EXTRATERRESTRIAL ABIOGENIC ORGANIZATION OF ORGANIC MATTER: THE HOLLOW SPHERES OF THE ORGUEIL METEORITE

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Abstract. Fragments of the Orgueil meteorite were macerated in mineral acids (HNO_3-HF-HNO_3) to dissolve the mineral matrix and separate the acid-resistant organic residues; a routine procedure in the extraction of pollen and spores from terrestrial sediments. Numerous spherical hollow objects were found, optically resembling the brown amorphous residual organic matrix of the meteorite. Their morphology, size-distribution, and chemical composition, revealed by electron microprobe with reference to carbon and phosphorus, are described, and evaluated in connection with criteria of biogenicity. The intrinsic criteria are satisfactorily met, but the extrinsic requirement of a sedimentary environment is not met. A review of the literature concerning the meteoritic environment suggests an explanation of these spheres based on the environment of their formation. It is proposed that they are organic coatings on olivine microchondrules, magnetite and glass globules, the mineral component of which has been dissolved by the acid maceration. They could have initially resulted from the polymerization of dispersed small organic molecules condensing on the surface of the microchrondrules. The latter were injected from a volcanic 'nuée ardente' into the dispersed cold primordial cosmic dust of hydrated silicates and organic molecules, around the meteorite parent-body. This presumably occurred before the cosmic dust accreted as the carbonaceous chondritic outer layer of the parent-body. Upsurging reducing hot gases from the 'nuée ardente' would polymerize part of the dispersed organic matter as the insoluble brown amorphous matrix, possibly the 'sticking' agent when the cosmic dust accreted. The spiraled form of several of the organic structures described here are suggestive of atmospheric heat microturbulences. Organic membranes and comet-form tails of spherical coatings suggest polymerization in the wake of injected microchondrules. These diverse organic structures would result in our view from the abiogenic thermal organization of organic matter in an extraterrestrial gas-solid system.*

Résumé. Plusieurs fragments de la météorite d'Orgueil ont été macérés dans des acides minéraux (HNO_3, HF, HNO_3), afin de dissoudre la fraction minérale et isoler la fraction résiduelle résistant aux acides. C'est là un procédé utilisé couramment en palynologie pour extraire les grains de pollen et les spores des sédiments terrestres.

De nombreux objets microscopiques, sphériques et creux, ont été mis en évidence. Ils sont optiquement similaires au résidu organique brun, amorphe, dans lequel ils sont enrobés. Leur morphologie, leur répartition en fonction de leur taille, et leur composition chimique élémentaire, analysée par la microsonde électronique, qui révèle la présence de carbone et de phosphore, sont décrites, puis évaluées en fonction des critères disponibles d'une éventuelle origine biologique. Les critères intrinsèques aux objets sont bien satisfaits, mais non le critère extrinsèque d'un environnement sédimentaire convenable.

L'analyse des hypothèses qui ont été avancées pour décrire l'environnement originel de la météorite,

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permet de suggérer une explication de ces sphères creuses organiques, qui repose entièrement sur cet environnement à l'époque de leur formation. Ce sont des revêtements organiques à la surface de microchondrules d'olivine, de globules de verre et de magnétite, minéraux de haute température appartenant à la fraction minérale de la météorite qui a été dissoute par la macération acide.

Ces coques organiques résulteraient de la polymérisation de petites molécules organiques dispersées, qui se seraient condensées à la surface de gouttelettes minérales en fusion. Ces dernières ont pu être éjectées par une nuée ardente volcanique issue du corps parent de la météorite, et projetées dans la poussière cosmique primitive froide en suspension autour de ce corps parent, composée de silicates hydratés et de petites molécules organiques.

C'est ensuite seulement que cette suspension de poussière primitive aurait subi l'accrétion pour former finalement la couche extérieure froide de matière éteoritique carbonée du corps parent. En outre, des gaz réducteurs à haute température, s'élevant de la nuée-ardente, ont pu polymériser en partie la matière organique en suspension, pour former la matière météoritique organique amorphe, résistant aux acides, qui a peut-être été l'agent agglomérant lors de l'accrétion.

Les formes spiralées de plusieurs des structures organiques décrites ici suggèrent des microturbulences atmosphériques dûes à la chaleur. Des membranes organiques, et l'appendice en forme de queue de comète d'une sphérule, suggèrent une polymérisation organique dans le sillage de la trajectoire de microchondrules. Selon notre opinion, ces diverses structures organiques résultent donc de l'organisation abiogénique sous l'effet de la température, de matière organique préexistante, plus simple, dans un système solide-gaz extraterrestre.

The present research started as a preliminary investigation using palynological techniques in 1962 at the suggestion of Friedmann [34]. The work was initiated by the first author who was unaware of the then current controversy about alleged life-like structures in the Orgueil meteorite. The work was resumed in 1968 in Paris and shortly after at Harvard University. The objective was to secure reliable information about one category of objects which was found by the first author in 1962 and believed at the time to be indigenous to the Orgueil meteorite. It might seem that there is an essential ambiguity in this investigation in that the search for organized objects in a carbonaceous meteorite is tantamount to a search for evidence of extraterrestrial life. Indeed, this approach is intriguing as long as 'a vigorous and healthy skepticism' [86], prevails. Indeed, Urey *et al.*, felt it necessary to clarify their position in this respect [93].

Because of the 'like-nothing-on-earth' environment of the meteorite at the time of formation an essential skepticism about the significance of morphological similarity to known terrestrial microstructures is prerequisite. Moreover, recent advances in the laboratory synthesis of abiological carbon compounds [72], more elaborate hypotheses on how life may have emerged on Earth [10, 64], and increasing awareness of the difficulties in recognizing criteria for 'biogenicity' of organic compounds (compare Hodgson and Baker [38] and [39]) all signify the necessity of an unyielding critical and skeptical attitude.

1. Previous Literature on Orgueil Organized Elements

The recent abundant literature about organized elements in carbonaceous chondrites, reflects roughly two opposite positions: (1) firm believers in extraterrestrial biogenic origin, and (2) unremitting opponents or skeptics. There were also diverse inbetween unconvinced researchers.

From 1961 to 1967, the conclusion of most of the descriptive and illustrated reports by microbiologists, algologists and micropaleontologists on organized elements was that they were fossils, biogenic, and amenable to botanical taxonomic nomenclature; Claus and Nagy [18], Staplin [81], Palik [68, 70], Van Landingham [95, 96, 97], Timofejew [88, 89]. There were subsequent complementary and in part disputationary papers by Claus and Nagy [19, 20, 21, 22], Nagy *et al.* [56], Nagy *et al.* [58], Claus and Suba-C [24, 25], Claus *et al.* [23], Palik [69], Ross [75, 76], Staplin [82, 83], Tan and Van Landingham [85], Urey [90, 91], Van Landingham [94], Nagy *et al.* [55], and Manten [44].

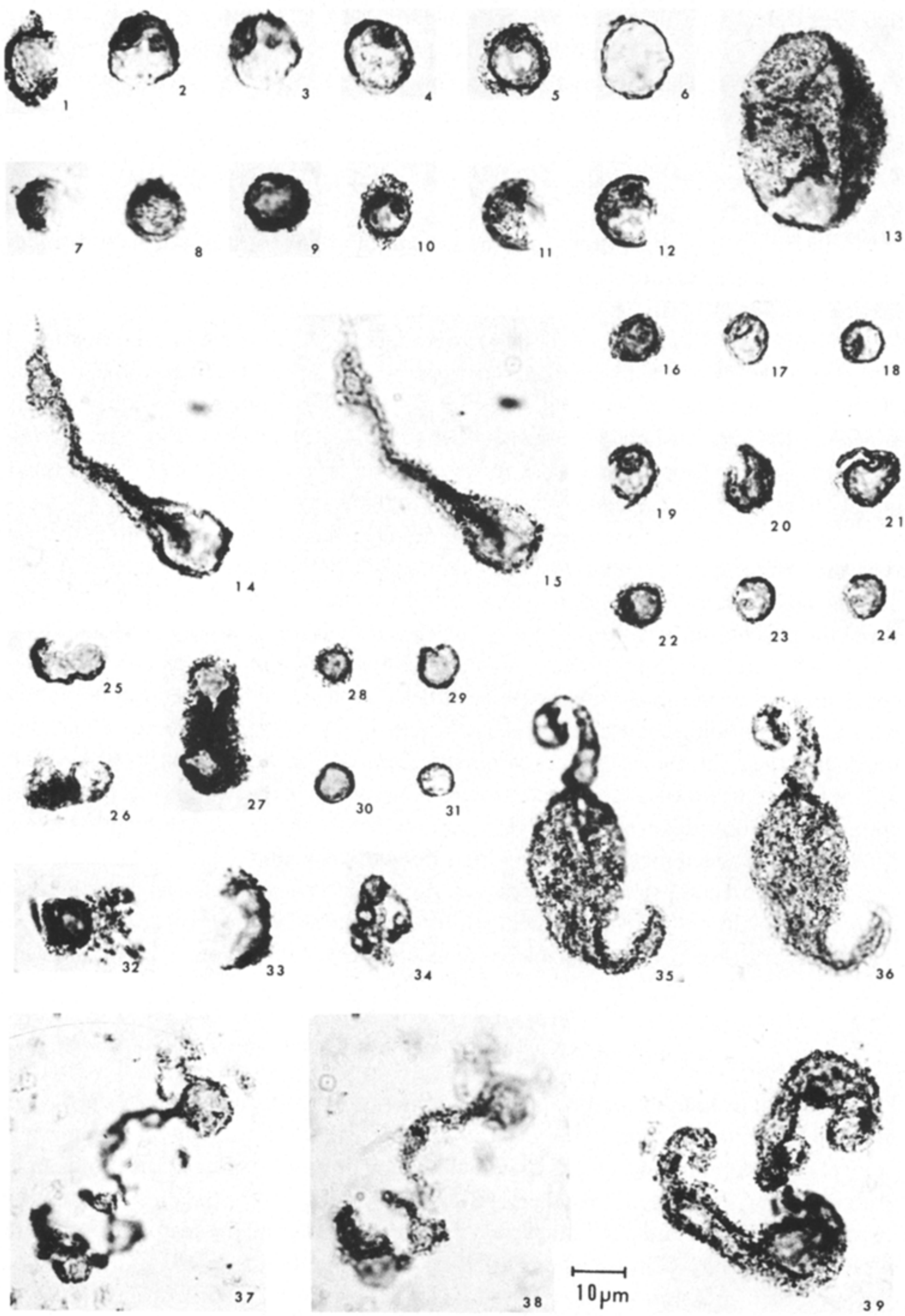
In subsequent criticisms that arose the skeptics and partisans of unconclusiveness insisted that the structures were terrestrial contaminations or minerals: Anders and Fitch [2], Botan [13], Briggs [14], Briggs and Kitto [15], Briggs and Mamikunian [16], Deflandre [27, 28], Fitch *et al.* [30], Fitch and Anders [31, 32], Friedmann and Rossignol [34], Gregory [35], Mamikunian and Briggs [41, 42, 43], Morrison [48], Mueller [49, 50, 51], Orcel and Alpern [66], Pearson [71]. Finally, the previous 'believers' felt it necessary to leave their later reports inconclusive: Nagy [61], Nagy *et al.* [63]. Previous 19th-century reports were those of Hahn [36, 37], and Vogt [98], though mainly overlooked in the new discovery and controversy.

Great care seems to have been generally used in the handling of samples during laboratory procedures. The problem of terrestrial contaminations both in the field and in the laboratory have been experimentally investigated: Claus *et al.* [23], surveyed the microbiota in soil and sediments of the Orgueil village area near Montauban, France; Anders *et al.* [5], showed that a particular fragment of Orgueil was purposely contaminated, probably during the 19th century. Tasch [86, 87], deliberately accepting laboratory contaminations, attempted through blank control slides to determine them and then subtracted the contaminants from the meteorite slides. Briggs and Kitto [15], reported recent contaminations on the surface of the Mokoia carbonaceous chondrite; Oro and Tornabene [67], tested bacterial colony growth from Murray, Mokoia and Orgueil meteorites and found none in Orgueil.

Claus [26] in a comprehensive review has surveyed the history of the contamination problem and, after culture experiments to test growth of microorganisms, concluded that contamination by terrestrial microorganisms has generally been overemphasized.

2. Description of the Hollow Spheres

Palynological techniques were applied to fragments of the Orgueil meteorite provided first by Dr Friedmann, then by Prof. Orcel, to isolate the acid resistant organic matter, amorphous or organized, from its mineral matrix. The outer surface of the first sample was cleaned by scraping, of the others, by use of freon gas under pressure. To eliminate the mineral fraction and partially bleach the organic material, the reagents used were $KClO_3$ plus HNO_3 (Schultzes reagent) at 20°C, concentrated HF (52%) at 60°C for one hour, then 20°C for 24 h, then HNO_3 at 20°C for one hour. No centrifugation was performed to avoid the disruption of any fragile structures; instead, the liquid reagents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were decanted and siphoned. The organic residue was then mounted, some of it in phenolated glycerin, some in phenolated glycerin jelly, and some in distilled water.

Under the white-light microscope, this residue appeared loosely spongy and from light to dark brown in color, but neither black nor yellow, and mostly agglomerated in very easily disrupted loose lumps of regular elongated hexagonal outline. Orcel and Alpern [66], have shown the homogenous distribution of carbon in Orgueil, and their electron probe image of a polished surface at magnification 200 x in the electron probe is not contradictory with our observations after removal of the mineral matrix. Some euhedral hexagonal opaque crystals of troilite, of different sizes have been observed [30, fig. 1A], [49], [31], [12].

We chose to deal only with microstructures which have identical color and optical density as the surrounding acid-resistant organic matrix, thus achieving a better guarantee of non-contamination. Careful and time-consuming scanning of the microscopic preparations revealed the not infrequent (Plates 1, 2 and 3) presence of spherical hollow bodies, with a well-delimited wall. These bodies have exactly the same color and optical density as the organic brown residue.

The inner surface of the wall is smooth while the outer is irregular. This irregularity is due to small and irregularly distributed granular particles of matrix which adhere to the wall. In surface view the wall of the bodies appears transparent between the adhering particles. Often the wall is broken, the break covering only a small portion of the surface of varying size and shape, occasionally splitting the sphere into two irregular halves. This is evidently the result of mechanical fracture. When the wall is extensively fractured, its elasticity is evident in that it coils inward toward the center of the spherule. Lowering of the optical focus from the upper surface of the bodies to the lower demonstrates that they are hollow, and empty.

Discrete structures other than spheres are present. These include membranes, which are light brown, transparent, and irregularly granular. They are also similar optically to the amorphous organic matrix. At times, the spherical hollow bodies are combined with the membranes (Plate 1, figs. 14, 15), showing a hollow sphere imbedded in a membrane shaped like a comet tail bearing a flaring, irregular, funnel-shaped extremity. Several hollow spheres are occasionally imbedded in line in an irregular tube-shaped membrane possessing extremities like arms spiraling in opposite directions (Plate 1, fig. 39). The spiral form appears again in another object of the

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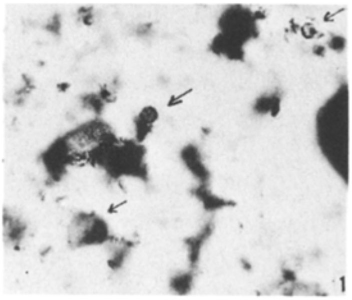
Plate 1. Organic hollow spheres and structures of the Orgueil meteorite. White-light micrographs. Magnification 1000x.

No. 1 to 12, 16 to 24, 25, 26, 28 to 34: spheres, complete or split, some showing the thickness of the wall (12, 21, 25).
No. 13: a full ovoid structure of organic matter.
No. 14, 15: hollow sphere with comet-shaped tail.
No. 27: two 'splitting' hollow structures.
No. 37, 38: spheres linked by filamentous structures.
No. 39: spheres imbedded in spiral arm-shaped structures.
No. 35, 36: full organic structure with spiral ribboned appendices.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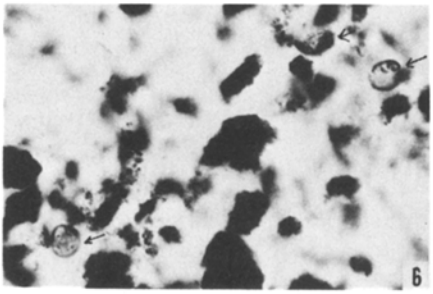
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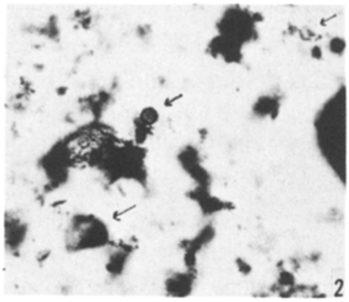
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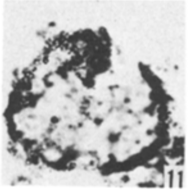
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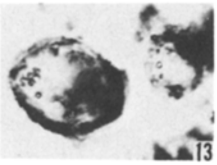
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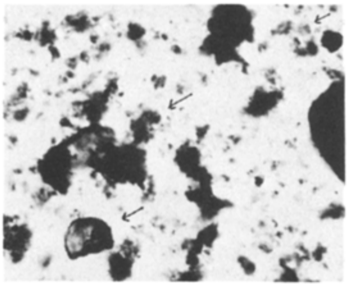


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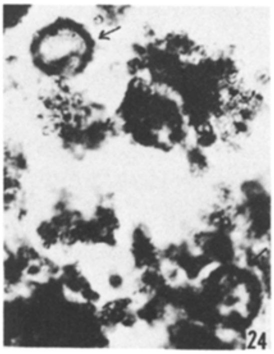
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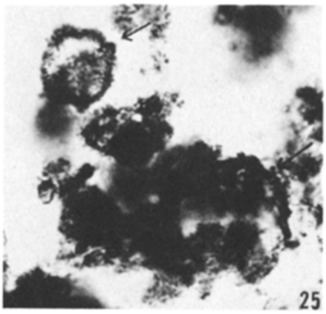
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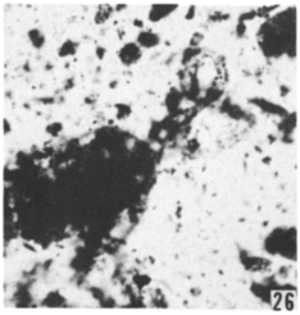
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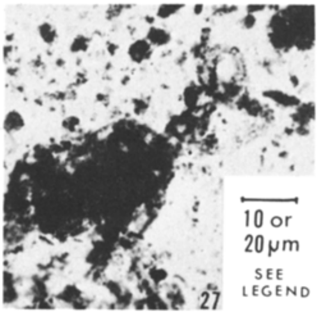
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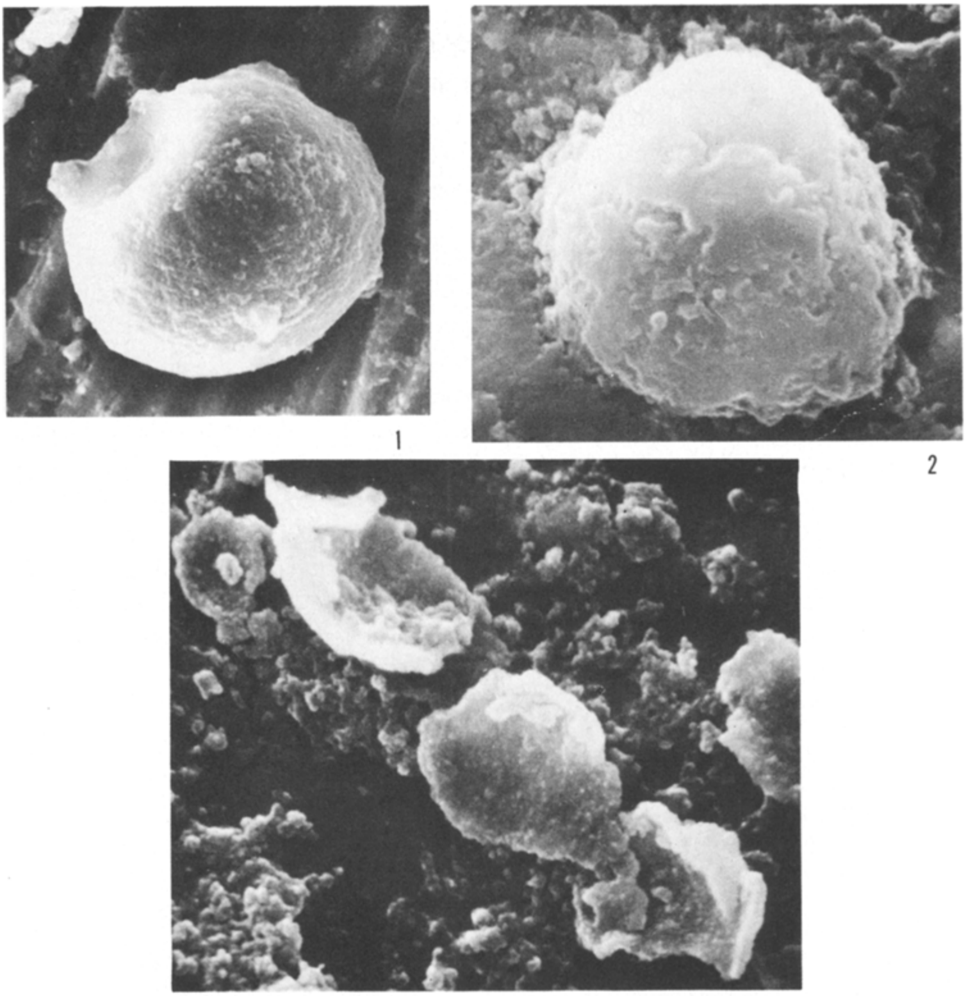

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Plate 3. Scanning micrographs of the hollow spheres of the Orgueil meteorite. Magnification 5000x.
No. 1: Hollow sphere with broken wall. No pattern appears on the surface.
No. 2: Hollow sphere covered by a thin layer of amorphous organic matrix.
No. 3: Split hollow sphere, showing the thickness and elasticity of the wall, and the cup shape of the three fragments.

Plate 2. Organic hollow spheres and microstructures of the Orgueil meteorite. White-light micrographs. Most pictures at magnification 1000x.

No. 1, 2, 6, 26, 27, 28, at magnification 500x.
No. 1, 2, 28: Three spheres in the same microscope field at different levels of focusing.
No. 6, 24, 25: Three spheres (No. 6), and two spheres (No. 24, 25), in the same field.
No. 14, 15: same split sphere.
No. 21, 22, 23: same fragment of sphere at different levels of focusing.
No. 17, 18: two joined spheres at different levels of focusing.
No. 5: broken sphere with wall coiling inward.
No. 3, 4, 7, 8, 9, 10, 11, 12, 13, 16, 19, 20: complete or split spheres of diverse diameters.
No. 26, 27: structure with tube-shaped appendice.

same color and optical density, but which is not hollow, and appears to be made of agglomerated matrix grains, a central ellipsoidal body, prolonged by ribbon-shaped arms spiraled in opposite directions; the surface of the central body bears a longitudinal fold (Plate 1, figs. 35, 36).

Neither the hollow spheres, nor the other structures, nor the matrix itself, can be stained with safranin dyes. It should be noted that safranin is readily adsorbed by recent terrestrial organic microfossils.

In summary it appears that there are two different types of objects composed of the same organic matter as the amorphous matrix, one of which appears in large numbers with a constant structure, viz., the hollow spheres; the other type consists of unique and irregular objects, membraneous and spiraled.

For examination with the scanning-electron microscope certain hollow spheres were isolated from suspension in water by hand with micropipettes under the white-light microscope and deposited on an aluminum pedestal. After positioning they were gold coated to a thickness of 500 Å. One sphere showed a small break, its surface being granular and devoid of any regular pattern (Plate 3, fig. 1). Two others were covered with amorphous organic matrix (Plate 3, fig. 2). Another sphere appeared to be broken into three cup-shaped fragments, in which specimen the thickness of the wall was appreciable. It may be concluded that the intrinsic elasticity of the wall persists even under conditions of desiccation (Plate 3, fig. 3).

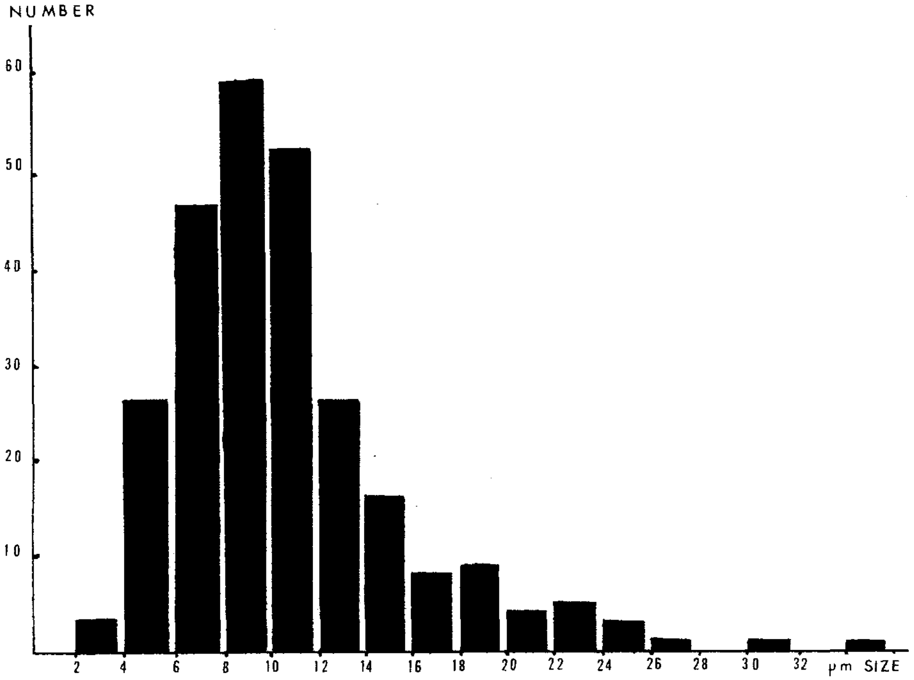

Fig. 1. Histogram showing the size distribution of the organic hollow spheres of the Orgueil meteorite.

The absolute frequency of these spherical bodies within the meteorite is difficult to evaluate. In Plate 2 (figs. 1, 2, 28) three of them appear at different focal levels in the same field of the microscope at a magnification of 500 x. It is probable that the spherules have an irregular distribution within the meteorite. Very approximately, a frequency of 300 to 500 per cm^3 seems reasonable in this particular fragment of the Orgueil meteorite. Their size distribution plots as an asymmetrical unimodal histogram (Text-Figure 1). This histogram curve is based on the measurement of 258 spheres. The peak is in the 7–10 μm range, only a few exceed 25 μm.

The organic chemical nature of these hollow spheres is evidenced by the technique of isolation, whereby mineral material has been removed. However, an attempt has been made to determine further their elemental composition. A sample was prepared for chemical analysis by use of the electron microprobe X-ray analyser. The spheres were hand-picked with a micropipette under a white-light microscope from part of the residue suspended in water and deposited on a round glass slide. They were located on the slide under a white-light microscope, and photographed for mapping at different magnifications to allow their re-location under the white-light optics of the electron microprobe. The preparation was aluminum-coated. The acceleration voltage of the electron beam was 20 kV, the beam focused to 1 μm^2, which gave a high precision concerning the identity of the probed object.

Three spheres and the surrounding organic matrix of each in the three nearby locations in the same microscope field were analyzed. The glass slide was likewise probed under similar conditions. This was done because the spheres and the matrix are not sufficiently dense as to prevent the penetration of the electron beam into the glass support itself. Five readings were made for each analysis. The following elements have been looked for: C, P, N, K, Ca, Si, Ni. The results are shown in Table I. The figures are non-corrected X-ray counts per time unit related to the X-ray intensity of the analyzed element, and, therefore, proportional to the amount of the element present.

TABLE I

Electron microprobe X-ray analysis of 3 hollow spheres, organic matrix and glass slide background. The figures are non-corrected X-ray counts per time unit; each value is the average of five counts. For the matrix, three points of different thicknesses have been probed around each hollow sphere. The variations from minimum to maximum reflect exactly the variation in thickness of the matrix.

Elements	Hollow Spheres			Matrix		Glass
	P-37-1	P-38-1	L-42-3	Minimum	Maximum	
C	267.13	619.75	784.25	165.50	2504	58.58
P	196.38	113.75	95.88	62.25	364.75	89.42
N	131.92	144.25	140	125	189	124
Si	10376.13			3170.25	11833.75	12872.75
K	564.38	1179.75	907.60	526	2674.88	453.57
Ca		2955.13	2913.25	720	3222.50	3135.88

The figures are generally low because of the very small size of the objects. However, reliable ratios can be computed. The hollow spheres are definitely carbonaceous: their carbon counts are significantly higher than those of the glass, which represents the background noise of the microprobe. In the spheres, the carbon counts increase with the thickness of the wall; in the matrix this carbon count increase with its thickness is also noted.

Phosphorus is present in the spheres and in the matrix; in both it is generally higher than in the glass. It seems to vary roughly parallel to carbon in the matrix, but inversely so in the spheres.

There is no silicon in the spheres nor in the matrix: their high silicon values reflect solely the underlying glass, and they are definitely lower than in the glass slide control.

Similarly the calcium response is from the glass, where it is higher than in the spheres and in the matrix. However, one sample of the matrix shows a positive response for calcium.

Potassium is much higher in the spheres and in the matrix than in the glass. The possibility exists that the relatively high K content may be residual from the $KClO_3$ used in the macerating solutions. This, however, seems highly unlikely in view of repeated washings after acid treatment.

The nitrogen counts are not conclusive inasmuch as the electron-probe accelerating voltage was set at 20 kV where 8 kV would have been preferable. Nickel was not detected. Vanadium could not be tested for lack of availability of a standard. Porphyrins, differing from those of terrestrial sediments, therefore indigenous, and presumably resulting from abiogenic synthesis, have been detected in Orgueil and other carbonaceous chondrites by Hodgson and Baker [39]. Ancient sediments on Earth contain only porphyrins in which nickel and vanadium are complexed.

The opportunity to carry out more extended chemical analyses was precluded by the minute mass of the spherules available.

The reasons why these spheres are not believed to be contaminations should be summarized: (1) their similarity in color, consistency, and chemical composition to the surrounding organic residue, (2) their negative reaction to the palynological stain safranin, (3) their uniform structure, (4) their relative abundance. Moreover, pollen grains, fungus spores, and textile fibres cannot be confused with them by any morphological criteria.

From our data certain generalizations may be made. Of these the similarity of elemental chemical composition of the insoluble organic matrix and the hollow spheres definitely points to their indigenicity in the meteorite. It has also been shown that carbon is a major constituent of both spheres and matrix and that phosphorus is consistently present.

This similarity makes it relevant to summarize here what is known of the composition of this insoluble organic matrix. This non-extractable acid-insoluble residue is amorphous carbon and organic matter; there is no graphite in Orgueil, as shown by X-ray diffraction [14, 31, 79]. That the residue is indigenous, has been brought out by

Smith and Kaplan [79] in Orgueil, and in 6 other carbonaceous meteorites, on the basis of δC^{13} values which fall within a narrow range, between − 14.8 and − 17.1 per mil (Orgueil: − 16.9) and outside the terrestrial range of kerogen from Precambrian sedimentary rocks and carbon from petroleum (− 25 to − 35 based on the PeeDee Belemnite standard). This insoluble material is probably a mixture of polymers of not highly condensed aromatic structures, according to Bitz and Nagy [11]. However, Smith and Kaplan [79] regard it as consisting of highly condensed aromatic structures, which in Orgueil, account for 57 % by weight of the total carbon. Among the carbonaceous chondrites this proportion ranges from 25 to 57 % for type I and II, and increases to 63 % in type III where the total carbon is lower. Brooks and Shaw [17], claim the chemical and biological identity of the Orgueil insoluble organic matter with sporopollenin, regarded as a class of undefined polymeric material with several common chemical characteristics. Whether or not these characteristics apply to a group of organic compounds of related structure, their common and only possible origin as biogenic material is a quite different matter. The elemental analysis of the insoluble organic fraction of carbonaceous chondrites given by Raia [73], does not mention phosphorus. The origin of the meteoritic material, both soluble and insoluble, is still a matter of speculation, and an enigma. More will be said further about it.

3. Discussion: Criteria for Biogenicity

Could these organic hollow spheres be biogenic structures? This, of course, is the crucial question. The nagging difficulty for the paleontologist in search of the oldest evidence of life on Earth to differentiate between once-living (fossil) and never-living structures, has been stressed by Rutten [77]. In the most favorable case the once-living structure is preserved in its minutest details by the 'replacement', of the original organic substance by minerals: 'actual preservation of organic substance is rare in the extreme'. This meteorite indeed provided organic structures, but, because of the particular and still speculative parent body environment, the approach must be much more critical than is necessary with terrestrial Precambrian organic structures.

Which criteria should be met to accept biogenicity of any structure? Some are intrinsic to the structure, some extrinsic.

Among the first, the degree and constancy of organization are both apparent at high magnification in the white light microscope as well as the scanning electron microscope. Indeed an extreme morphological simplicity is predicted for very early structures, should they be biogenic or pre-biogenic. The cooling of the meteorite parent-body is estimated to have occurred around 4.4 to 4.6 × 10^9 yr ago [4]. This simplicity is the case in early Precambrian microfossils (3.2 × 10^9 yr) from the South African Fig Tree Series [78, figs. 1–4]. In the sediments underlying the Fig Tree Series, (Onverwacht Series > 3.2 × 10^9 yr old), among acid resistant microstructures not believed to be biogenic, a well-developed sphere [62, fig. 1E] which is representative of a whole regionally delimited group whose size ranges between 6 and 20 μm, resembles the Orgueil hollow spheres.

Chemical composition is another intrinsic criterion for biogenicity, although not obligatorily organic. It is met here on the level of elemental composition. Electron microprobing of 29 more or less well defined 'organized elements' in Orgueil has shown iron and silicate mineralization, some negative analyses are interpreted as indicating carbon [58]. As we will show further, these may be related to the hollow organic spheres. Electron microprobing has not yet been attempted on isolated Early Precambrian organic microstructures.

The abundance of organic objects in Orgueil might be interpreted as evidence of biogenicity. The chances of fossilization for an individual organism are low for a small living population and increase with the size of the population, eventually resulting only in few fossils but these originating from many living individuals.

The size distribution of the Orgueil objects is indeed evocative of populations of Precambrian organisms as shown in histograms of the microstructures in the Precambrian Gunflint chert [7, fig. 9]. These present a very similar asymetrical skewed curve, which is considered as indicating a 'diversity of organisms or entities rather than a normal distribution of a single "species" or "entities"'. The size distribution [24, fig. 1] of 303 randomly selected type 1 and 2 'organized elements' of Claus and Nagy [18], has a similar range, but with a major peak at 4.5 μm and two smaller ones at 3 and 9 μm. Besides, its similarity to the size distribution of magnetite globules and glass globules [50, fig. 8], has been stressed [51], but the validity of the comparison has been denied [25]. These mineral organized elements were previously shown [31], to be soluble in HF-HCl.

In a terrestrial context these criteria are unequally compulsive; their values are interdependent. A complex morphological organization, characteristically referable to unquestionable organisms, is a salient clue to biogenicity. If it is missing, other criteria have to be met. The chemical composition need not be organic, but if organization is very simple, an organic composition is a better and safer clue than silicon, calcium carbonate or pyrite. A small number of individuals would not be an obstacle to acceptance of biogenicity if all other criteria are met. Size distribution plots are nondiscriminatory in resolving the question of origin of unknown objects.

It must be kept in mind that too little is known of the borderland between pre-life and life, on Earth and more obviously so in extraterrestrial environments, to decide if criteria derived from established terrestrial fossils or living organisms would be fully relevant in such speculative circumstances. But we have no other than these. On this basis, we can say that the *intrinsic* requirements for biogenicity of these hollow spheres might seem rather satisfactorily met.

But on the other hand, there are *extrinsic* criteria for biogenicity, related to the environment.

On the Earth, fossils are found only in sedimentary layers: it is a fundamental prerequisite as a function of deposition. There is no sedimentary structure in the Orgueil meteorite, as already pointed out [4, 65, 92]; the presence of liquid water on the meteorite parent-body during at least 10^3 yr has been admitted [3, 57], but considered at best as an interstitial filling of the accreted matrix, depositing magnesium sulfate in the veins, too scarce to postulate an aqueous sedimentary environment.

However, Urey [92] states that the texture of the Orgueil meteorite closely resembles terrestrial pyroclastic sediments such as volcanic tuffs, settling in water, since it shows localized preferred orientation of the clay minerals as might result from plastic flow. Orcel and Alpern [65] quote a fluid structure. There is also some similarity to a terrestrial loess. Against a long aqueous stage, Mueller [50] notes that olivine microchondrules are present, although on Earth, olivine decomposes completely in a few decades or centuries when exposed to atmospheric moisture. The presence of olivine microchondrules is indicated by Orcel and Alpern [65], on the basis of electron microprobe elemental analysis; it is discussed by Boström and Fredriksson [12], who do not rule it out. Therefore, the meteoritic texture does not conform satisfactorily with the usual sedimentary requirement associated with true fossils.

4. Environment of the Meteorite Parent-Body

From this point, in order to understand the genesis of the hollow spheres, the environment on the meteorite parent-body is the next aspect for consideration, although it is still hypothetical. The different theories which postulate the environment tend to agree on some points – and disagree on others. However, we shall try to find enough bases to allow us to propose an explanation of the organic hollow spheres consistent with speculation on the environment of the meteorite parent body in its history. Such considerations definitely exclude biogenicity.

Carbonaceous chondrites, as the other meteorites, are generally believed to come from one or several parent-bodies which accreted 4 to 5 billion years ago in the outer fringe of the asteroidal belt between Mars and Jupiter, at 3.9 AU, thus belonging to the solar system [4]. They could also come from comets. They are composed of two fractions: low-temperature fraction A, the oxydized matrix of hydrated silicates – maybe not its original state [12]–, with dispersed organic matter, and high-temperature reduced fraction B, microchondrules and chondrules of olivine, metal grains, magnetite and troilite crystals. In type I carbonaceous chondrites, as Orgueil, the fraction B is very small – no chondrules, but microchondrules – the meteorite is mostly the silicate matrix and organic matter, with veins filled with water dispersed magnesium sulfate and other salts. Several hypotheses have been proposed to account for the origin of each fraction, and for their mixing [4].

The origin of the silicate matrix is generally agreed upon as being the cold primitive cosmic dust which accreted last in the outer zone of the meteorite parent-body at temperature equal to or below 315 K [40], and was never strongly reheated [46, 74, 29, 99, 100, 101, 40, 6].

Larimer and Anders [40] suggest the hydrated silicates are primitive, their water beginning to condense about 350 K. But Boström and Fredriksson [12], suggest that the present silicate matrix (a chlorite, [57]), is not in original state: hydrothermal conditions formed first ferrous chlorite, troilite, and magnetite at high temperature, above 265 °C; later, oxydizing process at low temperature, below 170 °C, in aqueous environment rich in ferric iron, formed the present ferric chlorite; 'primordial dust must in the main have been characterized by rather reducing conditions, since the quantities of oxygen present were small'.

There is no general agreement about the origin of the high temperature fraction, specially the chondrules, which appear relevant to our considerations. The chondrules could be older [99], or more recent than the dusty matrix, and derived from it, directly by melting of the dust by lightning discharges in solar nebula before accretion (Whipple 1966 in [40],), or very indirectly, after accretion of the parent-body, magma formation and volcanism. The mixing of the two fractions could result from local equilibrium process (dehydration of cosmic dust and solid-state recrystallization of olivine chondrules by thermal metamorphism [46]), or, more likely, from mechanical process [4, 12].

This implies that the high-temperature fraction did not originate among the cosmic dust. Wood [99] suggested that chondrules condensed as liquid droplets from cooling solar gases during the formation of the solar system, later accreting into planets and asteroids; however, [101], he computed that the pressure necessary to this direct passage from gaseous to liquid phase (100 atm total pressure at 2000 K) would be too high for the conditions of the primordial nebula in the asteroid belt (10^{-5} to 10^{-4} atm); therefore, the primordial condensate was only dust, and chondrules resulted from the later melting of some dust by a high energy event, such as lightning discharge before or after accretion.

According to early views, chondrites are volcanic tuffs which originate in a 'nuée ardente' from explosive volcanism, and chondrules result from dispersal of lava as 'drops of fiery rain' [80]. They are accepted as such by Ringwood [74], Fredriksson and Ringwood [33], Fish *et al.* [29], Boström and Fredriksson [12]. The ignimbritic origin of chondrules [74, 33], implies that a magma was melted in the presence of volatiles, H_2O, CO_2 and reducing gases in the lunar-sized meteorite parent-body (there is not much agreement about the size of the parent-body); then, by lowering of pressure, the gases escaped, causing fragmentation and explosive dispersion of the magma; low viscosity liquid silicate drops at temperatures below 1000 °C solidified into chondrules and microchondrules. Mueller [50, 53, 54] accepted the hypothesis that the surface of the parent-body remained in a state of nuée ardente, a relatively stable state, because of slow degassing linked to relatively small size and low gravitational field. He pointed out the relationships between the juxtaposition of microchondrules, their coalescence into chondrules, and the volatile content of carbonaceous chondrites: in his view the microchondrules and chondrules, molten mineral spray and droplets, are analogous the fog condensates and raindrops; they are transported by the upsurging gases escaping from the nuée ardente and sorted by the gases, and by gravitation. By progressive cooling, the volatiles of the upsurging gases condense and enrich the still suspended cosmic dust, which after accretion, became the outer layer of the parent body and the source of the type I carbonaceous chondrites.

This cloud of primitive cosmic dust already contains a variety of low molecular weight organic compounds, synthesized in space by various possible mechanisms between which we cannot presently decide [8], whether by Urey-Miller type synthesis, Fischer-Tropsch reactions alone, or followed by equilibration process [84].

According to Larimer and Anders [40], in the solar nebula, above 1000 K at pressures less than or equal to 10^{-1} atm, CO is dominant. As the solar nebula cools and temperature falls below 1000 K, at 10^{-3} to 10^{-6} atm, several per cent of C can be condensed into materials such as CS_2, HCN, C_2H_2 and asphalt-like polycyclic aromatic hydrocarbons, if the C/O ratio is slightly above 1. Then organic matter increases with subsequent drop of temperature and Fischer-Tropsch type reactions occur, which result in straight chain high molecular weight aliphatic hydrocarbons. The high-temperature hydrocarbons might be destroyed as the temperature lowers, but would be regenerated by lightning discharges. The condensation of hydrocarbons was favoured by catalytic reactions on mineral grains surface at 500–600 K; at this temperature, aliphatic hydrocarbons tend to change to aromatic. Anders [6], suggests that the temperature condensation of the organic matter could have been equal to or below 550 K.

Briggs and Mamikunian [16] compare the meteoritic organic matter to the waxy coatings on cosmic nickel particles, discharge polymers synthesized abiogenically by radiation in space.

The outermost layers of the Orgueil parent-body, represented by type I carbonaceous chondrites, have the highest percentage of volatiles, both added and original, with the lowest volatilization temperature [52]. There is a parallel trend of increasing high-temperature microchondrules, abundance, size and coalescence [53], decreasing percentage of volatiles and increasing volatilization – and condensation – temperature of these volatiles, from type I to type III carbonaceous chondrites; the maximum temperature during cosmological history of Orgueil could have been 100 to 180°C, [52].

This postulated gradient supports that part of the hypothesis of Mason [46], which suggests that the type I carbonaceous chondrites are consolidated from the outermost layer of the parent-body, and the type II and III, from the deeper zones of this layer, closer to the former incandescent mantle.

The escaping hot gases polymerized a proportion of the already existing organic matter parallel to their decreasing temperature gradient, into the acid-insoluble organic matrix, which is in a dispersed state in the meteorite [60, 66]. Actually, Smith and Kaplan [79], showed that the percentage of insoluble organic matter to the total organic matter decreases from type III to type I, although the total organic matter increases from type III to type I.

5. Origin of the Organic Hollow Spheres

Therefore, the organic hollow spheres can best be explained as organic coatings on olivine microchondrules, globules of glass and magnetite; these microchondrules and globules have been dissolved by the acid macerations during laboratory preparation. The polymerization of organic molecules by upsurging gases in the still suspended cosmic dust outer layer of the meteorite parent-body, before the accretion of this layer, was even more effective at the contact of injected incandenscent spray and droplets of high temperature minerals. Thus, there was elaborated a polymerized organic coating around the microchondrules and globules with a definite membrane-like structure, possessing coherence and elasticity, and, in addition, organic membranes and comet-tail-like structures. It is suggested that local contact thermal metamorphism of organic matter on microchondrules produced the coatings here described as hollow spheres. The dispersed insoluble organic matrix, it is suggested, was directly polymerized by hot gases, and the organic membranes formed by a combination of both processes. In the dense gaseous suspension of cosmic dust, receiving hot gases from the inner layers of the parent-body, the organic spiral structures are the consequences of zones of turbulence, the lineated structures indicate zones of straight heat and gravitation gradient. Their juxtaposition in very small fragments of meteorite (about 1 cm^3) indicates that these structures resulted from very localized atmospheric microphenomena and, accordingly, they are irregularly distributed in different meteorite fragments.

Distribution-size curves of magnetite and glass globules, and olivine microchondrules in Orgueil, although not exactly similar to the histograms of size of the organic spheres, fall in the same size range with a peak around 5 μm [50, fig. 8].

The histogram of the sizes of the hollow spheres is reminiscent of a biological curve because in both cases there is probably a control by the environment. The maximum size of molten mineral droplets before solidification is limited by the physical parameters, and the optimum size of living cells and microorganisms is limited by metabolic and physico-chemical environmental factors.

Orcel and Alpern [65] described the aspect of the antigorite cement in Orgueil as granular, fibrous or lamellar, sometimes with an oriented structure, of fluid appearance, around microchondrules and metallic inclusions. Their figs. 1, 2, 3, show, in thin section, pear-shaped microchondrules oriented parallel to each other, each bearing a tail-like appendage and embedded in the cement which has a fluid-like appearance. These approximate the form of some of our comet-shaped organic structures, Plate 1. If this parallelism results from trajectories in a pre-accretion stage, the question of the mechanism in accretion of the chondrite is a most interesting one.

Smith and Kaplan [79], suggest that the accretion of the cloudy dusty layer of the parent-body into type I carbonaceous chondrites occurred very rapidly, even as a catastrophic event, because the range of δC^{13} values of the residual insoluble carbon is very narrow, 2.3 per mil: 'possibly under highly energetic conditions such as a high temperature (or plasma irradiation), from reduced carbon'. The 'sticky agent' of Urey could be organic matter. Wood [100], suggests electrostatic attraction, a charge unbalance on dust particles causing floculation or clumping. This could be linked to the intrusion of hot gases, and the newly condensed organic molecules would behave as sticking agents. The friability and porosity of chondrites show they have not been consolidated under pressure [45]. The accretion of the cold type I carbonaceous chondrite matrix seems then more comparable in terrestrial geological terms to that of a loess rather than that of a volcanic tuff, it having never been melted. Larimer and Anders [40] although they do not accept the ignimbritic hypothesis, suggest that the organic compounds may represent the sticking agent for accretion of cosmic dust.

It is fair to note that our organic hollow spheres are similar to already described structures. Fitch and Anders, [31, p. 510] describe two particles remaining in samples after HF-HCl treatment, but attribute them to terrestrial contaminants; however, their fig. 5A represents an object which is similar to our hollow spheres. Mueller [50], noted an organic coating on microchondrules. Boström and Fredriksson [12], cited plastic sheaths around limonite aggregates, and suggested they resulted from the distillation – evaporation and recondensation – of organic compounds initially present in the primitive matter, on walls of bubbles created by the vapors (several hundred degrees C) which formed the troilite euhedral crystals. These bubbles were later filled with limonite. They alternatively suggest that the organic compounds may have been adsorbed on the surface of limonite aggregates. Timofejew [89] described twenty rather similar empty spherical objects after HNO_3 maceration of the Migei meteorite, but compared them to Protosphaeridae which he compared to Precambrian algae. The hollow spheres may even be similar to some of the objects electron-probed by Nagy *et al.* [58]; some of the type I objects of Claus and Nagy [18], may indeed be intact microchondrules.

It is our view that the organic hollow spheres extracted from samples of the Orgueil meteorite are the coherent organic coatings on olivine microchondrules, magnetite and glass globules. They simulate biological structures of extremely simple spherical organization. From what has been postulated in models of origin of the carbonaceous chondrites their abiogenic nature is entirely consistent with the chemistry and physics of these models.

We hope that our investigation of these spherical structures will help to abate the former controversy over extraterrestrial life inferred from carbonaceous meteorites.*

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