FURTHER STUDIES ON MICRO-ORGANISMS AND THE PRESENCE OF SYNGENETIC PYRITE

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ABSTRACT. Various microscopic pyrite forms are described from Lower Jurassic and Carboniferous argillaceous rocks and it is confirmed that solution of the pyrite reveals abundant microfossils of a limited range of morphological variety which are described and figured. Two groups are distinguished, one showing clear association with plant material and the other with animal remains. Of the latter, occurrences of later geological and Recent ages are briefly noted and figured. Both forms were probably of a saprophytic nature and are thought to have been involved in the production of hydrogen sulphide and, consequently, the precipitation of the iron sulphide with which they are now found. The environment of this activity is discussed with particular reference to the Liassic deposits studied. No stratigraphic distinction is found for the microfossils within the inclusive period from Lower Carboniferous to Lower Jurassic.

THIS paper presents the results of research in continuation of that published earlier by the author (Love 1957). While elaborating the phenomena then described, it demonstrates their extensive occurrence in other strata and throws some light on the many problems previously left unsolved. These arose from the original demonstration that microscopic pyrite grains of generally characteristic sizes and shapes, occurring in various shales and a bituminous limestone of the Scottish Lower Carboniferous sequence, exclusively embodied small objects regarded as microfossils of a hitherto unknown type. On the basis of this association, these bodies were held to be the remains of organisms which themselves were producers of hydrogen sulphide and became filled and coated with iron sulphide as a result. The smaller of the two types of micro-fossil designated, *Pyritosphaera barbaria* Love 1957, was yielded by small uncompressed spherical pyrite grains with a characteristic framboidal surface texture. In size and form these resembled the pyrite grains already noted in the geological literature by many authors (see Love 1957, pp. 429, 437) from many other beds of dark shale, coal, and indeed certain other rocks more generally associated with their yield of metal ores. At the time, however, the investigation was not carried into these fields. The larger supposed micro-organism, *Pyritella polygonalis* Love 1957, appeared also to have counterparts in such rocks.

The present paper is concerned with furthering knowledge of the palaeontology and ecology of these organisms. Attention is mainly confined to the Rhaetic and Lias of Britain and some supporting studies on Carboniferous and more recent deposits. The Rhaetic and Liassic rocks were chosen as providing accessible and well-known sequences of suitable argillaceous rocks whose general conditions of deposition have already been studied in detail by other authors.

It had already been demonstrated (Love and Zimmerman 1961) that studies of sulphide spheres might with success be applied to such ancient rocks as the Lower Proterozoic Mount Isa Shales of Queensland, Australia, in suggesting the origin of the vast quantities of primary sulphide present there. The slightly metamorphosed condition of the rock, however, added difficulty to the work attempted and precluded much additional understanding being gained of the phenomena observed. In the case of the Permian Kupfer-

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schiefer of Germany, the classic ground of dispute over the origin of pyrite spheres in dark shale, it has been found possible to apply the findings of the present paper and to demonstrate much the same range of micro-organisms in a very similar environment; being concerned with ideas on ore genesis this is published elsewhere (Love 1962).

In the present work the small particle size of some of the organic material imposed a limit to the usefulness of the ordinary microscope, and it is likely that with improved methods and the use of more elaborate equipment much more information including chemical data will become available against which many of the conclusions so far reached may be tested. The subject-matter is of wide significance in the formation of an important type of sedimentary rock.

Full acknowledgement to the work of others is made in the text. Thanks are also due to all those who at various times have provided specimens for special study. The research was carried out in the Department of Geology of the University of Sheffield.

LOWER JURASSIC STUDIES

Source of material. The Westbury Beds provided the Rhaetic specimens and were principally collected from the Garden Cliff section near Westbury on Severn, Gloucestershire, and from St. Mary's Well Bay, Glamorganshire. Lower Lias rocks were taken from St. Mary's Well Bay and the Yorkshire coast at Robin Hood's Bay and near Redcar; Middle Lias specimens from the Yorkshire coast south-east of Staithes; and the Upper Lias succession of Grey Shales, Jet Rock, Alum Shale Series, and Peak Shales from shore sections around Whitby. Confirmatory specimens from boreholes near Whitby were kindly provided by Dr. J. E. Hemingway.

The South Wales and Gloucestershire sections of Rhaetic and Lias are described in detail by Richardson (1905) while the Lias of the Yorkshire coast is described by Hemingway (1934) in general terms and some of the sections in greater detail by Howarth (1955). All these authors paid close attention to faunas but only Hemingway recorded details of the finer microflora of these beds, referring (op. cit., p. 257) to the presence of spore cases, microspores and macrospores in the Alum Shales and Jet Rock Series. Although few preparations to reveal this microflora have been carried out by the present author (necessarily involving a different process from that described below), enough has been seen to show much material present in most of the beds studied, and Mr. D. Wall (personal communication) confirms the presence of spores, algae, and hystrichospheres in many of the Liassic strata.

Remarkably consistent results were obtained from the Rhaetic and Liassic rocks, from which the normal dark to grey, fine or only slightly silty shale and mudstone lithologies were analysed. In subsequent sections, therefore, it has not been found necessary to specify the particular horizons from which described material was obtained.

Description of the pyrite. The Jet Rock is typical of much of the Liassic and Rhaetic strata in the occurrence of pyrite, which X-ray examination showed to be the only sulphide present. There are pyritic replacements of delicate mollusc shells and thin coatings around concretions (Hemingway, *in* Hallimond *et al.* 1950, p. 60), as well as finely disseminated pyrite in the forms with which this paper is concerned, and revealed only by the microscope (Pl. 63, fig. 15). In other beds, such as the Alum Shales, larger irregular masses of pyrite occur. In nearly all the shales encountered, however, from highly to only slightly bituminous, or even slightly silty, the finely disseminated pyrite is abundantly present and it is clearly an important and widespread component of the rock. Although predominant as small single or compound rounded spheres, six types of occurrence are described. Some of these intergrade and precision in distinguishing them is not really possible. The significance of the forms will appear in a later section.

Pyrite type (*a*). Small grains up to 2–3 μ in size varying in shape between angular and spherical. The latter, at the upper end of the size range, may well intergrade with the material of type (*b*). In the case of the smallest grains even determination of their shape becomes difficult. All are scattered throughout the shale as may be seen either from thin sections or from mounts of a poorly disaggregated shale in which thin clots of material remain in their original state. The proportion of type (*a*) is undoubtedly much increased at the expense of other types, and also of concretions, nodules, and shell replacement material after crushing of the rock. The pyrite seen on spores is sometimes in tiny grains but due to their small size a relationship to the spore tissue can seldom be determined.

Pyrite type (*b*). Simple spheres ranging in size from 2–3 μ up to 15 μ or 20 μ. Often the framboidal texture so often associated with this form of pyrite can be observed in which the surface of the spheres is composed of numerous tiny grains or crystals of sulphide. This is a detail only clearly seen in reflected illumination from above a polished surface (see Love 1957, pl. 33, fig. 2).

Pyrite type (*c*). Multiglobular bodies of more complex outline. They show rounded bulges each of the same size range and texture as the individual spheres (type (*b*)) as if compounded of them. In individual isolated multiglobular bodies tested, the component parts are not easily prised apart with a fine needle, and certainly not without rupturing the structure. Their outlines are those of the bodies in Pl. 63, figs. 10, 12. Some of the compound bodies attain a nearly uniform equatorial outline, and only solution will reveal their nature. At the other extreme, however, some are difficult to distinguish from unconnected but touching single spheres. From the nature of the organisms released by solution of the sulphide it is unlikely that many of the instances in which spheres are seen just touching in thin sections and rather dense mounts of isolated material do in fact represent actual organic connexion rather than chance juxtaposition. It is certainly not felt safe to attribute to organic causes all the forms such as chains, rings, and even budding, that might be surmised purely from study of sections of the rock.

The surface texture of the multiglobular pyrite bodies, and also the single spheres at the larger end of their size range, may be rougher when seen in outline, and have a coarser surface texture of large pyrite grains or crystals, than is normal in the framboidal texture. They tend therefore to be reminiscent of the Kiesklümpchen (verezte Bakterien

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EXPLANATION OF PLATE 63

All figs., Liassic material.

Figs. 1–13. Micro-fossils released from pyrite grains in shales. 1, 2, 5–8, 10–13, ×2,000 approx. 3, 9, ×1,000. 4, ×3,000.

Fig. 14. Spore or pollen grains with pyrite coating, ×500.

Fig. 15. Thin section of *P. spinatum* zone shale normal to bedding in ordinary transmitted light; black grains are pyrite, ×250 approx.

Fig. 16. Plant spore with vesicle and corebody, cleared of pyrite, ×1,600.

Typ II) (mineralized bacteria) which Neuhaus (1940, p. 319, fig. 6) described from the Kupferschiefer. The material now described shows graduations between larger and smaller surface grains, possibly corresponding to some extent with the size of the bodies bearing them. Possibly, therefore, some of the Kieslümpchen-types are a particularly coarse extreme of the Kieskügelchen, Neuhaus's normal framboidal sphere.

When seen disaggregated from the rock, a number of multiglobular pyrite grains especially from the more bituminous shales may show shreds of organic tissue attached. It is very probable that from a highly compressed shale containing fragments of plant tissue of many kinds such material may remain adhering to a pyrite grain by chance, so apart from those cases which may appear significant from the prepared oxidation mounts, such occurrences are not pressed as sources of information.

Pyrite type (*d*). Undoubted pyrite on recognizable spores and pollen grains and on other bodies of a similar general shape and material. These bodies (Pl. 63, fig. 14), usually from 30 μ to 100 μ in size, and clearly exceptional in character, may show single isolated grains or spheres of pyrite (types (*a*) and (*b*)) or a mass of spheres to the extent of a complete cover of pyrite. The coarse Kieslümpchen texture of pyrite may be developed in partially covered bodies, perhaps explaining the nature of others of similar texture but completely pyritic. It is difficult to demonstrate by physical means the relationship of the pyrite to the spore or pollen grains. In some cases it is easily detached and leaves no mark or damage; in others an angular depression may be left; and again in others the pyrite may appear to be united with the wall material, for manipulation with a needle only ruptures the whole specimen.

In some of the shales studied spores, pollen grains, and similar bodies with adherent pyrite form a substantial part of that aspect of the microflora, an observation which Mr. D. Wall confirms. They are often individually of a sufficiently high aggregate specific gravity to appear in the heavy fraction of a bromoform separation. So too are many aggregrates of pyrite from the shale containing fragments of organic tissue, and generally of unidentifiable origin, but to which the general description of pyrite as for spores and pollen is fully applicable.

Pyrite type (*e*). Larger pyrite 'microberries' up to 120 μ in size. These are found when the finer fraction from a less intensely crushed rock is examined beneath a high-power binocular microscope. The descriptive name given by Macfadyen (*in* Love 1957, p. 437) is retained. They have a generally rounded shape and are clearly composed of large numbers of smaller spheres, 20–50 μ in size, which give the characteristic appearance. The microberry as a whole is not truly framboidal, but the smaller spheres are. On crushing, the berry breaks down readily into the separate smaller spheres, and these in turn break down into material between 0·5 μ and 5 μ in size, but of this the larger parts are found to be aggregrate, as yet uncrushed, of the 0·5–1·5 μ grains which are therefore basic to the whole microberry. In one specimen provided by Dr. Macfadyen calcite cement held the intermediate spheres together in the microberry; they became separated after effervescence in dilute hydrochloric acid but the intermediate spheres themselves did not react with the acid.

Pyrite type (*f*). Foraminiferids with pyrite infillings. The author's attention was first drawn to the occurrence of such specimens in the Lias by Dr. C. G. Adams. Through the moistened calcareous tests of various Lagenids pyrite could clearly be seen (Pl. 64, fig. 7) and on solution of the shells of such specimens accumulations of pyrite resembling the microberries were released from the chambers, whose internal shape they sometimes retained.

Experimental methods. The observations recorded in this paper concerning the bodies from the pyrite were obtained from two lines of investigation. One consisted of the chemical treatment of small *bulk* samples of pyrite concentrated from a rock, and provided adequate quantities of the final product for proper washing and preparation as permanent mounts on microscope slides. Comparison with a mounted sample of the untreated pyrite then allowed an inference to be made of the relationship between the pyrite and the cleared bodies. For the bulk process, the laboratory methods used were essentially those described by Love (1957, p. 431) and in greater detail by Love and Zimmerman (1961, pp. 883–4). Mechanical disaggregration of the rock sample by grinding was followed by separation of the heavy mineral grains, predominantly pyrite, by centrifuging in bromoform (s.g. 4·9). Unless it was to be examined for material which might be partly calcitic, the heavy residue was then treated for some days with warm hydrofluoric acid to remove traces of adhering light minerals, and when so cleaned it was oxidized in warm or hot concentrated nitric acid or other reagent. After removal of the acid and after several washings of water, the few remaining drops of suspension yielded small micro-organisms, as described below, in proportion to the original yield of rounded pyrite grains. The chemical treatment and the preparation of slides was carried out under strict precautions to avoid contamination. Though useful, bulk solution is not usually a rigorous enough method to determine some of the relationships and in many instances it was found necessary to carry out the process in such a way as to see it actually happening under the microscope.

Virtually continuous observation of the solution of the pyrite was made possible by use of the special cell, only developed late in the course of earlier work by Love (1957, p. 432) and subsequently described in detail by Love and Zimmerman (1961, pp. 884–5). It was possible to confirm that each form of micro-organism described could in fact be obtained in that condition from a particular grain of pyrite from the sediment. An inherent difficulty in this method, however, is that it is frequently impossible to preserve the material revealed and to make a permanent mount. To achieve this it is necessary, without losing a specimen perhaps only a few microns in size, to remove the excess reagent around it and to irrigate the chamber with water to prevent later crystallization of salts produced in the reaction.

This direct observation method obviates the need for elaborate statistical support for such assertions as are made, as was attempted in earlier work (Love 1957, pp. 431–2) and the method is essential if only a limited amount of material is available, for example part of the pyritic content of a single foraminiferid. In the bulk method, large numbers of foraminiferids would be needed for a single preparation. It is also possible to observe the actual position of the sulphide, for instance whether it occurs superficially or within organic material, and to watch for evidence of other soluble minerals such as calcite originally accompanying the pyrite.

Description of the micro-organisms released from the pyrite. The micro-organisms remaining after the removal of the pyrite are varied in appearance but lie in a limited number of groups, some of which are clearly interrelated.

Group 1. Spherical bodies (Pl. 63, figs. 1, 2) 5–20 μ in diameter occurring abundantly, resembling *Pyritosphaera barbaria* Love 1957 (p. 433). They appear to be composed of a tightly packed mass of small cells, 0·5–1 μ, each one of which can be seen during solution to contain a single pyrite crystal. Occasionally, however, the aggregate of cells is looser and often the larger bodies in the size range about 20 μ show this. Polished sections of pyrite spheres in the rock confirm a uniform internal structure. The very thin skin of transparent tissue around the individual pyrite grains does not appear to affect their reflecting power and has not been observed in the sections.

Group 2. Bodies as described above (*P. barbaria*) with a partial or complete thin outer envelope of smoother tissue. If complete (Pl. 63, fig. 4) in the polar view it hardly obscures the characteristic appearance of the microspinose body, but this is no longer clear equatorially. Careful focusing of the microscope at high magnification often makes it possible to demonstrate the complete continuity of the sac below and above the core body. Other bodies (Pl. 63, fig. 2) only bear a shred of such material. The microspinose core body may be considerably smaller than the outer sac (Pl. 63, fig. 6) and less characteristically developed, in many cases appearing as a less perfectly formed mass of microgranular character (Pl. 63, fig. 5). Masses like this, if found isolated, could be included in Group 1 above. Only one core body occurs within any one sac; but the latter often occur in tightly packed compound groups, up to six together being common as well as more complex formations. Here not all the sacs necessarily contain core bodies. The tissue of the sacs is described below (Group 3) and a similar outer wall to the group, as described there, is present here.

Group 3. Hollow sacs whose general appearance is like that described for the sacs with core bodies. Less common, they are transparent to translucent and of a variable smooth to rough surface appearance. In photographs at an equatorial level of focus the walls appear as a thin line or as a rougher, thicker layer of material. The number of cells in the group may again be from one (Pl. 63, fig. 8) to many (Pl. 63, figs. 10 and 12). The outer wall as a whole often appears to be stronger and thicker than the apparently common walls separating individual sacs. Furthermore, these inner walls may not be complete and some instances giving a distinct appearance of cells in division have been noticed. This may best be described as a 'dumb-bell' shape, with a constriction but no actual wall between the cells. Sometimes such adjacent cells may be more or less equal in size but often one is smaller.

Both simple and compound sac forms have been observed while becoming cleared of pyrite. The former come from single spheres. If no well-formed core body is present the pyrite inside the sac may, in its later stages of solution, be irregular (Pl. 63, fig. 13) but is sometimes idiomorphic, though not often appearing to have impressed this shape on the sac from within. Sometimes breakdown into small dark grains can be seen. These may leave either no remnant or microgranular organic material (Pl. 63, fig. 5); it must be stressed, however, that with the empty cells it is in most cases very difficult to decide whether absolutely no grains of core material are present, as distinct from any slight granularity or roughness of the wall. The compound sac forms are released from multi-globular pyrite grains of similar outline or from apparently single spheres, the coarseness of whose outer pyrite may obscure the detailed shape. These multi-globular pyrite bodies and the sac bodies from them appear uncompressed and usually are more or less symmetrical and free from attachment to any other material.

Group 4. Spores and pollen grains, recognizable at least by their general shape, showing, after solution of sulphide, patterns of irregularly developed high-relief ring markings on the surface, observed to correspond with the position of the sulphide. (This is discussed on p. 452.) In some particularly clear specimens examination at high magnification has revealed that the structures, which vary from circular to polygonal, and measure 3–10 μ are vesicle-like developments apparently within the surface layers of the exine, but often this is difficult to decide. Some markings merely appear to be the result of other components of the shale, often pyrite as spheres, pressing in during lithification. The vesicles originally contain pyrite and in some instances it has been possible to witness its disappearance from them during solution. In some of these vesicles microspinose spheres are found (Pl. 63, fig. 16) identical with the core bodies of the isolated sac-bodies, single and compound, described in Group 3 above. Other fragmental organic tissue, generally darker than the spores and of unknown origin, often also reveals round to hexagonal pits or blisters, lighter in appearance, some containing microspinose or roughly microgranular bodies.

Group 5. Small clear granules or particles 0·5–2 μ in size, yielded from the pyrite infillings of foraminiferids and from microberries (Pl. 64, fig. 8). The observation method has been used exclusively. There is no clear evidence of the presence within the pyrite of any organic structure larger than about 2 μ, which is the upper limit of the grains into which the material can be broken, and which form the profile of the intermediate round aggregates that make up the microberry as a whole. Very gentle or very rapid solution sometimes leaves them in large spherical groups, but these appear as loose clusters only and the particles readily drift apart, subject to the movement of liquid and air bubbles around them. Close observation in this freely moving condition is difficult but freshly prepared specimens appear to be small clear round cells, of a rather high refractive index and having no effect upon plane polarized light. Some are perhaps seen in a state of division but most are single, rounded to slightly elongated, and featureless.

Group 6. Many of the smallest grains of pyrite (type (*a*)) in the concentrates dissolve to leave small cell-bodies behind such as are described above, although others dissolve without leaving any trace as would be expected if they are derived from shell and nodule

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EXPLANATION OF PLATE 64

Figs. 1–6, Carboniferous; 7, 8, Liassic; 9, Eocene; 10, Recent material.

Figs. 1, 3. Bodies of *P. polygonalis* type, ×1,400.

Fig. 2. Micro-fossils released from pyrite grain in shale, ×2,000.

Figs. 4, 5. Parts of surfaces of spore material showing vesicular formations and a core body. 4, ×2,000. 5, ×2,500.

Fig. 6. Partly cleared intermediate sphere from pyrite microberry, Scremerston Coal Group, ×3,000.

Fig. 7. Pyrite-infilled foraminiferid, transmitted light, ×30.

Fig. 8. Dispersed cells from solution of pyrite infilling of a foraminiferid, ×2,000.

Fig. 9. Cells from solution of a pyrite microberry, probably internal cast of a diatom, ×2,000.

Fig. 10. Cells from solution of pyrite infilling of a foraminiferid, partly or completely cleared, ×2,000.

pyrite. Similar material has been observed in most bulk-prepared samples among the other organic residues but it is so small that it is not easily studied or recorded except when specially prepared. On some spores and other tissues tiny grains of pyrite have left such small cells but no particular relationship of either the pyrite or the small cells to the host material can be determined.

INTERPRETATION OF THE MORPHOLOGY OF THE MICRO-ORGANISMS

Within the variety of material released from the pyrite, the close relationships which clearly exist must now be discussed. It is convenient to consider Groups 1–4 of organic material together first, and Groups 5–6 afterwards, bringing in, where relevant, evidence obtained from sources other than the Liassic rocks.

Micro-fossils, Groups 1–4. Considering the material from isolated simple and compound pyrite spheres (pyrite types (*b*) and (*c*)) all gradations are to be found, firstly, from the single sac to the complex group and, secondly, concerning organic material within the sac, from the rough accumulation of small grains to the well-formed microspinose body of the *Pyritosphaera barbaria* type, apparently composed of similar grains. From this it is likely that representatives of two developmental series are present, the one from single to compound sacs and the other of growth of core material within the sac, this being then secondary to the appearance of the sac. It is not thought that the cores represent the residue of normal cell contents. The balance of evidence is against the original occurrence in the shale of the *Pyritosphaera barbaria* bodies free of external sacs, for even if observed solution of some spheres reveals no sacs or only shreds of attached material the grinding involved in the original preparation could have stripped off and destroyed them. Less intensely ground material showed more outer sacs. In transmitted light some isolated pyrite spheres show the presence of a very thin translucent skin at the periphery, external to the sulphide but not affecting the reflecting powers of this mineral on the upper surface of the sphere. Also suggesting the outer position of the sac, the appropriate Liassic sphere when its solution is watched does not usually show the loss of pyrite external to the sac. The tendency for the core body, when fairly well developed, to show attachment to one part of the sac wall, in an off-centre position (Pl. 63, fig. 6), is supported by the way in which many examples of *P. barbaria* have a residual polar cap or tail of attached material.

It is difficult not to regard as further manifestations of the same general phenomenon of sacs with core bodies the vesicles on Liassic spores and pollen which often contain *P. barbaria*. These formations have been shown to be equally associated with pyrite. In fact, in view of the discussion below, it may well be that the isolated saccate forms first originated in association with some plant tissue of which no other trace is now preserved.

Further interpretation and explanation will perhaps in due course be made since this aspect of the work also falls within the scope of current research and publication on the microbiological destruction of plant remains by the author's colleagues, Prof. L. R. Moore, Dr. R. Neves, and Dr. H. J. Sullivan, who are concerned with the effects of what appear to be rather similar ring or vesicle structures (present author's terminology) as one of the factors in the degradation of Carboniferous plants, spores, and pollen. The occurrence in some of these structures of core bodies recognizable as *P.*

barbaria strengthens the homology. Through the kind assistance of Dr. Neves the author was able to examine some typical Carboniferous rock material and observed that while normal maceration yielded abundant organic material of this type, when subjected to the methods described in the present paper a heavy fraction of the rock contained large partly or completely pyritized bodies whose characteristic size and shape showed them to be the same spores. Oxidation and solution of the pyrite revealed intensely vesicular exine previously not visible, with *P. barbaria* in some of the vesicles (Pl. 64, figs. 4, 5), but in the specimens actually observed during solution, while it was confirmed that pyrite occurred within the blister-like vesicles, it could not be ascertained whether any organic cores remained.

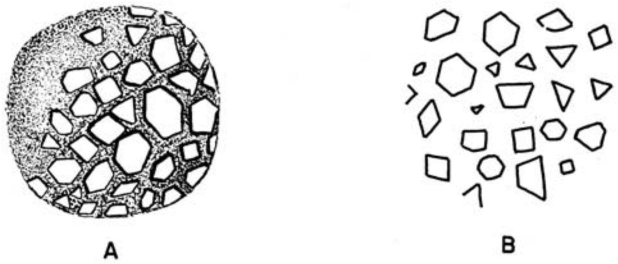
An attempt was made to determine the extent to which this pyritic condition in spores and pollen of both Carboniferous and Liassic age is to be associated with the particular secondary structure noted. The search was carried out on unoxidized material, disaggregated mechanically and by non-oxidizing mineral acids: bromine (Neves 1958, p. 3) was not used. Obviously the spores and pollen were not subjected to the usual cleaning and maceration to clarify their surface nature and observation in the detail required was therefore not easy, especially in Carboniferous material. Nothing was observed which demonstrated the existence of the vesicles in an unpyritized condition, and a close association with pyrite must be assumed.

Various Carboniferous marine and non-marine shales and coals gave large yields of the same range of micro-organisms, with abundant *P. barbaria*, as described above. Free-standing compound saccate bodies were, however, distinctly more common in the Lower Jurassic rocks; obvious core bodies were rarer in the Rhaetic. The cause of these variations is not yet understood. In general chambers free of core bodies, whether isolated simple or compound bodies or on other tissue, are few in relation to the occurrence of mature and poorly formed core bodies.

Reconsideration of Pyritella polygonalis *Love 1957.* After study of the more complex forms of isolated saccate body described above it was found necessary to reconsider the species *Pyritella polygonalis* Love 1957 (p. 434). Although both are obtained from pyrite masses, any similarity between the type material of *P. polygonalis* and the free multisaccate bodies is not immediately apparent. A link was observed, however, through a series of preparations of rocks around the Upper Carboniferous *Gastrioceras subcrenatum* marine band first made for Dr. R. M. C. Eagar (Love 1957, p. 440). Later, more exhaustive study of material obtained from pyrite revealed a range (e.g. Pl. 64, fig. 2) similar to that obtained from the Liassic rocks and also multicellular material showing the characteristics of *P. polygonalis.* This form, in contrast to the diagnosis of the species (op. cit., p. 434), was not always as complete rounded masses but also occurred on fragments of other tissue such as spore exine in the manner of the ring structures already described. Core material occurred in some specimens of this type, some well-developed microspinose bodies of *P. barbaria* being included. On one specimen illustrated (Pl. 64, fig. 3), a margin of unaffected material surrounds the area of polygonal pattern.

Further study of slides containing *P. polygonalis* from the Burdiehouse Limestone of Scotland (the type material) confirmed this observation (Pl. 64, fig. 1) among material which was originally thought to be broken and fragmentary parts of larger bodies. The regular form of the latter appears in all probability to be that of spores and some less-affected specimens of recognizable spores were in fact noted. The description of the cellular structure originally given, however, needs no amendment. So far, no core bodies have been found in this Burdiehouse Limestone material, although this discovery may yet be made. Their rarity here, and relative abundance elsewhere, is a factor not yet explained. The effect of these observations on the taxonomic status of *P. polygonalis* is discussed below.

The particular feature of the polygonal shape to the vesicles, well shown in Pl. 64, fig. 1, and also on the spore material in Pl. 64, fig. 4, appears to be secondary to the formation of the vesicle, derived from the idiomorphic crystal shape taken up by pyrite. The *P. polygonalis* material when pyritized has shown the remarkable Kiesklümpchen texture figured by Neuhaus (1940, p. 319, fig. 6). Here, in distinction from the small regular closely packed small pyrite crystals of the Kieskügelchen (pyrite spheres) the structure is of large, well-spaced angular crystals, up to 10 μ across. In an earlier study of the type material of *P. polygonalis*, where it abundantly occurs, comparative camera lucida drawings (text-fig. 1) were made in reflected light of pyrite crystals from a typical Kiesklümpchen body, and of the polygonal vesicles in a *P. polygonalis* body from the same rock. That the pyrite outlines were all possible random cross-sections or equatorial outlines of the pyrite cube, octahedron, or pyritohedron makes it not surprising that later it was possible, on observing actual solution, to confirm that each pyrite crystal while becoming steadily smaller, at an early stage revealed the polygonal cells in whose centre, under a covering membrane, each one was situated. At the early stage the outline of the chamber corresponded closely with the shape of the pyrite grain and the process often took place without rupture of the surface membrane. In another instance ultrasonic vibration failed to release pyrite from these bodies (or from normal framboidal spheres), again demonstrating its non-superficial position on them.



TEXT-FIG. 1. Comparative camera lucida drawings on same scale of A, *Pyritella polygonalis*, by transmitted light; B, sulphide grains on a Kiesklümpchen body, by reflected light.

In contrast to these forms free of core bodies, the large pieces of spore material such as in Pl. 64, figs. 4 and 5 showed a mixture of pyritic forms from large angular crystals to normal framboidal areas. The latter would be expected where core bodies are present and particularly when they are large. From some present-day data (see Love 1957,

p. 434) it is probable that the pyrite is secondary to some other sulphides and its crystallization may well be governed by physical factors in its immediate environment, to give either a single large crystal in a cell or a framboidal group. Why isolated sacs tend to be filled in the latter way while the vesicles in masses of vegetable tissue tend to the single polygonal form is not understood. Even where a single grain fills a free sac the polygonal shape does not often persist, perhaps because the material is less rigid.

Conclusions concerning Groups 1–4. The two developmental series recognized for the isolated *P. barbaria* and sacs have now been extended to include the other material released from pyrite, some of it clearly secondary developments on plant debris. Of the two possible relationships between the two series the first is that the core material represents the fossilized remains of some investing parasitic or saprophytic micro-organism. In this case the sac is in the position of host. In principle such activity is common at present among lowly organisms, often with a preference for a particular host. Geologically, records are few; that noted by Kidston and Lang (1921) of supposed fungal investment of Devonian plant tissues shows no similarity to the forms under discussion here. In the present instance the host material could perhaps be small algae in the case of the simpler isolated sacs, while in the spores and pollen it could be a pre-existing secondary structure developed in the exine. The somewhat variable proportions of those forms described in different rocks is perhaps a factor in favour of the hypothesis, but the inclusion of saccate forms in which no investing material has conclusively been observed supports the contrary view. This other possibility is that the development of the sacs or vesicles is followed, in a distribution whose controls have not yet been recognized, by development of the core material to form the *P. barbaria* body, all as part of the life-cycle of a single organism.

In either case, both the limited range of the organic forms involved and the environmental evidence to be discussed later imply that a special agency was employed in the pyritization. It may be noted here that over a period of some years the characteristic form of *P. barbaria* has never been obtained by the author from unweathered rock except by the solution of sulphide. The case has already been argued (Love and Zimmerman 1961, p. 891) that the formation of the sulphide ion was part of the life activity of the organisms rather than, after their death, being of external origin or essentially based on the decomposition of their tissue. Furthermore, a distinct association with the degradation of plant material is thought to be shown, in contrast to the subject of the next section.

On morphological grounds, however, the evidence is insufficient to allow a conclusion to be reached on the nature of the organisms. It is not known even to what extent the fossilized material resembles in detail, in its microscopic appearance, its original form. Furthermore, in the combined development series favoured here on the balance of evidence but not regarded as conclusively proved, it is not clear whether the core body has a final separate existence from its sac perhaps as some form of spore. It would not be valid to suppose that it does from the observations so far made, and this would then appear to preclude the individual cells of the core body from being the starting-points of new cycles, the origin of which remains obscure. The observation that the largest forms appear to have a rather looser structure than the others is not accompanied by any evidence of breaking up and dispersing, which would indeed seem impossible in the undisturbed muds below the interface. Alternatively many lowly organisms form resting bodies under certain, usually unfavourable, conditions. Yet these barely seem appropriate as an explanation of the enormous abundance of the material in dark shales where the activity of the organisms has been so intense.

Taxonomically, any grouping together of the microfossil forms described would involve considerable emendation of the existing genus *Pyritosphaera* and the species *P. barbaria* together with probable suppression of the genus *Pyritella* and inclusion of *P. polygonalis* in the emended species of *P. barbaria*. However, because of the complete lack of knowledge of the systematic position of the whole group, together with uncertainty about the various stages within it, and in view of the fact that current research on present-day sediments may ultimately indicate a solution to these problems, it is proposed that no taxonomic emendation should as yet be made.

Micro-fossils, Groups 5–6. Two problems arise in the interpretation of the micro-fossil material obtained from pyritic foraminiferid infillings and microberries and also from widely scattered minute sulphide grains. One concerns distinction between the former group and the pyrite which yields *P. barbaria* and associated forms. The second problem involves, in more detail, the distinction between the individual granules of organic material from the pyrite of foraminiferids and microberries and the granular material of *P. barbaria*. Two factors help to differentiate the material in the first case. The saccate condition does not seem to exist for the 20–60 μ intermediate spheres forming the pyrite masses in the foraminiferid tests and the microberries, although it commonly occurs with the spheres of the *P. barbaria* series. Furthermore, the core body of the latter, when well formed, remains unbroken after the treatment with acid, as if it were a discrete body, albeit a granular one. The opposite is the case for the spheres from the tests and microberries. It is tentatively concluded that different organisms are under consideration.

This is supported by similar evidence from Carboniferous rocks. From crushed and ground marine shale from the *Gastrioceras cancellatum* horizon of the Axe Edge locality, Derbyshire, many unopened ostracods, mostly pyritized, and also goniatite spat smaller than 100 μ in size were picked out, together with microberries. From within some of the closed shells clusters of pyrite spheres were found lying completely out of direct contact with the rest of the rock material and appearing quite different from the smooth pyrite of the shell walls, from which, in the case of the goniatites, they were separated by a delicate tissue of wall membrane. Also perfect internal moulds of goniatite spat were found composed entirely of spheres and devoid of shell. Material of all these types, and also microberries, gave on solution great quantities of 0·5–1·5 μ clear light granules (Pl. 64, fig. 6), comparable with those described from the Lias. Flattened microberries from Eocene strata, provided by Dr. J. W. Murray, and giving a similar product on solution (Pl. 64, fig. 9), were clearly the infilling of diatom tests of characteristic shape. Current work by Love and Murray demonstrates that pyrite infillings of recent foraminiferids, giving a similar product on solution (Pl. 64, fig. 10) may develop soon after death at the mud–water interface whenever de-oxygenated conditions obtain. It is believed that identification of the micro-organisms involved here may be applicable to the Liassic and Carboniferous test-infillings, so consistent is the appearance of the pyrite and the organisms within it. The origin of the spherical microberries, of whatever age, is not yet clear. If they are internal moulds of tests well filled with pyrite, no such tests were found in the rocks examined. It is possible, however, that if the test was originally of some soft organic tissue it may have completely decayed.

The small individual granules of microfossil material (Group 6) obtained from isolated and unattached individual small pyrite grains could with equal likelihood have originated as the partly developed cores in the *P. barbaria* series, or as crushed-down material from animal tests broken or otherwise removed even within the rock, or from microberries which may also have such a source. No means is at present known of recognizing their particular origin.

It is clear that on the basis of the evidence of the pyrite material which must have developed within the tests of animals that a particular association with the degradation of animal material is attributable to the micro-fossil or present-day micro-organisms obtained from it. Again there must be some lowly organism involved and indeed the individual granules are of a size range appropriate for bacterial cocci. It might reasonably be expected that differing micro-organisms would be involved in the decay of plant and animal material. It is not proposed at present to attempt to define the taxonomic position of these micro-organisms.

It is likely that the granular organic remains found, after solution of pyrite, within the central chamber of specimens of *Tasmanites* from Liassic rocks may be assigned to this group. These large and distinctive bodies frequently contained small amounts of granular pyrite and could be picked out from the rock material for individual treatment. In all cases observed the pyrite contained organic material, and similar contents of *Tasmanites* appear to have been observed by Eisenack (1958) but no recognizable body of the *P. barbaria* type was found.

ENVIRONMENT OF FORMATION OF THE SULPHIDE

As a control on the validity of any conclusions concerning the micro-organisms drawn from geological or present-day evidence, the available geological information on the environment in which they flourished must be considered. Whatever their nature, they are very abundant, recognizable as simple pyrite spheres, and most characteristic of sedimentary rocks of the dark shale and sapropelic environment (Deans 1948, pp. 348–50). The value of applying such a simple criterion as the presence of micro-fossils to more complex rock groups has already been demonstrated (Love and Zimmerman 1961). Again, Deans has observed pyrite spheres in lesser quantity in the Northampton Sand Ironstone where Taylor (1949, p. 83) regards the limited pyrite formation as having been controlled by the temporary development of stagnancy.

It may be necessary to modify the view put forward earlier by the author (Love 1957, p. 434) that the spheres and associated forms of pyrite and micro-organisms are associated with bottom conditions entirely inimical to aerobic life. Bottom conditions in fact appear to have differed from rock to rock. In the case of the Pumpherston Shell Bed and Oil Shale of the Scottish Lower Carboniferous sequence evidence was put forward that no aerobic benthonic life existed in the area since only free swimming or floating organisms occur as fossils. In all the beds abundant pyrite spheres were found while the very fine lamination of the sediment was undisturbed.

Related conditions are envisaged by Hemingway (*in* Hallimond *et al.* 1950, p. 68) for the Jet Rock of the Yorkshire Lias. The beds are regarded as representing muds of slow accumulation under marine conditions in waters which at depth were deoxygenated and rich in H_2S; as the sea extended at least to Germany the stillness must have been due to the bottom waters lying below the reach of major wave action in the open sea and therefore at some hundreds of feet in depth. The waters were fully marine, even if somewhat isolated from the main ocean. Similarity with the present state and recent history of the Black Sea is apparent.

But it must be considered whether some property of very fine sediments rich in organic matter might enable a strong resistance to deformation to be attained sooner than is generally thought to occur. Unpublished evidence from the author's collection of thin sections of Carboniferous shale sediments suggests that disturbance of a shale layer before deposition of the next may give small disarranged pellets with characters such as the cryptophyllite texture (Dunham *in* Eagar 1952, p. 358) already in their final relationship to the lamination of the pellet rather than that of the undisturbed shale. There is evidence from present-day organic-rich dark muds (J. W. Murray, personal communication) that even at the mud–water interface fine organic material binds up the clay sediment into a relative firmness. Furthermore, mucous-covered burrowers, such as *Arenicola*, which are normally prodigious ingesters of sediment, are strongly inhibited by the presence of much fine clay (Reid 1929). After deposition of the mud these factors might lead to an initially localized deoxygenation whose products would in turn lead to the beginning of the more generally foetid conditions by which other varieties of aerobic burrowers were also inhibited. It may not be necessary therefore to invoke very great depth, or alternatively, unusual climatic conditions such as the absence of all but the gentlest of winds. Some climatic factors, however, may have induced other conditions favourable to the maintenance of long-standing stagnancy over a wide area.

Hemingway regards the Jet Rock as the initial part of an Upper Lias cycle of sediments produced by the shallowing of the deep sea basins which had originated under tectonic control. At the top of the Upper Lias cycle the sands of the Blea Wyke Series yield 'frequent moulds of lamellibranchs, *Pentacrinus* debris and *Serpula* and are characterised by an abundance of worm tubes which ramify through the rock' (op. cit., p. 69). This indicates intense scavenging activity able to remove all digestible organic material from the sediment, an activity which is normal in nature when no restraint is present but which must have been absent in the muds which ultimately gave the shales at the bottom of the sequence. These shales, however, show a steady gradation upwards (op. cit., pp. 68, 69), becoming less pyritic, less well laminated, less dark, and less bituminous; they were probably formed 'in a marine environment which was becoming less stagnant and presumably less deep.' In the Alum Shale Series microscopic pyrite is still found but occasional sideritic mudstones and calcareous concretions appear. Counts of grains in thin sections of typical specimens of the Hard Shales above the Alum Shales, and the Bituminous Shales below, show that the proportion of pyrite spheres and multispheres is about half the value in the Grey Shales, Jet Rock, and Jet Rock Shale. The Alum Shale Series is compared by Hemingway with the Grey Clay of the Black Sea, laid down in an environment appreciably shallower than that of the sapropelic muds and nearer the shore.

There are two ways in which residual organic material may occur in a sediment, giving a sapropel. Firstly, rapid deposition of organic material without much sediment at a rate in excess of that at which it can be utilized by mud-grubbing and burrowing organisms may cause some to be buried below their reach. Van Andel and Postma (1954) note that the maximum formation of sulphide in the Gulf of Paria at the present time is associated with such conditions, even where fairly strong currents renew the bottom waters. The same is recorded by Neeb (1943) from the East Indies. Secondly, toxicity in the sediment, due to the effects of stagnancy and lack of circulation, will inhibit or prevent at least aerobic organisms. This is due to the exhaustion of available oxygen for oxidation processes by organisms and the ensuing establishment of a chemically 'reducing environment'. It is believed that in the case of the shales in the Upper Lias sequence rapid deposition was not the case: nevertheless the toxicity must have been confined to the well-laminated and undisturbed sediment, for undoubtedly a bottom surface-dwelling fauna was present, however peculiarly limited in variety.

Besides free-swimming ammonites and pelagic micro-plankton living in the upper water, *Inoceramus* is prominent among thin-shelled lamellibranchs, but not common, although it increases in abundance in the Bituminous Shales 'where some horizons become oyster beds' (J. E. Hemingway, personal communication). In the Alum Shales *Nuculana ovum* is found.

In the Rhaetic beds at Garden Cliff and the corresponding ones at Lavernock, Richardson (1905) has presented clear evidence of a bottom-dwelling fauna even from the black shales, including *Pteria contorta*, *Pecten valoniensis*, *Protocardium rhaeticum*, *Gervillia praecursor*, and *Modiola minima*. Hemingway (*in* Hallimond *et al.* 1950) regards this cycle, initiated by the Westbury Beds and passing up through the Sinemurian into the Lower Pliensbachian ironstone shales, and also the Middle Lias cycle culminating in the Cleveland Ironstone Series, to be less well developed than the Upper Lias cycle. Both Hemingway (op. cit., p. 70) and Raymond (1955, p. 16) regard the Rhaetic sea as having been shallower and more restricted in area than that depositing the Jet Rock.

It is probable, then, that the zone of formation of the forms of pyrite described in this paper is within the mud rather than at the mud–water interface of the sediments giving the Lower Jurassic succession. Although a 'sulphuretum', a zone charged with hydrogen sulphide and inimical to aerobic life, may have been present this need not necessarily have lain in the bottom waters if oxygenation of these was sufficient to remove that proportion of the toxic dissolved hydrogen sulphide which passed upwards out of the mud. Perhaps the limited fauna of the Jet Rock is indicative of the effect of the most extreme conditions of toxicity reached in this Series and undoubtedly conditions relaxed in the succeeding Bituminous Shales. Some parallel to this may perhaps be seen in the study by Eagar (1953, p. 341) of the non-marine lamellibranchs in shales of the Upper Carboniferous Lower Foot Mine succession of Goyt's Moss, in which much of the microscopic pyrite occurs as the small spheres under discussion in the present paper. In a series of beds of uniform grain size and composition, distinct changes of shell shape are related by Eagar, with 'probable significance', to the maximum occurrence of pyrite. Despite the latter, the benthonic population of the area appears to have continued. In fact the change downwards from aerobic to anaerobic conditions can take place at any level in the mud and in this connexion references to studies on the behaviour and effect of bacteria in muds were made in earlier papers (Love 1958, p. 435;

Love and Zimmerman 1961, p. 891). Whatever the present organisms may be, the only available records concern bacteria and for these it is clear that at depths of up to fifteen feet into fine subaqueous muds laid down in the absence of current action it is probable that sufficient circulation or migration of iron can occur to allow for much biochemical change and that the chemical system is not a closed one over the distances involved.

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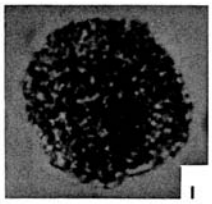
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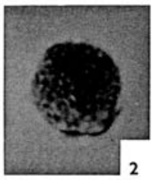
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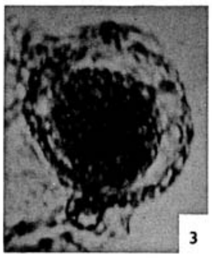
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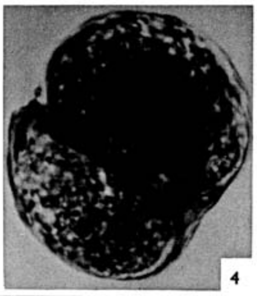
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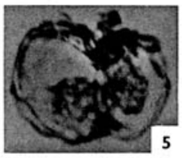
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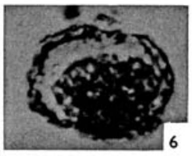
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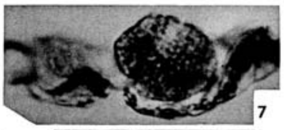
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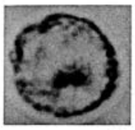
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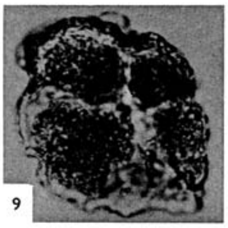
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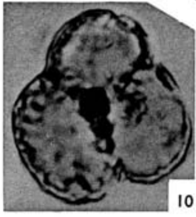
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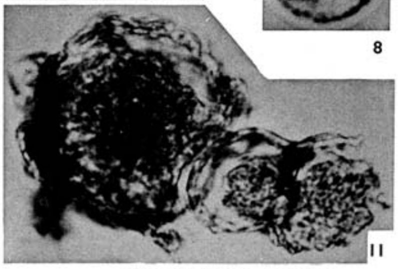
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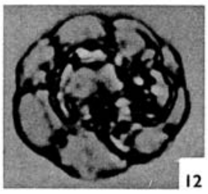
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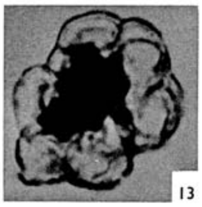
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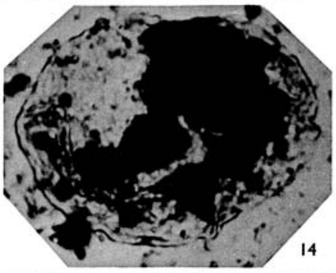
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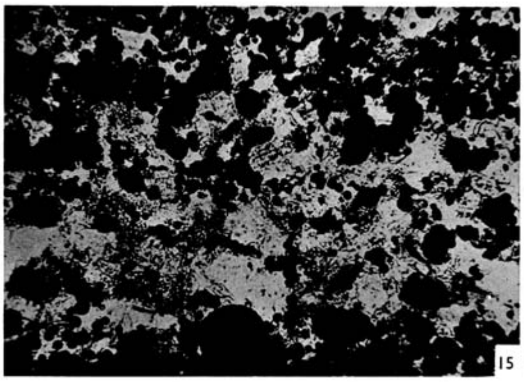
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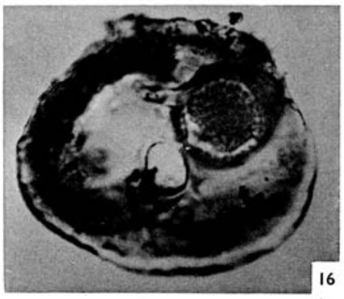
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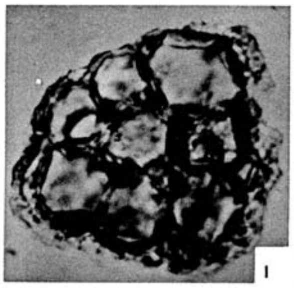
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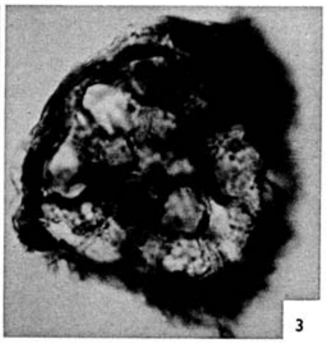
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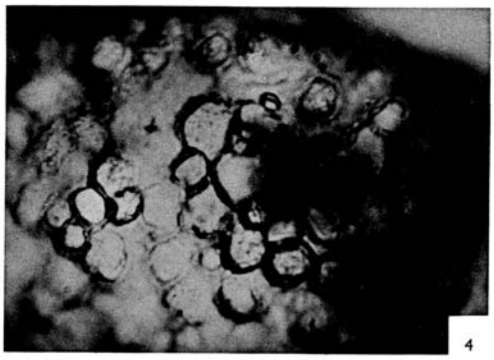
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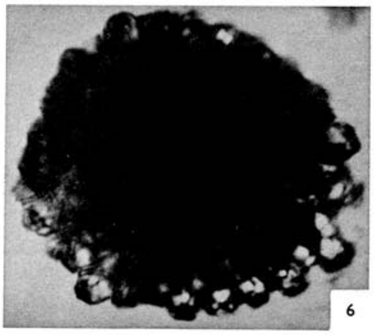
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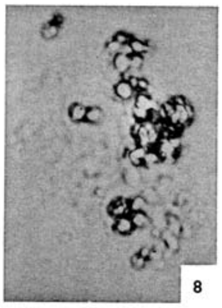
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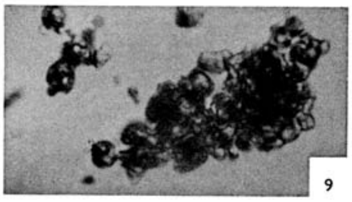
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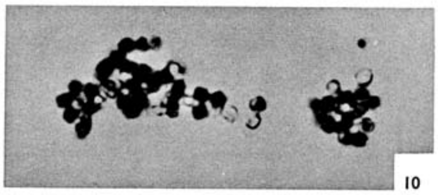
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