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4. Spherulites and Allied Structures

Part I.

BY

W. H. BRYAN, M.C., D.Sc.

Spherulitic Crystallization as a Mechanism of Skeletal Growth in the Hexacorals

BY

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SPHERULITES AND ALLIED STRUCTURES.

PART I.

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SPHERULITES AND ALLIED STRUCTURES.

Part I.

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(PLATES III., IV., V., AND 8 TEXT FIGURES.)

(*Read before the Royal Society of Queensland, 30th September*, 1940.)

I. INTRODUCTION.

This paper may be regarded as an extension and elaboration of a study entitled "Some Spherulitic Growths from Queensland" published a few years ago (Bryan, 1934).

Since the earlier work was completed a much larger and more varied collection of specimens has been assembled and other occurrences have been investigated in the field. The material available for study now includes many hundreds of individual spherulites, ranging in size from miscroscopic examples to giants over 3 feet in diameter. The complete collection has been obtained from south-eastern Queensland.

II. DEFINITIONS.

The term "spherulite" is used in this paper in the original sense of Vogelsang in 1872 as interpreted by Holmes (1928) as follows:—"A radiating and often concentrically arranged aggregation of one or more minerals, in outward form approximating to a spheroid, and due to the radial growth of prismatic or acicular crystals in a viscous magma or rigid glass about a common centre or inclusion."

This definition is satisfactory in so far as it excludes many growths that are often loosely described as "spherulites," but for which the more general term "radial aggregates" is perhaps more appropriate. These are commonly seen in such minerals as wavellite and tourmaline. The definition rightly excludes, too, concretionary and oolitic structures closely analogous to spherulitic growths, but having a very different origin. For these a special nomenclature has been suggested by Bucher (1918).

On the other hand, a rigid application of the definition excludes a number of structures that in physical properties and in origin are quite obviously related to the spherulites proper. Thus insistence on radial structure would exclude similar growths in which the component crystals were merely divergent or quite parallel. Similarly, insistence on spheroidal form would rule out a host of allied forms of various shapes.

These difficulties may be met by using the term "spherulitic" in a somewhat liberal sense, but a more serious difficulty remains. There are found closely associated with spherulites proper and formed under similar circumstances bodies in which the shape is not determined by the growth in the way the definition demands. Such bodies may take various shapes (including spheroidal) and exhibit various crystalline structures (including spherulitic). For these bodies the word "spheruloid" is suggested as a convenient and appropriate term.

III. FORM AND STRUCTURE.

(A) GENERAL CONSIDERATIONS.

In a spherulite *sensu stricto* there is a simple and essential relationship between the internal radial structure and the external spherical shape. The one actually produces the other. Similarly, in all the admissible variations of the spherulite there is a definite causal relationship between the several internal structures and the respective external forms. In all cases, this relationship shows itself as a surface everywhere at right angles to the growth of the component crystals. Thus just as spheres are formed by radial growth about points, cylinders (axiolites) will be produced by divergent growth along axes and tabular forms by parallel growth upon plane surfaces. Varying combinations of these regular structures may give rise to many irregular forms in no sense spherical, produced by structures in no sense radial, yet in all of which the form is controlled by the structure, and to all of which the term "spherulitic" may be applied.

In many spherulites the outer surface consists merely of the distal ends of the radial fibres and is thus a ragged or even a prickly structure, firmly embedded in the ground mass. Such indented spherulites may be found both in natural and artificial glasses (figure 3a). But in many other cases and particularly in larger spherulites the outer surface consists not of radial fibres but of a concentric "skin" that effectively encloses the spherulitic material (figure 3b).

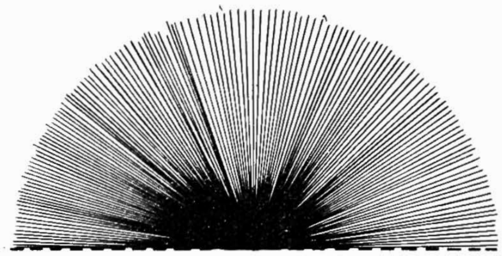

Fig. 3a.

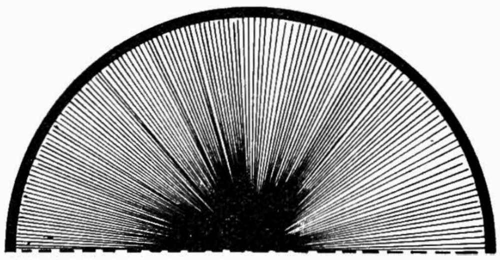

Fig. 3b.

TEXT FIGURE 3.

Considerable variation in spherulitic structures is brought about by the arrangement of the fibres. These may be arranged either as *simple* growths giving unit forms showing uniformity of pattern within the spherulite (figure 4a) or as *composite* growths in which the spherulitic forms are made up of numerous mutually interfering repetitions of the unit pattern (figure 4b).

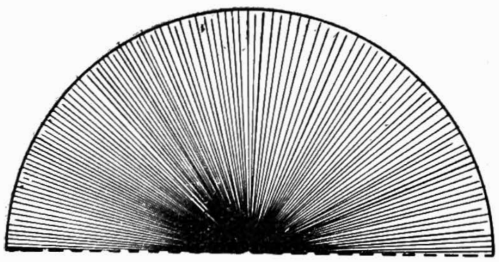

Fig. 4a.

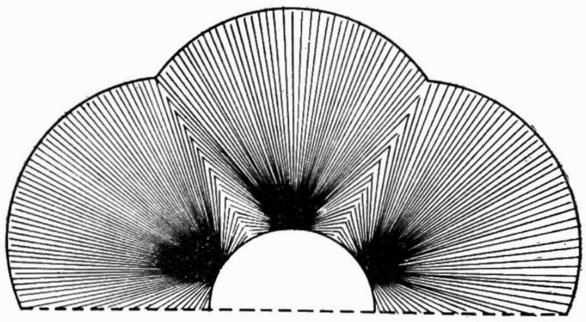

Fig. 4b.

TEXT FIGURE 4.

Since it would be tedious to consider all the possible variants of spherulitic growth, attention will be focussed on the more regular forms, consideration being given first to their production as a result of simple spherulitic growth.

(B) SIMPLE SPHERULITIC STRUCTURES.

The simplest type of radial growth is the simultaneous extension of numerous slender crystals to produce an open structure like the quills of a porcupine. Such growth is approached by some small spherulites of both natural and artificial glasses, but is not characteristic of the larger spherulites.

In simple spherulitic growth, as usually developed, the radiating fibres multiply in number as they grow outwardly. This may be due to the progressive insertion of new individuals in the spaces between the older fibres, but more commonly it appears to be due to the branching of the outer ends of crystals already formed. The branching may be simply dichotomous, thus increasing the number of fibres by geometrical progression, or may be less regular, resulting in a somewhat complex radial tissue.

The spherulites formed in these ways range from those in which the fibres are so closely arranged as to give a solid appearance, to loose spongy spherulites, while every intermediate type is to be found. True (spherical) spherulites show radial structure and branching of fibres in all planes corresponding to freedom of growth in all directions (figure 5a).

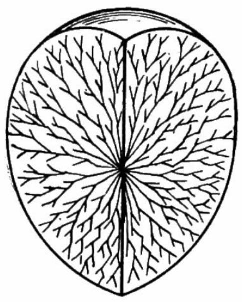

Fig. 5a.

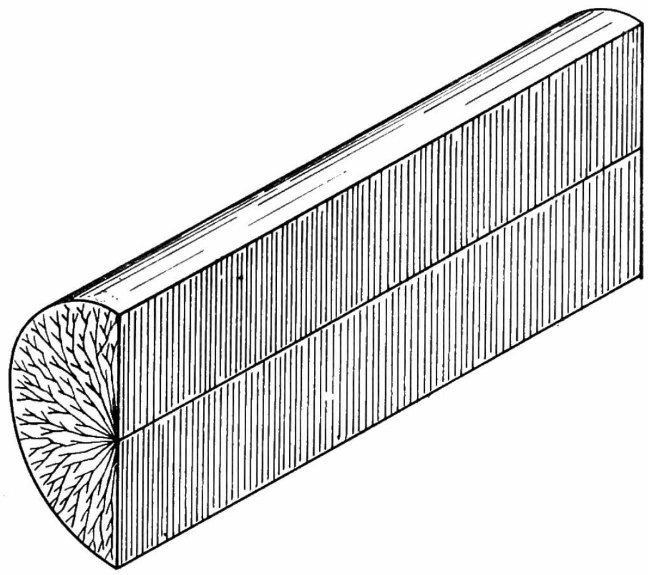

Fig. 5b.

TEXT FIGURE 5.

Simple spherulitic growths about axes, although theoretically possible, are uncommon, as they require the simultaneous initiation of crystallisation at all points along the axis. When this does occur there results an internal structure differing from that of the simple sphere, in that the radial growth is confined to planes transverse to the axis, longitudinal planes showing parallel growth. For the same reason, branching of the crystal fibres can take place, too, only in transverse directions. Such a structure, combining radial and parallel growth, may be described as *divergent* (figure 5b).

Simple spherulitic growth upon a plane surface is even more rare. When it does occur there is no sign either of radial or divergent growth, the component crystals showing a strictly *parallel* arrangement. In this case there is no possibility of the branching of crystals in any plane.

(c) Composite Spherulitic Structures.

These consist essentially of simultaneous spherulitic growths from a number of points scattered along an axis or upon a surface. (Obviously, growth of this type cannot take place about a point.)

In the earliest stages the growth about each of the several centres will tend to be independent of that about neighbouring centres, and will follow the lines indicated for simple spherulitic growth. But a stage will soon be reached when mutual interference between adjacent growths will be set up, and the spherulite will grow thereafter as a composite structure.

Composite growth about an axis is commoner by far than simple growth in the same circumstance. The result of such growth is to produce the caterpillar-like structures that are so characteristic of the larger axiolites (Harker, 1909)* (figure 6a).

Composite growth upon a plane is also far commoner than simple spherulitic growth in such a position, and results in the production of numerous closely appressed parallel columns, commonly hexagonal in cross section. These may be capped by cupola-like structures (figure 6b) or may merge into one general surface (figure 6c).

Whether they be initiated along an axis or upon a surface the number of centres about which growth will begin and their arrangement may well be haphazard, in which case a somewhat irregular structure, composed of closely appressed columns of different sizes, would result. But it must be remembered that spherulitic structures are, in fact, often amazingly regular and possess an almost perfect geometrical symmetry. In this connection, it is of interest to consider the case of composite spherulitic growth upon a spherical surface, and to inquire into the nature of the pattern that would result from regular spherulitic growth about a number of points evenly and symmetrically distributed about the surface of the initial sphere. There would appear to be many possible schemes of distribution that would satisfy these conditions, but a further requirement would also need to be fulfilled—namely, that each component growth be as nearly circular as possible, for each component would, if unconfined, tend towards the form with circular cross section. In other words, the polygons bounding the component growths should each have the largest possible number of sides.

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* Harker doubted the existence of axiolitic growths and suggested that such as had been reported might really represent cross sections of spherulitic growths about plane surfaces. Composite axiolitic growths are, however, common in the collection under review.

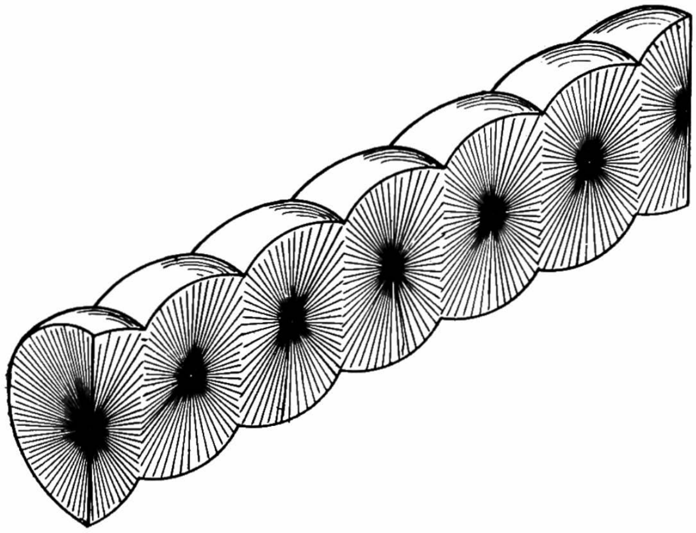


Fig. 6a.

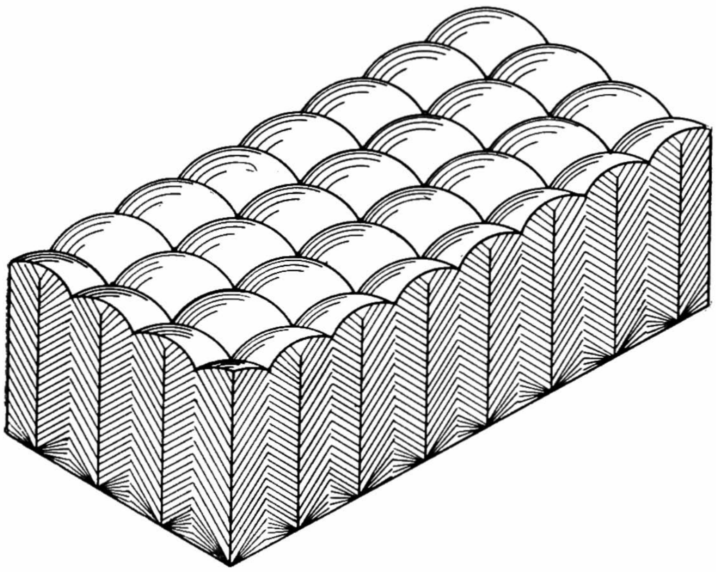


Fig. 6b.

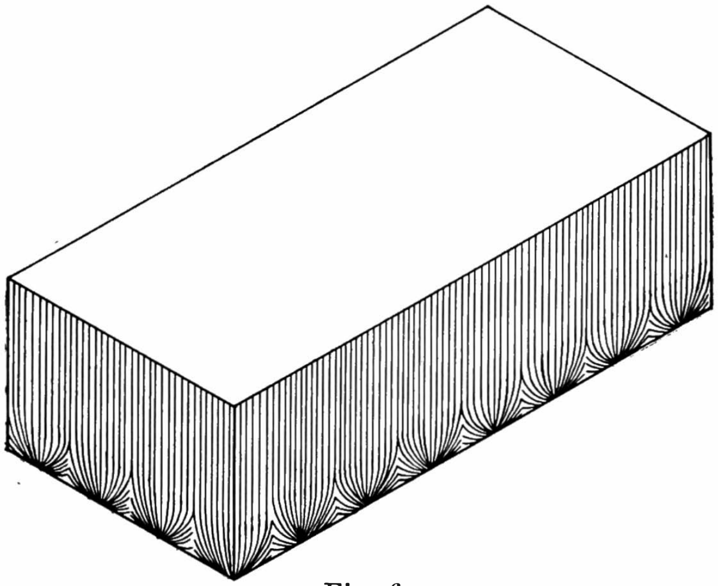
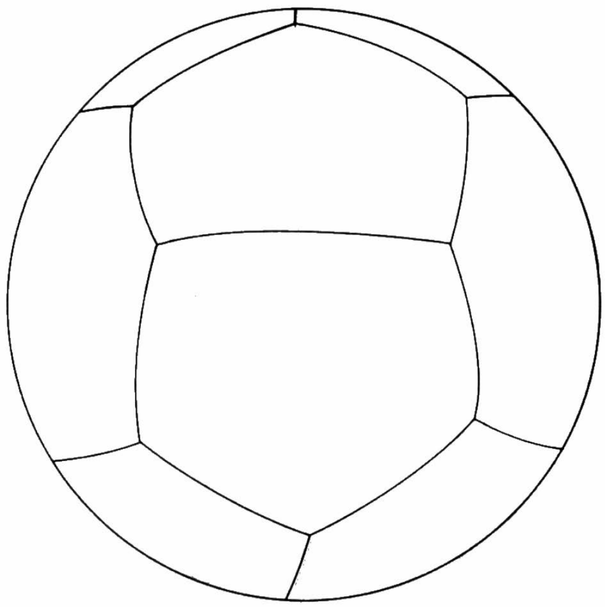


Fig. 6c.
Text Figure 6.

In so far as distribution alone is concerned, considerations of symmetry suggest that the hex-octahedron or the tetra-hexahedron (projected upon a sphere) would form a suitable basis of likely patterns, but these do not satisfy the second condition as, in both cases, each face has only three sides. The rhombic dodecahedron is better with four sides to each face, but (in the absence of any available symmetrical figure with six-sided faces) we are left with the pentagonal dodecahedron (pyritohedron) as most nearly satisfying both conditions.

Some of the specimens in this collection closely approach this ideal geometrical structure, being made up of twelve closely appressed pentagonal columns, while other specimens can readily be regarded as imperfect examples of the same phenomenon (figure 7).



TEXT FIGURE 7.

Composite spherulites when examined in section often show an intricate and apparently confused mass of spherulitic tissue. This intricacy is due to irregularities of growth of many kinds, some of which may be traced to the mutual interference of adjacent components. Such interference manifests itself in a variety of ways, but for the most part these may be regarded as variations of two distinct types of growth. These may be termed *tufted* and *plumose* respectively.

In growths of the first or tufted type, the centres from which the radial growths are directed remain fixed in their original position, so that as composite growth proceeds the lengthening radiating fibres of neighbouring components become more and more nearly parallel, and the whole growth moves toward a unity and homogeneity that resembles more and more closely the simple spherulitic growth. At the same time, the outer surface becomes more and more nearly spherical (figure 8a).

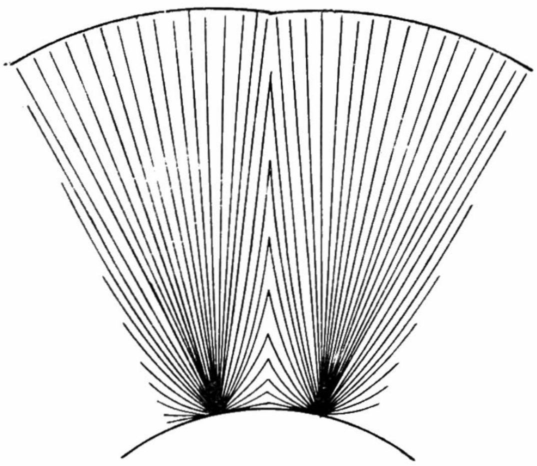


Fig. 8a.

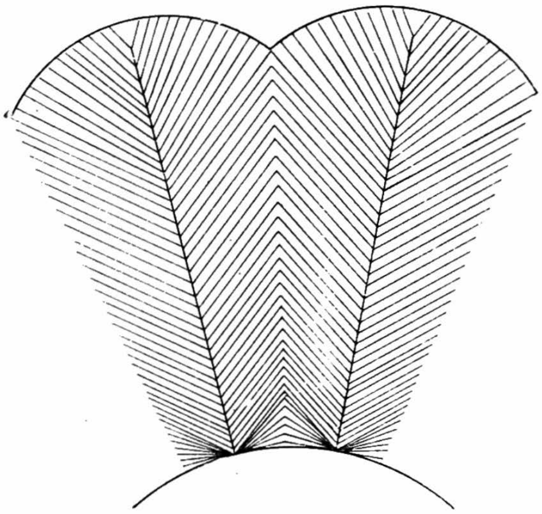


Fig. 8b.

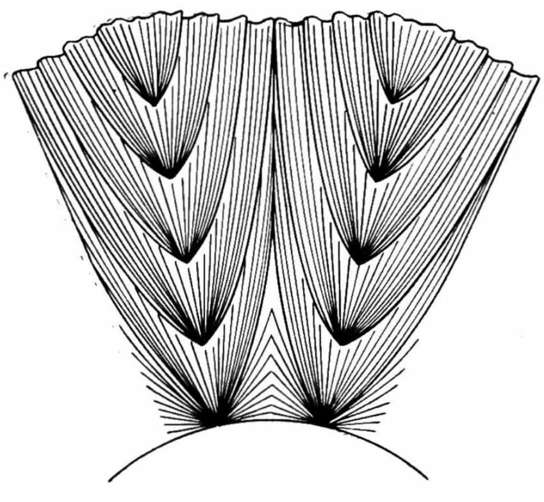


Fig. 8c.
TEXT FIGURE 8.

In examples of the second, plumose, kind the apparent centres of radial growth move progressively outward from their original positions. As a result of this movement, neighbouring components remain as antagonistic as when they first interfere and the structure continues to grow as an obviously composite one with a characteristic bulbous outer form (figure 8b).

Many variants, both of the tufted and plumose types of composite growth, are to be found. One of the most interesting of these occurs when the centres of radial growth move outwardly in spasmodic fashion producing tufts, each embedded in an earlier tuft, the whole giving rise to external patterns of the rosette type (figure 8c). Another feature of this variant is the ease with which the outermost tuft may be removed, leaving behind a conical cavity.

(D) RELATIONSHIP BETWEEN SIMPLE AND COMPOSITE SPHERULITIC STRUCTURES.

For the purpose of the foregoing analysis, simple spherulitic structures and composite spherulitic structures have been treated separately, as though distinct and mutually exclusive, but this is far from the fact. It is true that numerous observations show that spherulitic growths about points are essentially simple in character, whereas spherulitic growths about axes, planes and curved surfaces are dominated by composite structures.

Nevertheless, change from simple to composite structure during the growth of a radial spherulite is common enough, while the change from composite to apparently simple structures is by no means rare. Changes of both types are particularly marked immediately after pauses in the growth of the spherulite.

Although all strictly radial spherulites must be initiated as simple spherulitic growths, it sometimes happens that, in their later stages, new centres of growth are set up within the tissues of the ever expanding spherulite, and from these centres outwardly divergent growths consisting of sheaf-like clusters, or conical bundles of crystals, are insinuated (figure 9a). In the outermost parts of a large radial spherulite these may be so numerous as almost to have obliterated the simple radial structure.

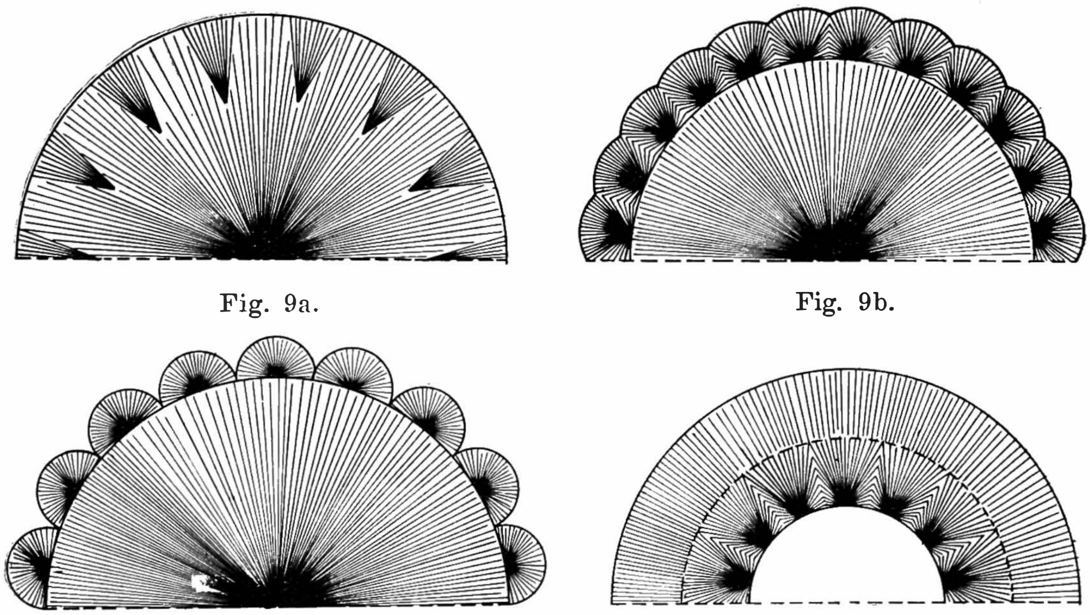


Fig. 9a. Fig. 9b.

Fig. 9c. Fig. 9d.

TEXT FIGURE 9.

An even more marked change is sometimes brought about after a pause in the growth of a simple spherulite. After such a pause, growth may be continued as a series of composite structures (figure 9b). In extreme cases, and probably after a longer or more serious interruption, the later growths appear as a second generation having little in common with the original spherulite (figure 9c).

On the other hand, adjacent spherulitic growths, as has been shown earlier in the paper, may sometimes become more and more nearly parallel and in the end give rise to what appears to be a uniformly homogeneous simple spherulitic structure (figure 9d).

IV. CONDITIONS OF GROWTH.

(A) RADIAL GROWTH.

In as far as spherulites are essentially radial structures they are essentially growths of a single mineral. The presence of a second mineral in no case contributes towards the radial growth. At best, the second mineral may possibly accommodate itself to the structure of the growing spherulite and thus reinforce it. Commonly, it may modify the radial outgrowth, by emphasizing concentric or annular structures. At worst, it may actively interfere with, and ultimately inhibit, the radial growth, replacing it with an inter-growth of the two minerals concerned.

It is generally agreed, as a result of observations in many parts of the world, that the radial structures characteristic of spherulitic growth are due to rapid crystallization in a highly supersaturated and very viscous solution. Little advance on this position appears to be possible on the available geological evidence, but the work of Morse, Warren, and

Donnay (1932) on "Artificial Spherulites and Related Aggregates" is clearly relevant and their conclusions definitely significant to our problem.

These authors point out that "spherulites can be formed of many substances if the reacting solutions are allowed to mix by diffusion avoiding all convection." They state, further, that: "Little is known regarding the factors which cause a substance to crystallize as a spherulite as against a number of normal crystals or a single crystal. It is, however, suggestive that the presence of a gel appears to be highly favourable to the growth of artificial spherulites. Furthermore, it appears to be true that the radiating fibrous masses of a number of minerals have also developed from a gelatinous state. The fact that the spherulites of volcanic rocks as well as those found in slowly cooled artificial glasses must have grown in media of high viscosity at once suggests the possibility that viscosity has an effect somewhat similar to that of gels in producing the spherulitic habit. It is altogether possible that the effect of the gel or of viscosity is to impede or perhaps prevent convection during crystallization."

(B) Concentric Growths.

Concentric structures, although not essential to spherulitic growth, frequently accompany it.

These show so many variations that they may presumably be caused in any one of several ways, but all are due to interruption of some kind to the otherwise continuous and uniform process of radial growth.

Structurally, the various concentric growths appear to fall into three categories. In the first, we may place those in which there is a sudden change in the nature of the radial growth, but in which there is no structural hiatus. In the second group, there is definite but irregular discontinuity separating several apparently independent concentric shells. In the third category come those examples where successive shells of spherulitic material regularly alternate with some intervening mineral substance.

The three types of concentric structure may be referred to shortly as (*a*) Interrupted, (*b*) Lithophysal and (*c*) Rhythmic.

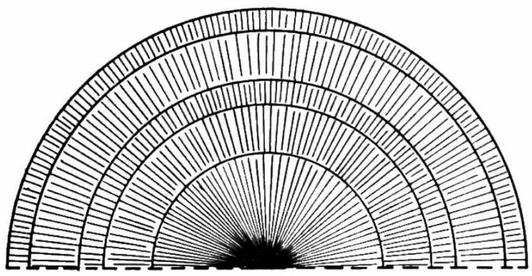


Fig. 10a.

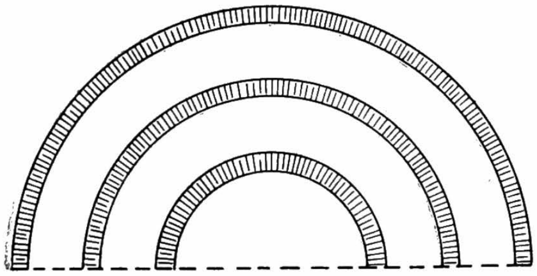


Fig. 10b.

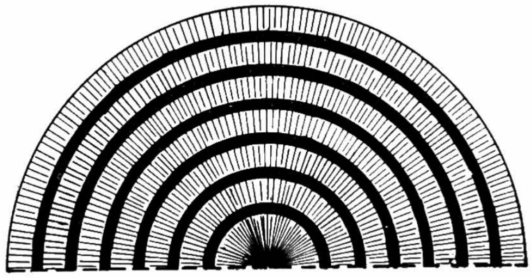


Fig. 10c.

Text Figure 10.

The origin of concentric growths in general, and in particular, the question as to whether the three different categories are due to three different processes, may now be considered.

In the case of the first type (figure 10a), slight pauses in the process of crystallization would seem an adequate reason for changes in the nature of the radial growth striking enough to introduce a concentric element into an otherwise dominant radial pattern. Such pauses might be due to rapid crystallization having brought the immediate source of supply down to saturation point. In this case, crystallization would be resumed as soon as diffusion of supplies through the surrounding medium had brought about the necessary concentration.

Interruptions such as these were observed by Morse, Warren, and Donnay (1932) who noted that pauses in the growth of artificial spherulites were often accompanied by a change in the size of the constituent fibres.

Concentric structures of this first kind are to be expected in natural spherulites, too, in view of the rapid rate of spherulitic growth and the slow rate of diffusion in the viscous medium.

But concentric growths of the second kind (figure 10b) cannot be explained as simply due to slight pauses in crystallization following transient changes in the immediate environment. They appear to differ from the growths found in the first category, not merely in degree but in kind. In particular, and as distinct from the preceding class, the concentric structure is far more conspicuous than the radial.

The best known examples of this second group are to be found in the so-called "lithophysae." In their simplest form, these consist of a hollow thin-shelled structure, such as are to be seen in the "hollow spherulites" of Cross (1891) and the spherulitic "bubbles" of the writer (Bryan, 1934). More commonly, they are composed of a series of such shells arranged concentrically, each shell exhibiting radial spherulitic growth and being separated from its fellows by a notable gap. Such abrupt discontinuity of structure must represent serious discontinuity of process. Indeed, where in the first type of concentric growth the radial structure is almost continuous, in this second type the hiatus between successive shells is such as to suggest that each individual shell is to be regarded almost as an independent unit.

The origin of lithophysae has been the subject of much debate and several different hypotheses have been advanced. These have been canvassed elsewhere (Wright, 1914), and it is unnecessary to go over the ground again, but it would seem that a simple and adequate explanation of the single-shelled bubble-like lithophysae would be to regard them as films of spherulitic material deposited about gas bubbles escaping from the cooling lava. But here one should recognise the possiility that part, at least, of the gas contained in the central cavity may have been liberated from the spherulitic material during crystallization and that the bubble is to that extent self-inflated.

The exact manner of growth of the more complex lithophysae cannot be so simply explained, but it would seem to have depended largely on the liberation (perhaps the rapid liberation) of gas from the lava. Indeed, some of the lithophysal masses in the collection on which this paper is based approach, in their thoroughly cavernous nature, pumices and similar rock froths.

In the third type of concentric growth (figure 8c) the most conspicuous feature is due to the regular alternation of concentric shells of radially crystalline felspathic material with shells of a second material that shows no sign of radial growth.

This type of structure has been described in detail by Mourant (1932), who regards it as due to rhythmic precipitation of the felspar. According to Mourant's argument, the necessary conditions for such precipitation would not usually be attained, so that rhythmically banded concentric structures should be uncommon. This is in keeping with the writer's experience, for in a collection of several hundred specimens very few good examples have been found.

Of the three kinds of concentric structures dealt with above, all may be explained, more or less satisfactorily, in terms of interrupted growth of one mineral. But the possible effect of the simultaneous crystallization of a second mineral should not be neglected in view of analogous structures in other fields supposedly having been produced in this way.

In this respect, the work of Schade (cited by Bucher, 1918) on gallstones is of interest. After pointing out that both radial and concentric crystalline growths are found in gallstones, he states that the difference between spherites of radial and concentric structure depends on the amount of other substance thrown out simultaneously with, and mechanically enmeshed in, the growing structure. Schade found that natural cholestrin gallstones, when 80 to 90 per cent. pure, show a radial crystalline structure, while gallstones containing 25 per cent. or less cholestrin exhibit perfect concentric lamination.

It would seem from these researches that the simultaneous crystallization of a second mineral might not only interfere with the radial growth of the first, but might actually bring about concentric growth, although the operating mechanism is far from clear.

The science of metallography, too, furnishes interesting structures that may possibly be analogous. Thus, Van der Veen (1925), in dealing with eutectic mixtures of metals, states that "The deposition of both components may follow simultaneously when for both the factors of crystallization as e.g., the nuclei numbers and the linear velocities of crystallization are the same. If these factors differ, one of the phases will separate first and deposition will proceed periodically alternatingly." Again, "Oscillating between the two points of supersaturation alternate layers or thin rectiplanar lamellae will form." Such an arrangement is exemplified by the eutectic "pearlite" in carbon steel.

Rosenhaim and Haughton (1935) point out that "pearlite" in its characteristic alternate structure "bears out its analogy with the normal eutectics which are also typically laminated."

(c) Graphic Growth.

Although not represented in the material in the writer's collection, the close relationship of spherulitic crystallization with the development of graphic and granophyric structures has long been known.

Harker (1909) was of the opinion that "The spherulites of the acid igneous rocks fall into two chief classes, according as the radiate growth is constituted (*a*) by graphic intergrowths of felspar and quartz, or (*b*) by felspar fibres only."

Teall (1888) argued that the graphic intergrowths commonly seen in acid rocks indicated simultaneous crystallization of eutectic mixtures of quartz and felspar, and that since spherulitic structure was often associated with graphic structures it, too, indicated eutectic crystallization.

The metallographers, using a similar argument, also interpret the combination of spherulitic and graphic structures as due to the simultaneous crystallization of two metals present in eutectic proportions, the one showing spherulitic growth being regarded as the "dominant" partner. Thus, Rosenhaim and Haughton state that "In the case of the lead-tin alloys the tin is the predominant metal, and each of the crystals of the eutectic is in reality a radiating structure, known as a 'spherulite,' of tin carrying the lead in its interstices."

But, as Harker (1909) long ago pointed out, it is wrong to think of a spherulite as forming at a point of time, for it represents growth from a centre. It appears to the writer that the very existence of spherulites is disproof of simultaneous crystallization, whether in rocks or alloys. It would seem that both the "graphic spherulites" of Harker and the lead-tin and other alloys that show a combination of spherulitic and graphic structures may be explained simply in terms of one substance (felspar in rocks, the "dominant" mineral in alloys) starting to crystallize alone, and after having established a spherulitic structure being accompanied by the second substance. Under these circumstances, the spherulite might continue to grow with crystals of the second substance in its interstices, but in some cases the simultaneous crystallization of the two substances would express itself as a graphic structure that, as it developed, gradually interfered with and ultimately took the place of spherulitic growth.

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DESCRIPTIONS OF PLATES.

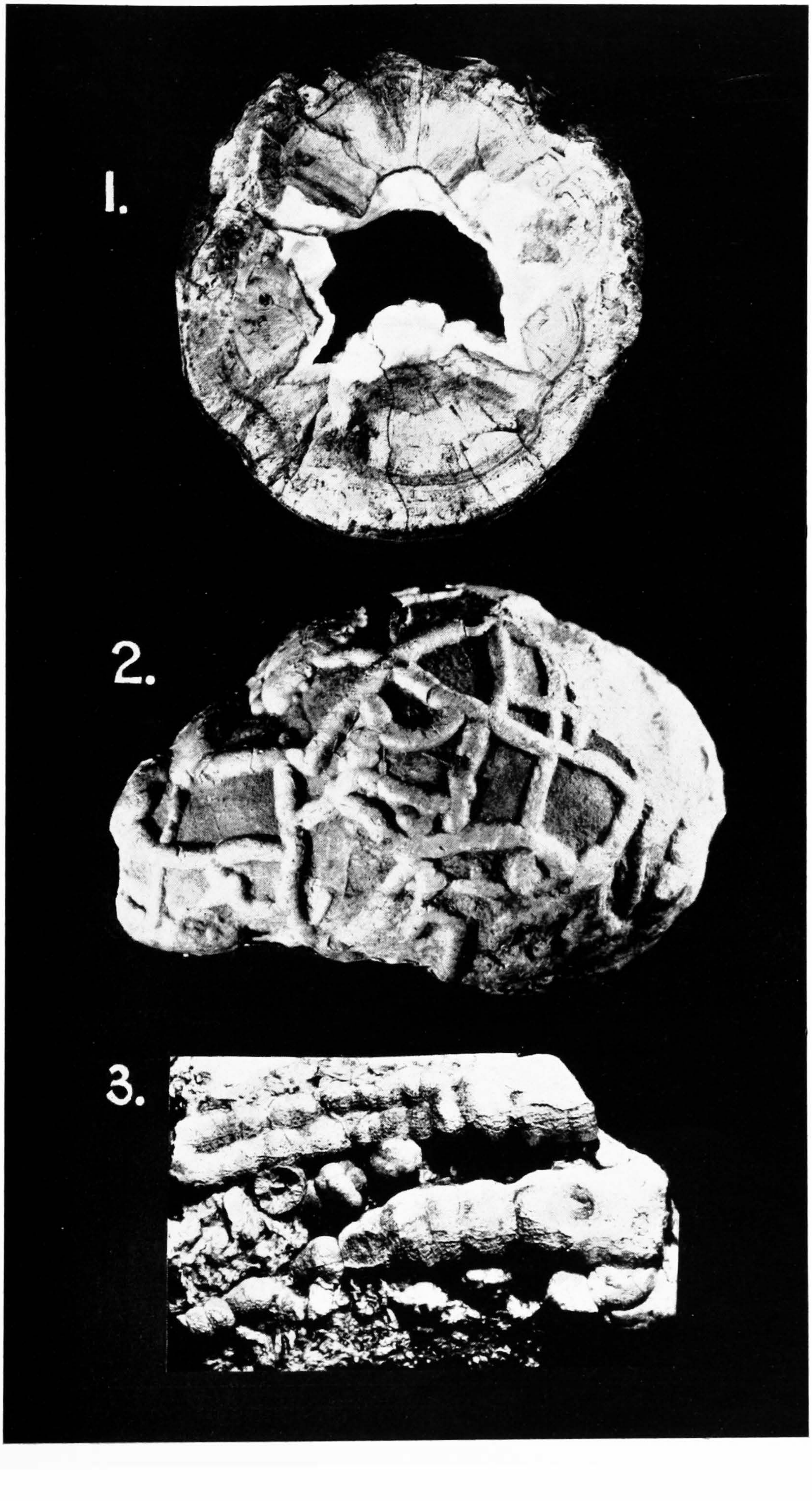
Plate III.

All specimens were collected from the northern end of Tamborine Mountain, south-eastern Queensland. Photographs by E. V. Robinson.

Figure 1.—Individual spherulite, showing combination of radial and concentric structures. Natural size.

Figure 2.—Secondary axiolitic growths upon a spherulite. Natural size.

Figure 3.—Composite axiolites. Natural size.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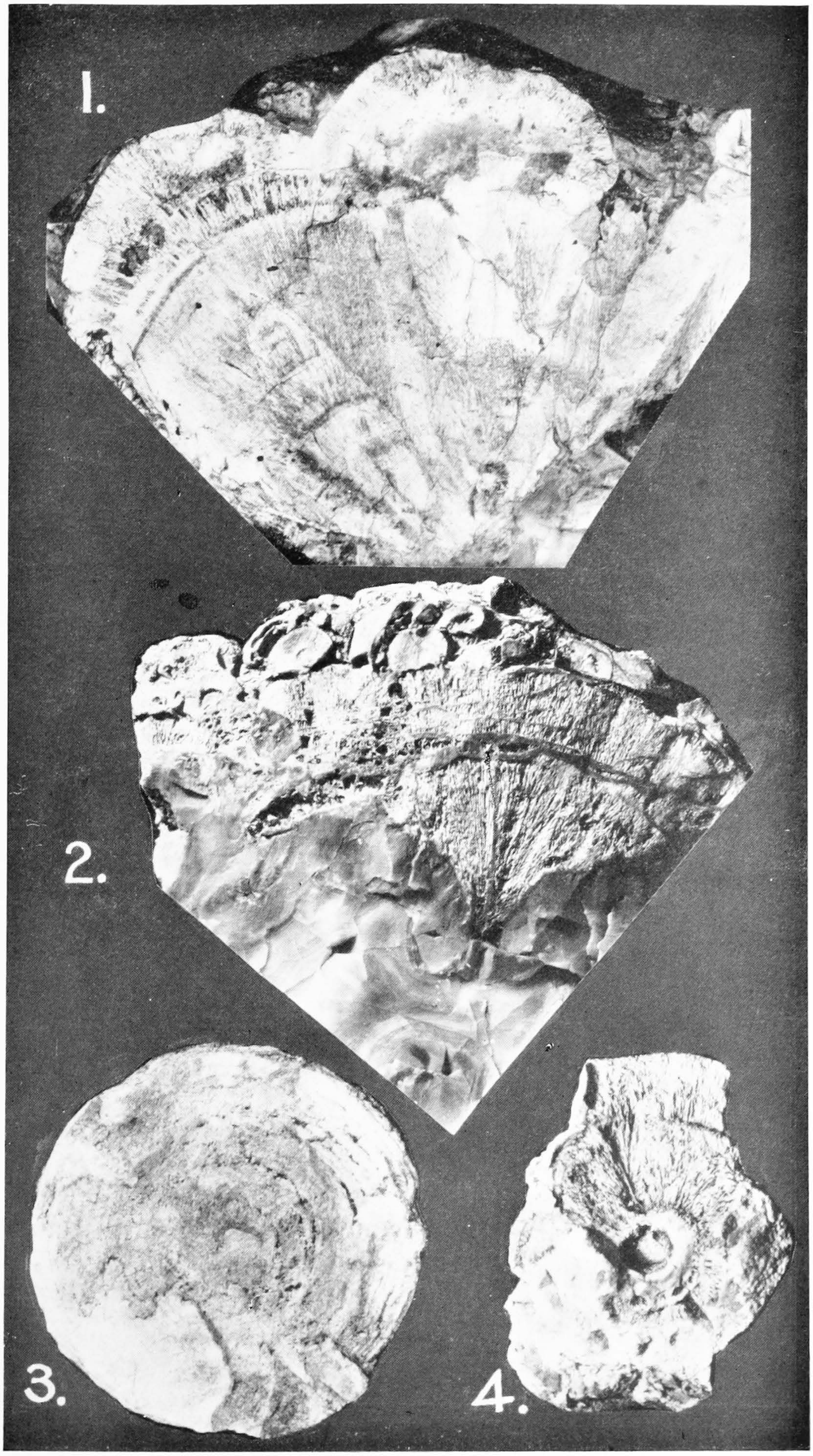
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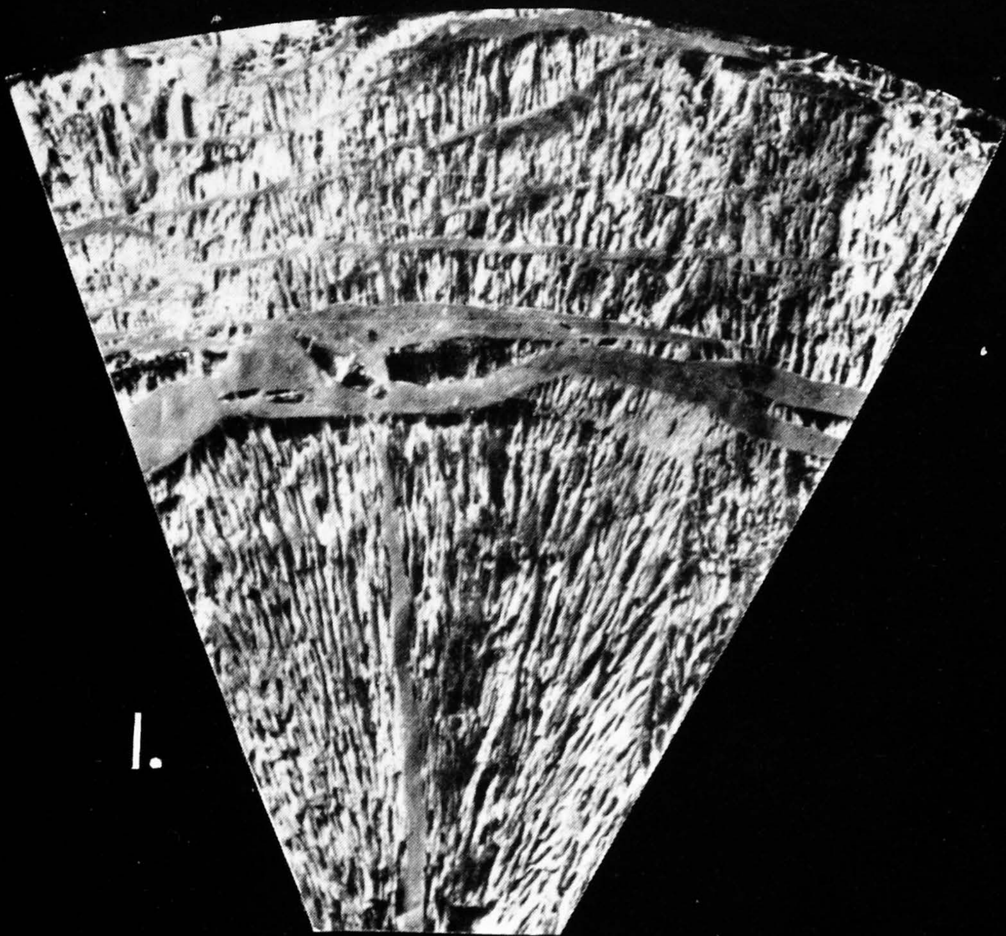

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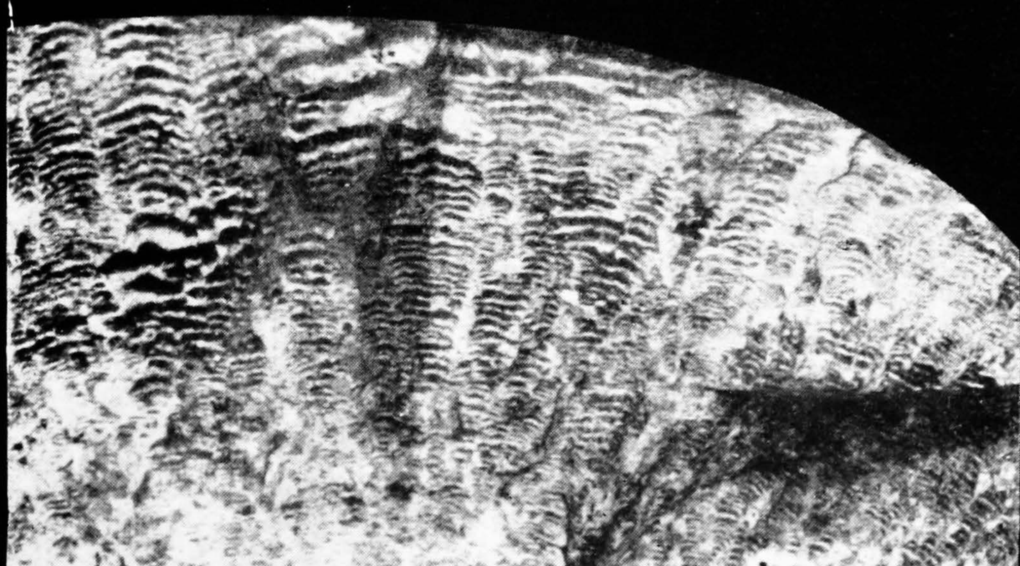

2.

PLATE IV.

Figure 1.—Portion of a composite spherulite, showing radial and concentric structures. Natural size.

Figure 2.—Segment of a disrupted spherulite surmounted by secondary spherulites. Natural size.

Figure 3.—Spherulite, showing concentric structures of a rhythmic nature. Twice natural size.

Figure 4.—Partial spherulite, showing combination of radial and concentric structures. Somewhat enlarged.

PLATE V.

Figure 1.—Part of a composite spherulite, showing radially arranged spherulitic tissue. About three times natural size.

Figure 2.—Part of a spherulite, showing concentrically arranged spherulitic tissue. About three times natural size.
