneously, bear a remarkable resemblance to those commonly considered to be microfossils, and the conditions under which they are grown are not dissimilar to the chemistry of geological formations of other eras, particularly Archean (or Hadean) times. Our materials are sparingly soluble carbonates crystallised in the presence of silica (present as one or more amorphous silicate species) under alkaline pH conditions. In the presence of silicate, the morphology of barium and strontium carbonates is transformed in a remarkable way (García-Ruiz, 1998). We focus here on the barium salt.

The experiments are trivial. Their chemistry is related to that of silica gardens but the morphologies and textures of the resulting precipitation products are quite different. A solution of soluble  $\text{BaCl}_2$  (typically 0.01M concentration) is introduced to a silica sol or gel. The silica solution is prepared by diluting water glass to the desired concentration (viscous  $\text{Na}_2\text{SiO}_3$  solution, pH ~11.8). The pH of the mixture is adjusted by the addition of small aliquots of HCl or NaOH solution. To form our biomorphs, the pH of the silica sol or gel needs to be maintained at pH values in the range of 8.5-11. We have prepared the samples in cells of various types, from (microscope slide) glass sandwiches of variable thickness (between 2mm and 5 cm) to plastic circular wells of about 5 mm diameter and 5mm depth, to Petri dishes of diameter 10 cm. The observed morphologies are not dependent on the geometry of the sample cell, or proximity of growth to the cell wall.

The morphologically interesting aggregates that form in these glass or plastic cells contain as major constituents the sparingly soluble compounds barium or strontium carbonate. Slow diffusion of CO<sub>2</sub> into the cell leads to a build-up of carbonate ions at the elevated pH values. Eventually, the concentration of Ba<sup>2+</sup> and CO<sub>3</sub><sup>2-</sup> species leads to precipitation. The kinetics of precipitation depend on the silica concentration, temperature and pH of the mixture. At room temperature, precipitation in the sols commences within a couple of hours, and the morphologies evolve over a period of approximately one day to give stable forms that have been preserved for months without change. The morphologies of the precipitates depend critically on the pH of the system. We focus here on the pH window that yields the most biomorphic aggregates, typically around pH 10.5. The aggregates have been extracted from the glass or plastic cells, and examined by optical microscopy, SEM (the Hitachi S4500 Field Emission Scanning Electron Microscope (FESEM)) and X-ray diffraction (Nonius Kappa 4-circle diffractometer with CCD).

The growth process of gel-grown aggregates appears from optical microscopic observations to proceed in two stages (Figures 1). The first precipitates, that form closest to the site of salt introduction and consequently made at the lower pH range (8.5 < pH < 9.5), are often fractal-like dendritic forms with an approximately radial symmetry showing continuous non-crystallographic branching whose diameter is at the scale of mm. The resulting cauliflower-like morphologies are made of a radiating array of barium carbonate crystals, which successively branch at small, non-crystallographic angles, forming a 3-D space-filling structure. The building blocks of these structures are micron-size crystals. This 3-D morphology can be understood as the result of the crystallization of witherite crystals in the presence of polymeric silica which is pushed ahead of the growing crystal, forming an impurity boundary-layer and thereby triggering the observed fibrillation, as explained by Keith and Padden (Keith and Padden, 1963). A similar mechanism applies to low temperature aqueous solution growth of silicate cements (Williamson, 1968); similar forms have also been seen in other composite systems, such as fluorapatite-collagen growths (Kniep and Busch, 1996).

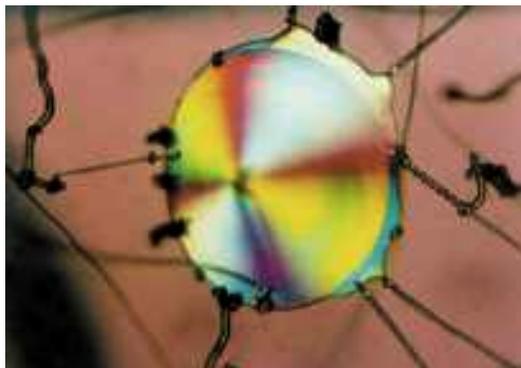
Later growths resemble those formed in the sol, discussed below. We characterise these structures at the  $\mu\text{m}$  scale first. Growths can be broadly classified as 3-D, 2-D and 1-D objects, with the dimensionality decreasing systematically as time proceeds. In the sol, where salt and silicate are mixed from the start, small clusters of deformed spherulitic aggregates (3-D) appear first, suspended from a thin film at the sol surface. The overall form of the clusters ranges from radially symmetrical, resembling a fleshy flower or star anise fruit, to irregular globules. Spherulites then sprout complex arrays of intergrown sheets, often reminiscent of the cell walls in a foam. It is worth noting that the sheets are occasionally observed to meet along triple line junctions, with dihedral angles of *ca.*  $2/3$ , as in soap films that adopt that this Plateau configuration (Aste and Weaire, 2000) under the action of surface tension.

After some delay, single sheets are extruded from the less regular clusters described in the previous paragraph. These sheets invariably emanate from the original clusters by selective growth of one member. These sheets have variable curvature, though slightly saddle-shaped ones predominate (negative Gaussian curvature). In some cases, trumpet-like forms are observed, that consist of sheets resembling the pseudo-sphere, a surface of constant negative Gaussian curvature. The margins of the

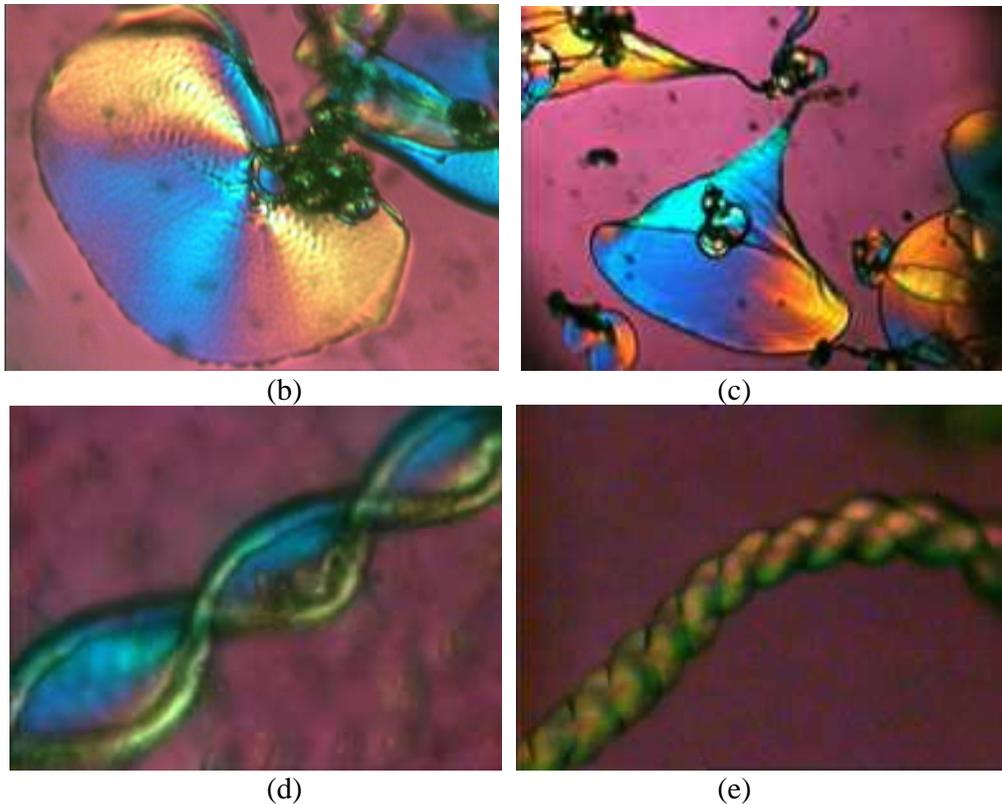
sheets often adopt cardioid forms, presumably due to slight azimuthal variation in radial growth rate. Rarely, perfectly flat sheets with circular rims are seen. Sheets grow in radius by accretion at the rim, evidenced by concentric corrugation bands, and their thickness does not appear to change significantly with time. Usually, the sheets eventually develop scrolled margins clearly evident as “lips” around the rim.

The sheet rim often shows one or more cusps that indicate discontinuities in curvature at the margin, and appear to mark the boundaries of distinct radial growth sectors. Cusps can also be created when two differently-curved sections of sheet lip confront one another as scrolling of the lip propagates around the sheet rim. At a cusp, there is a tendency for radial growth of the sheet to transform into linear (1-D) growth along the pointing direction of the cusp. If the sheet rim segments on each side of the cusp are curled in opposite senses, the change to linear growth preserves the chirality of the twist, and growth proceeds by a mutual winding of the two sheet lips about each other along the cusp axis. This stage of growth thus produces helically-wound rod-like objects, with cross-section normal to the long axis resembling a letter “S” or its reverse. Note that there is no preference for one enantiomer over the other – racemic mixtures are observed. These 1-D objects can be sub-classified into three principal morphologies (a broad classification, that simplifies a continuity of forms). They are distinguishable by the degree of overlapping of successive whorls of the helical form and the extent of lip curling. The first examples (“twisted ribbons”), contain a substantial helicoidal ribbon, twisted along the helix axis but flat normal to the axis, separating the tightly scrolled lips; these resemble intertwined ropes in the optical microscope. The second case has no distinct helicoidal central ribbon – it is almost all lip – and resembles a double helix. In the third case, the lips are large but less tightly scrolled, and overlap each other to the extent that only a single helical wind is discernible. These structures resemble the irregularly wound appearance of pastry croissants. Extreme overlap of whorls and shortening of helical pitch can result in such aggregates developing a spheroidal “head”, which could be regarded as a final reduction of dimensionality to zero. These helical forms are very similar morphologically to Lehmann’s myelins.

It is stressed that continuous evolution from one extreme of the morphological progression to the other has been observed in single aggregates, but the progression appears to be continuous with time and presumably with change in growth conditions. Selected time-lapse growth (QuickTime) movies can be accessed on the WWW at [http://www.rphysse.anu.edu.au/~sth110/Astropaper\\_extra.html](http://www.rphysse.anu.edu.au/~sth110/Astropaper_extra.html).



(a)



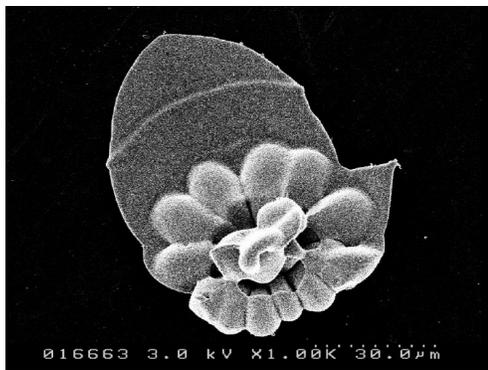
*Figure 1: Optical micrographs of silica-witherite aggregates, viewed between crossed polarisers (with a sensitive tint plate in place). The continuous colour changes within the aggregates is evidence of preferred orientations of optically anisotropic material within the aggregates (witherite crystallites). A variety of morphologies are found, dependent on the pH of the starting  $\text{Na}_2\text{SiO}_3 - \text{BaCl}_2 - \text{NaOH}$  solution and aging period between mixing and aggregate formation. (a) low magnification image, showing helical tubules emerging from a sheet (field of view ca. 1mm) (b) Initial stages of aggregate formation, showing saddle-shaped sheets growing from globular aggregates (600 $\mu\text{m}$  field of view). (c) trumpet-bell shapes sheet (600 $\mu\text{m}$  field of view) (d) close-up of a twisted ribbon, with a helicoidal sheet spanning double helices (90 $\mu\text{m}$  field of view). (e) Tightly-wound braid (90 $\mu\text{m}$  field of view).*

The material of which these forms are constituted is a composite. Energy-dispersive X-ray analysis (EDXA) in the FESEM has shown that both silicate and carbonate phases are present. These may be removed selectively by alkaline or acid dissolution respectively. X-ray and electron diffraction confirms the presence of crystalline carbonate (e.g. witherite,  $\text{BaCO}_3$ ) crystallites within the aggregates. Absence of Bragg diffraction from other phases implies that the silicate component is amorphous to X-rays. EDXA indicates that the amorphous silicate is not pure silica, but contains some large cations such as Ba and Na balancing the charge on an anionic polymer. Raman spectra are consistent with that structural assignment

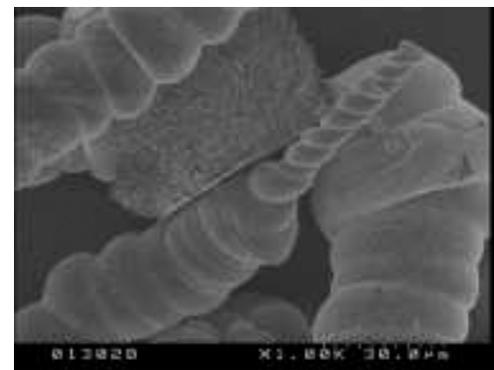
These micron scale morphologies exhibit complex self-organisation at still smaller scales. Observations of the aggregates through the optical microscope with crossed polars confirm the presence of a remarkable degree of orientation ordering

within the aggregates. The optically anisotropic barium or strontium carbonate crystallites are preferentially aligned to a degree that imparts birefringence to the whole aggregate. The orientation of the slow axes in the crystallites can be deduced from the interference colours observed with a sensitive tint plate in the microscope column. The continuous variation of colours within the aggregates indicates a continuously varying *orientation* of the carbonate crystallites with position. For example, the crossed-polar optical micrograph of the flat circular sheet indicates a continuous azimuthal variation of orientation, that is independent of the radial coordinate.

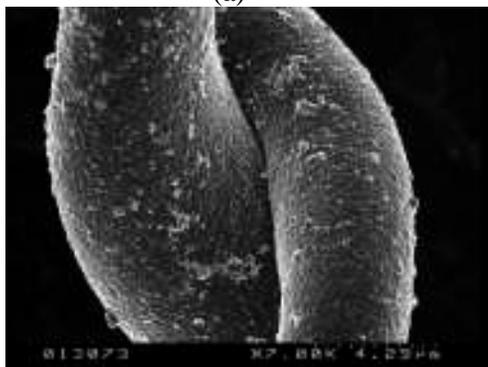
In the FESEM, aggregates appear to consist of a bulky core region that makes up the major volume fraction of the object, consisting of aggregated elongated rods, that describe oriented textures, occasionally overlaid by a distinct skin composed of a single layer of spheroidal and occasional cylindrical units, also displaying clear orientational order (Figures 2).



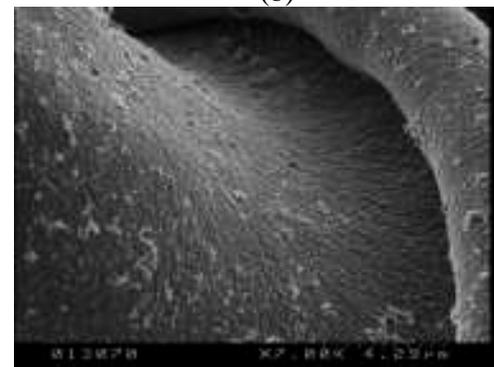
(a)



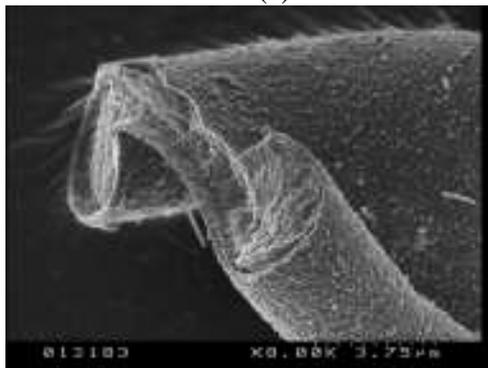
(b)



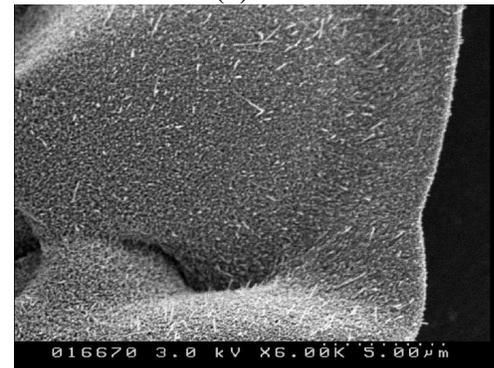
(c)



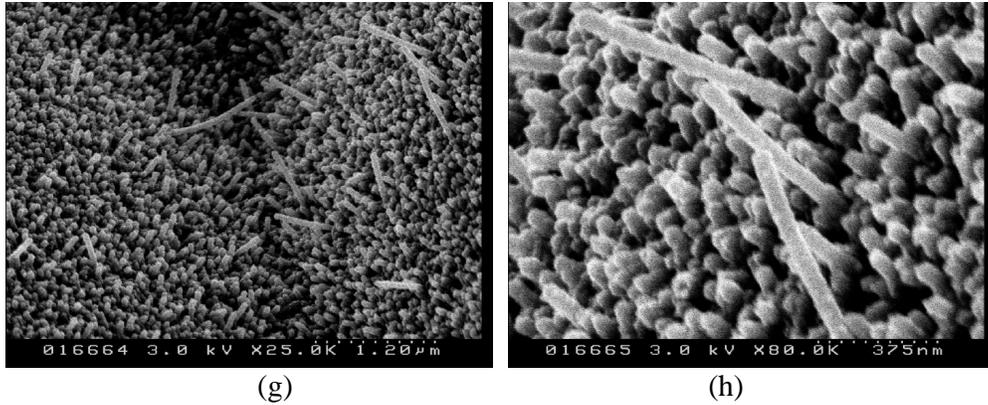
(d)



(e)

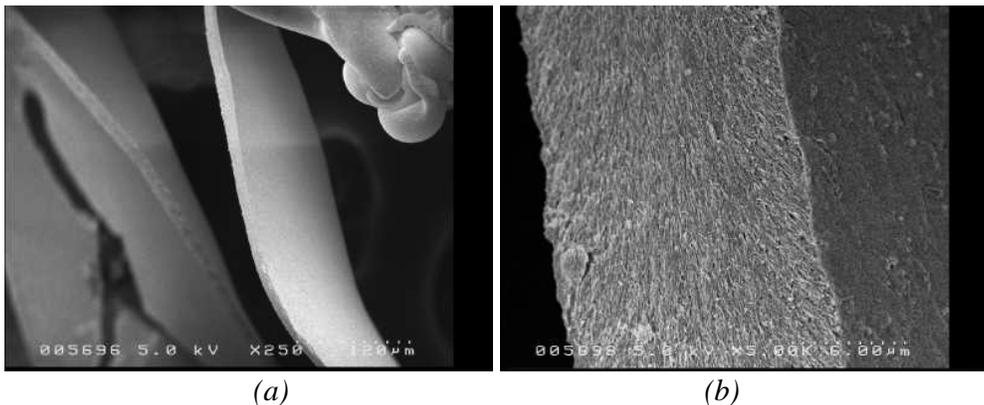


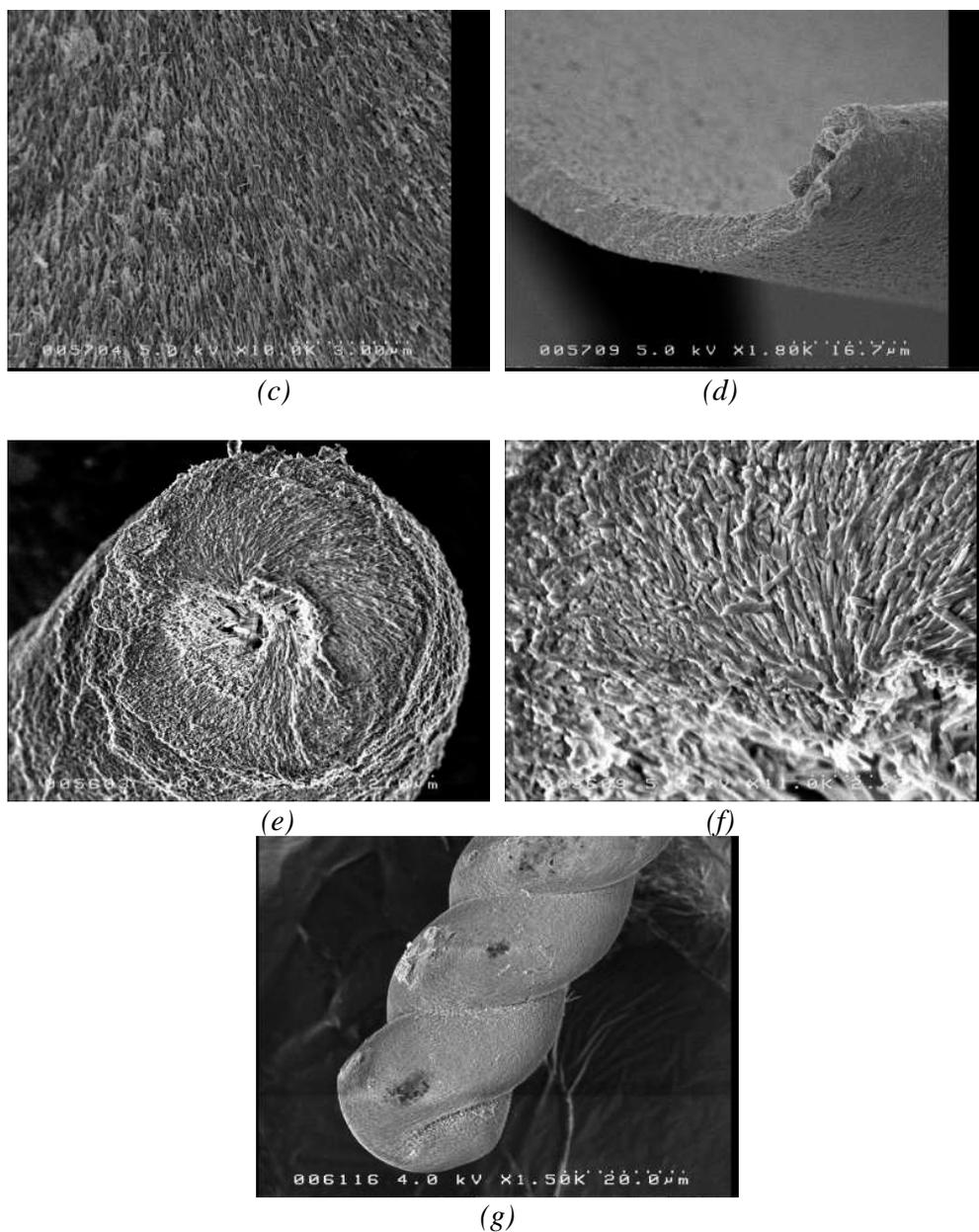
(f)



*Figure2: FESEM images of sol-grown silica-witherite aggregates, extracted from the sol by hand and dried (after washing in water and ethanol): (a) globular form with extruded sheet ; (e) sheets (with curled lips at their edge) and twisted ribbons of various pitch are formed (b-e). Images (f-h) are a sequence of increasing magnification, showing details of the surface texture.*

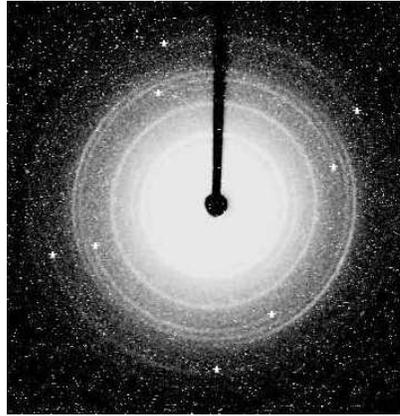
Closer examination indicates that the textured core of the aggregates consist of tightly-packed, approximately cylindrical forms, typically 2 nm wide and 200 nm long. These lie nearly parallel, though coherently misoriented, with respect to their immediate neighbours, giving rise to the observed texture in the optical microscope. The optical observations indicate that the long directions of the tubules are coaxial with the **c**-axes of the carbonate crystallites. The radial texture seen in optical micrographs is clear at the submicron scale, with continuous and slowly varying twist of the director with relative position in the aggregate. The degree of twist between adjacent rods is approximately uniform. In many cases, the rods are coated with amorphous silica skin. The degree of coating is dependent on the original silica concentration as well as aging time. The orientational ordering is better displayed in FESEM images of aggregates whose silicate has been largely removed by slow dissolution in NaOH solution (Figures 3).





*Figure 3: FESEM image of bare carbonate aggregates, with most adsorbed silica removed by immersion in dilute NaOH. (a-d) show images of sheets at various magnifications. The oriented twist texture is clearly evident, with bilateral symmetry in the flat sheet (“bilayer” texture) (a-c). The curled lip is accompanied by the thinning of one side of the bilayer (d). (e-g) show helical aggregates. The presence of a twist orientation field is also evident in these aggregates.*

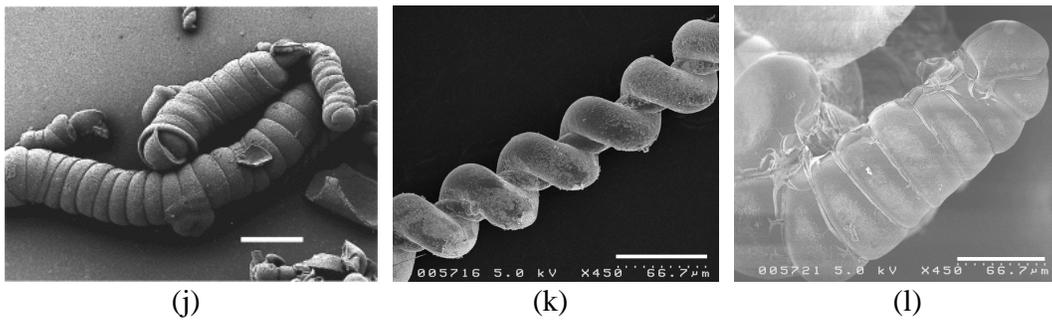
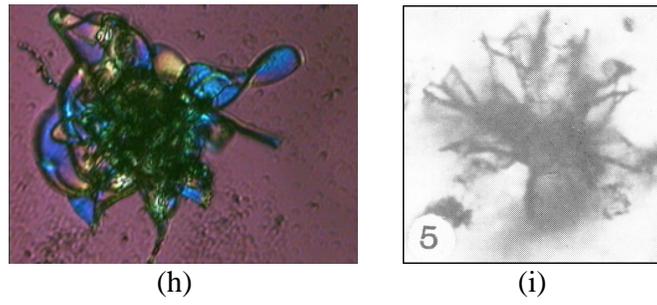
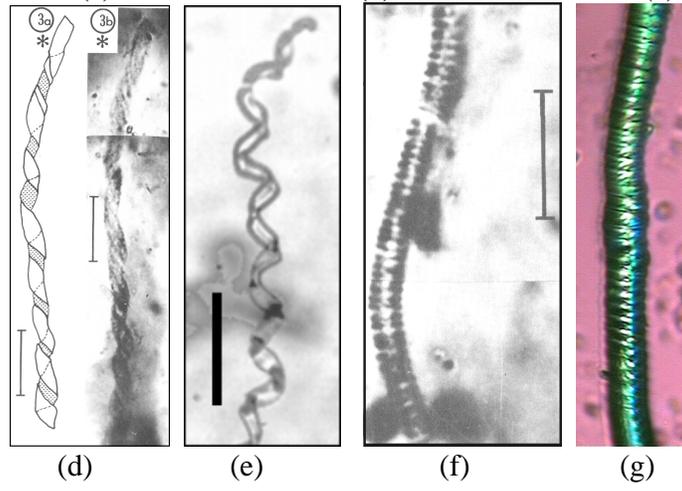
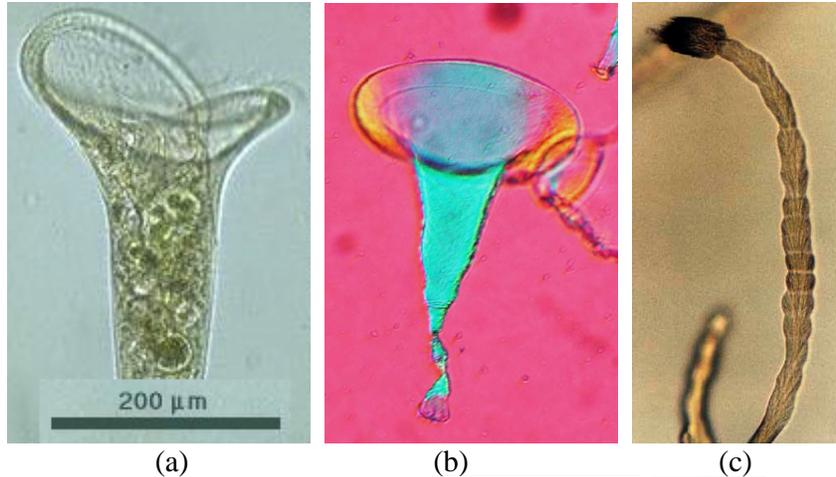
The orientational ordering is sufficiently strong that only a restricted range of crystallite orientations occur in the more symmetrical aggregates such as helically twisted ribbons and radial-textured sheets. X-ray diffraction patterns from carefully oriented stationary samples recorded on CCD plates show strong and limited arcing of some powder lines, consistent with this orientational correlation, rather than homogeneous and random crystallite orientations (Figure 4).

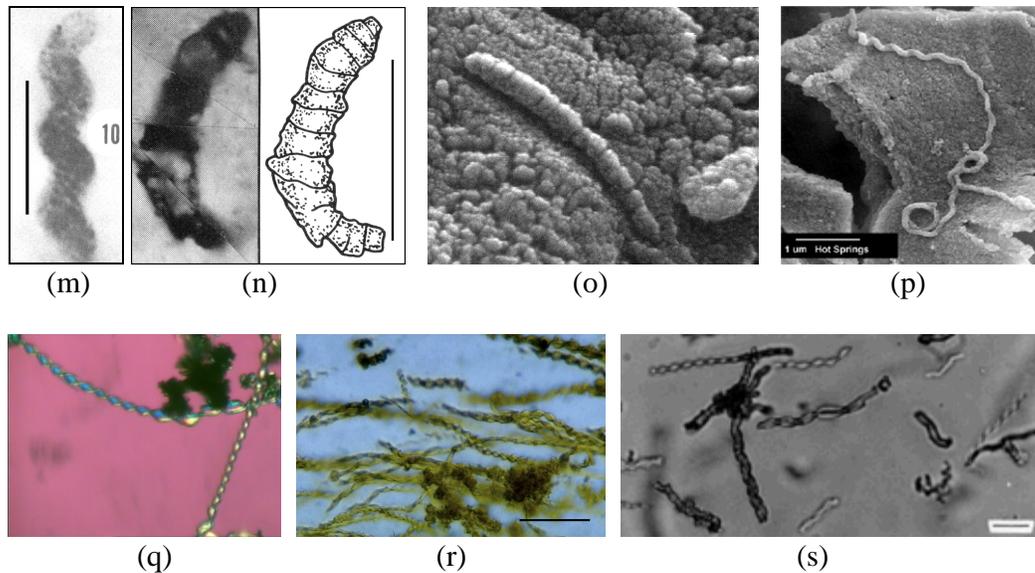


*Figure 4: X-ray diffraction pattern from a witherite-silica sheet. The diffraction lines can be indexed to the standard witherite crystal structure. The pattern is powder-like, indicating a range of orientations in the witherite crystallites. However, some lines exhibit a finite range of arcing (delimited by asterisks), while others extend through  $2\pi$ , forming full circles. That pattern is consistent with a strictly allowed range of crystallite orientations, set by the orientational order within the aggregate.*

The diffraction spacings index to conventional witherite, conforming the notion that these biomorphs are assemblies of witherite crystallites, coated (to a greater or lesser extent) with amorphous silica. Work is continuing to identify other stoichiometries that can lead to these biomorphs. It is noticed that the metal ions of carbonate salts tested that crystallise in the calcite structure under these conditions (Ca, Mg, Cd) fail to give similar behaviour. However, strontium carbonate, which is isomorphous with the aragonite-type structure of barium carbonate does give similar forms.

The twisted ribbons, sheets and trumpets bear a remarkable resemblance to simpler organisms, both ancient and modern. Some examples are shown in Figure 5. They are strikingly similar to the most primitive terrestrial microfossils (García-Ruiz, 1994), and their formation is geochemically plausible. The trumpet forms resemble the modern protist *Stentor*. Twisted ribbons and related forms are morphologically indistinguishable from putative Bitter Springs (Schopf, 1968) and Warrawoona microfossils from Australia (Schopf et al., 2002), (Ueno et al., 2001), filamentous goethite assemblages from Breiddalur, Iceland, presented as likely microfossils by Hofmann and Farmer (Hofmann and Farmer, 2000) as well as the modern bacterium *Gallionella ferruginea*.





*Figure 5: Animal, vegetable or mineral? A suite of images that demonstrate morphological convergence between living bacteria, putative microfossils and abiotic biomorphs (described in the text) precipitated in vitro.*

*(a) optical micrograph of protist Stentor roeseli (200µm scale bar) (reproduced and modified with permission from (Tsuuji));*

*(b) optical micrograph of a silica-witherite biomorph (ca. 100µm field of view);*

*(c) optical micrograph of a silica-witherite biomorph (ca. 600µm field of view);*

*(d) reconstruction and optical micrograph of the Bitter Springs (central Australia) late Precambrian microfossil Heliconema australiensis (10µm scale bar) (reproduced with permission from (Schopf, 1968));*

*(e) optical micrograph of a silica-witherite biomorph (200µm scale bar);*

*(f) optical micrograph of the Bitter Springs (central Australia) late Precambrian microfossil Calyptothrix annulata (10µm scale bar) (reproduced with permission from (Schopf, 1968));*

*(g) optical micrograph of a silica-witherite biomorph (ca. 60µm field of view);*

*(h) optical micrograph of a silica-witherite biomorph (600µm field of view);*

*(i) optical micrograph of the Middle Precambrian Duck Creek (Western Australia) microfossil Eoastrion (ca. 25µm diameter) (reproduced with permission from (Knoll and Barghoorn, 1976));*

*(j,k,l) scanning electron micrographs of silica-witherite biomorphs (scale bars 100µm, 67µm, 67µm resp.);*

*(m) optical micrograph of the Bitter Springs (central Australia) Late Precambrian microfossil Heliconema australiense (10µm scale bar) (reproduced with permission from (Schopf and Blacic, 1971));*

*(n) optical micrograph and reconstruction of the Apex chert (Western Australia) Archean microfossil Primaevifilum conicoterminatum (20µm scale bar) (reproduced with permission from (Schopf, 1993));*

*(o) scanning electron micrograph of a carbonate aggregate in the Martian meteorite ALH84001 (available at*

*<[www-curator.jsc.nasa.gov/curator/antmet/marsmets/alh84001/ALH84001-EM1.htm](http://www-curator.jsc.nasa.gov/curator/antmet/marsmets/alh84001/ALH84001-EM1.htm)>;*

*(p) scanning electron micrograph of a calcitic microbial filament collected from Hot Springs (1µm scale bar) (reproduced with permission from (Allen et al., 2000)) ;*

*(q) optical micrograph of a silica-witherite biomorph (600µm field of view);*

(r) *goethite filamentous microfossils (50 $\mu$ m scale bar)* (reproduced with permission from (Hofmann and Farmer, 2000));  
(s) *optical micrograph of the bacterium Gallionella ferruginea (10 $\mu$ m scale bar)* (available at <[pubs.usgs.gov/publications/text/Norrielist.html](http://pubs.usgs.gov/publications/text/Norrielist.html)>).

What is chemically required is an alkaline environment with high silica concentration and a moderate source of alkaline earth cations. Local scenarios fulfilling these conditions have been described in the contemporaneous Earth, e.g. the African alkaline lakes. Suitable geochemical environments are equally plausible at earlier stages of Earth's history, when basic silicates were dissolved by condensed water in the absence of silica removing organisms and organic matter (García-Ruiz, 2000). For example, recent detailed re-analyses and mapping of the Apex chert formation, containing the Warrawoona microfossils strongly point to a hydrothermal origin for this formation, with a preponderance of silica with barium and iron cations and sulfate and carbonate anionic species (Brasier et al., 2002). Clearly, formation of morphologically complex silica-witherite aggregates from that geochemical soup is eminently reasonable, provided the temperature and pH values are suitable. A recent analysis of a nearby deposit (also containing similar "microfossils") led the authors to conclude that the formation conditions were mildly alkaline (pH 7-7.5) with a formation temperature of ca. 200°C (Kojima et al., 1998). Such pH estimates are likely to be crude, but afford useful ballpark parameters for synthetic mimics. We have grown similar aggregates at atmospheric pressure, elevated temperatures and alkaline conditions. Although experiments to date have not exceeded 65°C, higher temperatures will promote silica-carbonate aggregation kinetics lowering the silica concentration threshold required to precipitate inorganic biomorphs. Clearly, the geochemistry of the Apex chert genesis is compatible with the genesis of microfossil-like silica-witherite structures. The morphological convergence of Warrawoona microfossils with witherite-silica aggregates can be accounted for invoking only inorganic processes. A number of authors have pointed out that in addition to *morphology*, *monodispersity of size* is characteristic of biogenic materials (see, for example, Hofmann and Farmer, 2000). That criterion too, fails to distinguish silica-witherite biomorphs from microfossils. The dimensions of biomorphs depend on the physico-chemical environment during precipitation. Thus, the diameters of ribbons found in sol-grown aggregates (whose chemical environment is more controlled than found in gels) are tightly constrained for fixed pH, concentrations, etc.

Modern analytical chemistry techniques are sufficiently accurate to allow for *in-situ* microchemical analyses, including "molecular fossil" recognition by mass spectrometry (Brocks et al., 1999) and micro-Raman spectroscopy (Schopf, 1968), (Brasier et al., 2002). This offers further characterisation of putative fossils, complementary to morphology. Recent Raman spectral mapping of Warrawoona samples has established a carbonaceous, kerogenous stoichiometry for the microfossils (Schopf et al., 2002), (Brasier et al., 2002). Much debate has been occasioned by the stoichiometric origin of a broad Raman peak (ca. 1350  $\text{cm}^{-1}$ ); a debate that we defer to others. To date, the only certain conclusion is that the microfossils comprise polymeric organic (carbon-rich) species, such as condensed polyaromatic hydrocarbons (PAH's), amino acids, or perhaps living tissue residue.

The presence of carbonaceous material in inorganically fabricated silica biomorphs can be explained without difficulty, given that polymeric silica is a known

trap for organic compounds. Indeed, preliminary data offers a reasonable scenario – involving only physical processes – that is consistent with the morphology *and* the stoichiometry observed in (e.g.) Archean putative microfossils. The morphogenesis involves a number of stages, the first of which is the witherite-silica precipitation described above. Two further steps ensue, whose chronological order is indifferent, account for the presence of carbonaceous matter in the aggregates. One is the dissolution of the witherite crystallite “template”, achieved in the laboratory by the addition of mild acid (eg. 0.03M acetic acid solution). It is worth emphasising that dissolution occurs *without any alteration* of the morphology; a silicate sheath remains that retains the complex sinuous form of the original aggregate (Figure 6). (The complete sequence (QuickTime movie) can be accessed at [http://www.rspysse.anu.edu.au/~sth110/Astropaper\\_extra.html](http://www.rspysse.anu.edu.au/~sth110/Astropaper_extra.html).) Its chemical nature is principally siliceous, with trace metal ions.

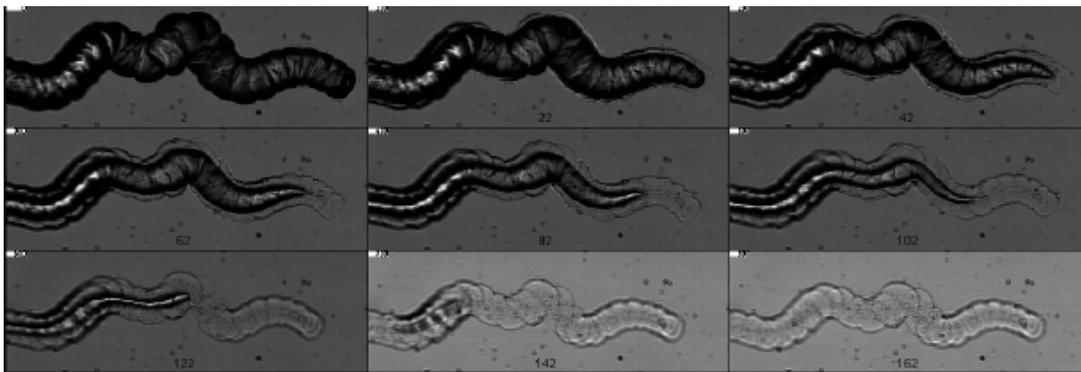


Figure 6: Time-lapse sequence (100 sec. between frames) showing dissolution of witherite crystallites within a biomorph (on addition of 0.03M acetic acid solution), from top left to bottom right. Note the conservation of the complex morphology in the silicate aggregate left (bottom right).

This silicate biomorph can act as an active trap for any hydrophobic species in the system, such as PAH's. For example, fluorescence microscopy demonstrates that anthracene, introduced into the sample in ether solutions, aggregates on the silicate sheath over a few hours (Figure7).

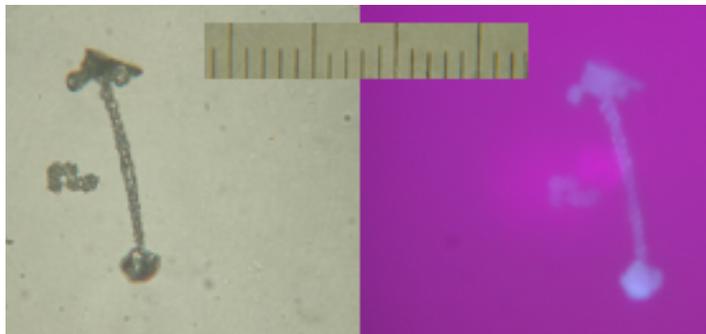


Figure 7: Left: Optical micrograph of a silicate biomorph, 5 hours after addition of anthracene-ether solution at 25°C, viewed in white light. Right: Fluorescence micrograph, indicating anthracene coating of the silica biomorph. (Microscope image collected under excitation at 365nm, emission >397nm; scale bar shows 20µm intervals between finer gradations).

Quantitative kinetic studies reveal that a range of amino acids are also strongly absorbed onto the silicate aggregates within hours (Ng and García-Ruiz). It is entirely plausible, though yet undemonstrated, that the resulting organic aggregates can then remain in isolation, following dissolution of the secondary silica substrate. In this fashion, sequential replacement of the original inorganic biomorph by organics via absorption onto the silica can occur, with retention of the original biomorphic form. We note that this dissolution sequence need not be confined to witherite dissolution. Equally plausibly, silica dissolution may occur (eg. under mildly alkaline conditions) leaving overgrown biomorphs via absorption onto the witherite following silica redissolution and later witherite removal. The original witherite-silica assembly can act as a “scaffold” for formation of biomorphs with a variety of stoichiometries, both inorganic and organic. Thus, a plausible non-living pathway for genesis of some putative microfossils is clear, involving accretion, growth, absorption and dissolution processes. They may well have formed under the inexorable directions of non-living processes, rather than the more complex processes of the biology.

The claimed advance in fossil recognition from micro-chemical analyses is a real one for relatively young samples where one can expect preservation of lipid or other cell-wall molecular detritus. However, stoichiometric analyses are unlikely to obviate the need for morphological studies for other samples, particularly Archean ones. For – just as there appears to be a morphological convergence between living and abiotic residue in these samples – their chemical residue is likely to be no more complex than that of the prebiotic earth, which included a wealth of organic molecules, from simple PAH's to more complex amino acids.

From the astrobiological perspective, it is clear that the composite materials described above demonstrate the dangers of invoking morphology as a biomarker. It is today clear that, even at the microscopic length scale, abiotic mechanisms yielding non-faceted and even non-crystallographic morphologies are wide. The selective adsorption of impurities on crystal surfaces may provoke the formation of non-crystallographic faces (rounded unfaceted crystals), which disguise the underlying atomic structural symmetry. At higher concentration of impurities, particularly in the presence of surfactants, three-dimensional nucleation leads to crystal aggregates with self-organised banding, a feature elsewhere considered specific to biomineralization (Dominguez-Bella and García-Ruiz, 1987). When ordered at the mesoscopic scale, these structures are shaped by the arrangement of two-dimensional defects such as disclinations (Yang et al., 1997). The remarkable kerite structures found by Yushkin in chamber pegmatites might have been formed by this mechanism (Yushkin, 1996). These boundary structures may also mediate the aggregation of crystallites, sometimes co-operatively, e.g. the precipitation of calcite in the presence of poly(aspartate) (Gower and Tirrell, 1998). When these boundary structures are semipermeable, as in chemical gardens (Vail, 1952), the precipitate shapes are governed by osmotic pressure. The resulting morphologies often bear startling resemblance to geological (Cairns-Smith et al., 1992), (Russell et al., 1989) and biological forms, with filamentous and bulbous forms similar to those of numerous simpler organisms as well as nanobes (Uwins et al., 1988). An interesting feature of these gardens is the huge variation of length scales possible, depending on the precipitate and membrane chemistry (Coatman et al., 1980), (Collins et al., 1998), (Double and Hellowell, 1976) presumably extending down to the nm scale

of nanobes. Finally, the remarkable structures described in this manuscript, where the existence of strict orientational order at the scale of the building carbonate crystallites (typically at the nm scale) is accompanied to a strict geometrical precision at the scale of the whole structure (typically at the mm scale) set by a local twist constraint. This brief directory of inorganic precipitates displaying non-crystallographic shapes must be added to those more classical mineral but biomimetic structures correctly interpreted by geologists as abiotic in origin.

Finally, the case of stromatolites, often considered the oldest remnants of biological activity on the earth, deserves some discussion as it sheds further light on the complexity of deducing biogenesis from morphology. As pointed out by Walter's comment (Walter, 1996) on the work of Grotzinger and Rothman (Grotzinger and Rothman, 1996), the issue of biogenicity of Precambrian stromatolitic structures exposes the difficulties in deducing the duration of terrestrial life from synthesis of fragmentary evidence from different disciplines. Stromatolites, made by cyanobacteria, form dome-shaped biogenic structures whose cross-sections display sinuous fractal-like outlines. The recent introduction of fractal geometry offers a useful morphological quantifier for biological and geological patterns, including stromatolites. Unfortunately, in some cases, the implications of fractal quantification have been extrapolated beyond reason. As with any morphological measure, fractal dimension alone does not offer proof for a specific formation mechanism. To use a simple Euclidean example, could one infer that tennis balls derive from trees because, like oranges, they exhibit a fractal dimension of three? Evidently not: geometry alone does not determine genesis. The same objection holds when comparing non-integer fractal shapes. Even in those few cases when geometric analysis yields a fractal dimension characteristic of a well-known mechanism, the issue for Natural Historians remains far from closed. For instance, detection of an exponent characteristic of diffusion limited aggregation (DLA) (Witten and Sander, 1983) allows the physicist to conclude that a Laplacian-type mechanism is most likely to be responsible for growth. However, given that Laplacian growth may be due to a Saffman-Taylor instability, diffusion controlled accretion phenomena or electrical conduction (Ball, 1986), the Natural Historian, concerned with the origin of biological or geological structures, must go further (Checa and García-Ruiz, 1996). In some cases, identification of fractal dimension may admit a reasonable hypothesis for the (physical) *class* of possible formation mechanisms, but rarely the mechanism itself. Thus, for example, Grotzinger and Rothman offer a bundle of four possible abiotic mechanisms consistent with the observed fractal properties of stromatolitic structures. This final example illustrates again that understanding of biogenicity must involve a synthesis of concepts across a range of disciplines, spanning physics, chemistry, biology and geology. The persistent use of morphology as a signature of biogenicity is unlikely to be replaced. To use that technique more gainfully, we must certainly understand the biological mechanisms involved in morphogenesis of primitive forms of life. But in parallel, we must delineate more adequately the possible variety of forms that can emerge from purely abiotic processes. Studies summarised here make it clear that that variety is considerably broader than current belief.

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