## The spherulitic rhyolites of Jersey.

## (With Plate XI.)

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[Read March 15, 1932.]

THE spherulitic rhyolites of Jersey show a development probably unequalled in the British Isles. They are particularly notable for the large size of the spherulites and for the variety of structures which they present. The object of the present paper is to describe some of these structures and to discuss their origin.

The first worker to publish a description of spherulitic rhyolites from Jersey was Davies (2).<sup>1</sup> Their field relations were described by Noury (4), while De Lapparent has discussed their petrology (5), (8) and the vexed question of their age (7). Parkinson (10), (12), (14) has given admirable and detailed accounts of the Bouley Bay rocks. A recent detailed survey of the Jersey volcanic rocks by the writer, details of which will be published elsewhere, shows the following sequence and approximate thicknesses :

3.	Non-porphyritic rhyolite	•••	1200	metres.
2.	Porphyritic rhyolite	•••	900	"
1.	Andesite	•••	600	,,

Spherulites are mostly confined to the non-porphyritic series and they are best developed near the top of it.

The non-porphyritic rhyolite is a rock of felsitic texture; its colour is purplish-chocolate, or, less commonly, pale yellowish-green. It contains extremely rare and small phenocrysts of quartz and microperthite. An almost constant feature of the rock is the presence of strongly marked flow-banding, chiefly shown in the hand-specimen by alternate bands of different colours, evidently due to differences in the state of oxidation and perhaps in the abundance of iron present. The bands are usually about one millimetre thick.

Under the microscope the non-porphyritic rhyolite is seen to consist of finely granular orthoclase enclosed poikilitically by a patchy quartz

<sup>1</sup> Numbers in parentheses refer to the bibliography at the end of the paper.

mosaic (pl. XI, fig. 6). Hyndman and Bonney (9) have shown that the Bouley Bay rocks are soda-rhyolites, and it is thus evident that the orthoclase, here and probably throughout the series, is of a sodic variety.

The chief features which distinguish adjacent flow-bands in thin section are the relative abundance of sericite, a little of which is present in all the rhyolites, and the differing grain-size of the quartz Along many of the bands elongated masses of quartz and felspar. mosaic frequently occur, up to 1 mm. in thickness, evidently of late crystallization. The walls bounding these masses are usually lined with a fringe of earlier orthoclase or perthite. All these features are attributable to original irregular distribution of water. Variations in the state of the iron are more evident in the hand-specimen than in thin section. In a discussion of flow-banding Fuller (19) has suggested that the requisite differences in the condition of the iron might be produced by vapours penetrating a flow-breccia. Remelting and further flow would give the vari-coloured streaks. Rocks arrested in various stages of this process almost certainly occur in Jersey. This theory would, moreover, account for the differences in watercontent.

The Spherulitic Rhyolites.—Small simple spherulites are common throughout the non-porphyritic rhyolite, but large spherulites and complex spherulitic growths are only known around Bouley Bay and in one small rock in St. Catherine's Bay.

The spherulites of Les Hurêts, overlooking the west side of Bouley Bay, have been described by Parkinson (10). In addition to simple spherulites up to about 1 cm. in diameter, there are found larger globular forms, only slightly complex, up to about 10 cm. Complex lenticular bodies, 5 to 20 cm. thick, merge into continuous bands with spherulitic structure and nodular outer surfaces. The total thickness of the spherulitic flow or flows at Les Hurêts is unknown, but a thickness of about 50 metres of lava is exposed without any obvious break. The different types and sizes of spherulites occur in separate bands parallel to the flow-banding, so that the rock looks in places like a bedded conglomerate.

Spherulites of several interesting types are found in a rock about 50 metres east of the south end of the long sea-wall of St. Catherine's Bay. Relatively simple bodies about 1 cm. in diameter occur locally, while complex lenticular spherulites, mostly from 5 to 20 cm. thick, are abundant throughout a thickness of about 10 metres of rhyolite.

The latter have in places coalesced into continuous sheets parallel to the flow-banding, similar to those at Les Hurêts. One such sheet was found to be over 30 cm. thick.

Petrology of the Spherulitic Rhyolites.—While studying the petrology of the spherulites the writer has been privileged to examine, in addition to material of his own collecting, a number of thin sections from Bouley Bay, kindly lent by Dr. Parkinson and mostly already described by him.

While superficially variable, the structure of the spherulites shows little fundamental variation. The primary spherulitic growth consists of radiating and more or less branching felspar fibres, often with a detectable negative elongation. Superimposed on these fibres is a patchy mosaic of quartz grains which enclose them poikilitically as the quartz does the granular felspar of the non-spherulitic varieties of rhyolite. The very small spherulites often consist of solid felspar but the larger ones show a complicated quartz mosaic, considerably influenced by the earlier felspar growth. The quartz grains are elongated in the radial direction, to which their optic axes are very roughly parallel. The quartz is, however, by no means as regularly arranged as the felspar.

Though the radial fibrous growth of the felspars is sometimes almost perfect, it is difficult to photograph or even to see in ordinary transmitted light. Pl. XI, fig. 3, shows the meeting of two spherulites in a specimen from Les Hurêts and is taken with oblique lighting from right and left: only those fibres which are nearly perpendicular to the plane of incidence are shown as illuminated. A few radial fibres of iron oxide<sup>1</sup> are seen as well as a quartz band (dark) representing a halt in the growth. Fig. 4 shows the same field as it appears under about the same magnification between crossed nicols. The polarization effects seen are almost entirely due to the patchy quartz. The parallelism of the quartz crystals is clearly far less perfect than that of the felspar, both as to external form and as to optical orientation. In this example the radial elongation of the quartz is much more pronounced than usual.

More nearly normal in the last respect are some small spherulites from St. Catherine's. In pl. XI, fig. 7, one of them is seen in ordinary

<sup>&</sup>lt;sup>1</sup> Fibres of iron oxide are often found between those of felspar and appear, at first sight, to belong to the primary spherulitic growth. They are, in fact, probably a late development as they sometimes terminate against veins of secondary quartz.

transmitted light. The spherulite shows at its centre a more or less continuous growth of felspar, probably originally radial, though the radial structure is now chiefly shown by iron oxide. Farther out the felspar is in concentric bands suggestive of rhythmic growth. The felspar is set in 'patchy' quartz which is present nearly everywhere but is best seen between the dense felspar bands. Even in these, however, some felspar is present. A devitrified glass surrounds the spherulites and contains perlitic cracks, formed after the spherulites. Some of the small circles of the perlite are in turn occupied by bodies suggestive of rudimentary spherulites. The main spherulites are red with iron oxide. The perlite is green and finely granular with patches of a greenish-yellow micaceous mineral and with quartz forming a sort of disperse mosaic. In fig. 8 the poikilitic structure of a spherulite in this rock is seen between crossed nicols.

The 'patchy' or poikilitic structure, seen both in spherulites and in normal non-porphyritic rhyolite, seems, as suggested by Bonney and Parkinson (14), to be a combination of primary and secondary crystallization. The felspar, at least in the spherulites, is primary, while the quartz is of later growth and probably secondary. The rejection of iron oxide, in some examples, along nearly all the sutures of the quartz 'patches', shows almost beyond doubt that the silica first crystallized as quartz, since where tridymite is known to have occurred in the rhyolite each crystal has formed several differently orientated quartz crystals. 'Patchy' quartz has, however, formed in the near neighbourhood of original tridymite (now quartz but recognizable by its external form outlined in iron oxide). The tridymite is itself of late growth, yet, from the temperature relations of these minerals, it must be older than the quartz or the 'patches'. The latter is thus of very late formation.

Lenticular Spherulites and Spherulitic Bands.—There is little but a formal distinction between the large lenticular spherulites and the spherulitic bands which are clearly formed by their coalescence. Parkinson (10) has claimed that, at Les Hurêts, crystallization in them began at the edges and spread inwards.

An examination of the rocks in the field strongly suggested to the writer that both lenses and bands crystallized outwards from internal centres, and the slice represented in figs. 3 and 4 was cut from one of the bands at Les Hurêts to discriminate between the two possibilities. The upper part of the figures lies nearest the outside of the band: it is clear, from the way in which the two sets of fibres meet, that recrystallization spread from the inside of the band. Dr. Parkinson has kindly lent the writer a thin section which at first sight seems to show inward growth. One part of the outer surface of the band is a concave curve with felspar fibres everywhere nearly perpendicular to it. Closer examination, however, shows that the fibres actually radiate from a large number of centres inside the band and meet in a similar manner to those of fig. 3.



F1G. 1.



FIG. 1. Natural section across a complex lenticular spherulite from St. Catherine's Bay, showing lines of growth etched out by weathering. a, nuclear bodies.

FIG. 2. Natural section across a complex spherulite from St. Catherine's Bay. a, b, compact lenticular spherulites, b showing rhythmic structure. The quartz crescents are shown dark, as they appear in the hand-specimen. c, partial spherulites forming the outer part of a large complex body. d, e, flow-banding.

In the lenses and bands of St. Catherine's Bay the lines of growth have been weathered out in a rather striking manner. The large lenses originate either in a compact lenticular growth, occasionally rhythmic, or in an irregular but roughly spherical body suggestive of Parkinson's central bodies (10). Arranged as though piled up in all directions around this centre is a cluster of crescentic partial spherulites. Such an arrangement is shown in text-fig. 1 which represents the lines of growth weathered out on an exposed surface. The outside of each crescent is part of a single sphere, while its inner surface is moulded on those of a number of its neighbours. The outer simple surface is clearly that of free growth.

The growth lines in the lenticular spherulites where they coalesce

into a band show that crystallization started almost simultaneously at a number of points in a flow-plane. The main spherulite b of textfig. 2 throws light on the origin of the central lenses. If we accept the little quartz crescents as growth-lines, while postponing consideration of the cause of their rhythmic arrangement, it is evident that crystallization spread outwards from any given point in the median plane except in so far as it was interrupted laterally by crystallization from other centres. Simultaneously a wave of nucleus formation was spreading in the plane.

It is not denied that crystallization in certain bodies can take place from without inwards but, as far as Jersey is concerned, the writer has only found it in fragments in breccias.

The median plane of a lenticular spherulite is often in a strongly marked flow-band of a pale colour, suggesting the presence of sericite, while secondary quartz is also visible at times and the iron is probably less highly oxidized than elsewhere. In accordance with the explanation adopted above, such bands are those that have been least acted upon in a flow-breccia and have thus retained their water and not been highly oxidized. This is in agreement with observations on a thin section of a loose pebble from St. Catherine's Bay, consisting of a very fine grained breccia remelted and partly drawn out. Those lenticular streaks which are most vesicular are also most highly spherulitic. Iddings (3) and Parkinson (12) have agreed that the production of spherulites is favoured by the presence of water.

Rhythmic Phenomena.—Comment has already been made on the rhythmic alternation of spherulitic felspar and 'patchy' quartz in the small spherulites at St. Catherine's. Parkinson (12) has described a further development of this structure at Les Hurêts in which the felspar zones alternate with bands of pure quartz, but much more striking examples of rhythmic structure occur in St. Catherine's Bay. The structure is not common but seems to appear anywhere at random in the spherulitic part of the flow and in almost any part of the large complex bodies, though generally in the more compact central lenticular spherulites.

The lenticular spherulites are sensibly symmetrical about flowplanes but the rhythmic structure is sometimes found only on one side of an otherwise symmetrical body. Cases of perfectly symmetrical rhythmic bodies do occur, and asymmetry is probably mainly an accidental result of the rarity of rhythmic structure. The asymmetry does, however, at times seem to be systematic, as in the case illustrated in text-fig. 2, which represents part of one face of a hand-specimen. The main spherulite is almost symmetrical except that rhythmic trains of alternating quartz and felspar arcs only arise from one side of the median flow-streak.

The structure of a rhythmic lenticular spherulite from St. Catherine's as seen in thin section is shown in pl. XI, fig. 9. It is very evenly developed and extends with something like regularity through a large part of a spherulite some 15 cm. thick. The cloudy granular areas in the figure represent felspar together with a little quartz and some finely divided iron oxide; the white areas are clear quartz. The felspar is arranged in parallel or slightly diverging piles of arcs, capping one another and separated by quartz. The arcs are about 0.4 mm. long and the distance from the inside of one to that of the next is about 0.14 mm. The concave side of each felspar arc is sharp, while on the outside it is ragged with needles running out into the This is a very constant feature where quartz bands quartz area. occur in spherulites, whether rhythmic or not, and shows that a given phase of felspar crystallization began sharply but died out gradually.

Iddings (3) has described and figured examples of rhythmic spherulites but none of his cases approach the best Jersey ones in delicacy. More nearly similar to the latter is a structure figured by Delesse (1, pl. 2, fig. 6) but dismissed in a few lines of description.

The arcs of felspar (cloudy in most of the figures) are usually separated by homogeneous quartz resembling vein-quartz. In a rhythmic spherulite from the cliffs north of Les Hurêts (fig. 5), however, we find that the larger spaces between the felspar arcs show late crystallization on the walls—first felspar, then tridymite, now seen as quartz outlined in iron oxide. Upon this grew a thin coating of quartz or more tridymite and the cavity was then filled with yellowish sericite, occasionally followed by more quartz.

This, allowing for the alteration of the Jersey examples, is approximately the order of crystallization in the cavities in the spherulites at Obsidian Cliff, Yellowstone National Park. In the latter case Iddings (3) has supposed that in the process of crystallization, rhythmic or otherwise, the water present in the lava first separated as vapour and later condensed and promoted the recrystallization of the confining walls.

The establishment of the Reaction Principle has, however, drawn attention to the frequency with which equilibrium is shifted with fall of temperature in a crystallizing magma so that recrystallization of earlier formed minerals takes place. Such a recrystallization seems to have taken place in the Jersey rock last described: it is unnecessary to assume, as Iddings does, that water was temporarily restrained from activity by removal to another phase—especially since cases are found where there is no clear quartz but merely increase and decrease in the proportion of felspar (fig. 7).

Of course the original lava did contain water, but it seems rather that when the primary felspar crystallized there was left a liquid consisting mainly of water and silica with perhaps a little felspathic material. Such a liquid would be much more mobile than the lava, while yet probably being miscible with and grading into it. On the other hand, it would readily produce the observed effects of crystallization in a cavity.

The Mechanism of Rhythmic Recrystallization.-The regularly repeated felspar arcs are similar to Liesegang rings and some similarity probably exists between their modes of formation, though there is no question of diffusion of a reagent producing double decomposition in the rhyolite. Liesegang rings are, in the typical case, regularly spaced concentric circles of silver chromate produced when a drop of silver nitrate solution is placed upon a film of moist gelatine containing a Wilhelm Ostwald (13) explained their production soluble chromate. by supposing that the precipitate did not crystallize out the moment the solution was saturated but when it reached the supersolubility point (metastabile Grenze). The whole neighbourhood was then depleted of chromate down to the normal solubility, with the result that the precipitant had to diffuse some distance beyond the first precipitate before it could again raise the concentration of the precipitate to the supersolubility point and produce another ring of the substance.

The supersolubility point is a well-defined phenomenon, thoroughly investigated by Miers and Isaac (15). Miers (16) has used it, on the same lines as Wilhelm Ostwald, to explain the rhythmic crystallization of potassium dichromate from aqueous solution. The later theories regarding rhythmic precipitation of Wolfgang Ostwald (18) and of Bradford (17) are more complicated and only apply to cases of double decomposition: they do not concern us here directly. Hedges (20), in a masterly survey of the whole subject, reaches a conclusion which embraces Wilhelm Ostwald's theory but is of more general application. He considers that 'the essential condition for periodicity appears to be the existence of some critical condition determining a change which proceeds to completeness once the critical value is reached'.

Several previous theories of the development of rhythmic spherulites demand consideration.

Cross (6) supposed that most of the structures seen in spherulites were the result of forms taken up by amorphous silica (opal). Most of the facts adduced in the present paper, however, point to felspar as the primary builder of structures.

Iddings (11) held that pulsating crystallization was due to 'the lowering of the saturation of the surrounding mother-liquor caused by the sudden liberation of heat in the act of crystallization and to the rapid extraction of crystallizing molecules'. This theory is practically the same as that of Wilhelm Ostwald and is, with important modifications, adopted by the writer.

Parkinson (12) holds that the latent heat, if any, of felspar is probably inadequate for the process suggested by Iddings. The crystallization of felspar from the lava on cooling, however, proves that its latent heat of solution in the lava is negative. However small it may be, conditions will almost certainly occur from time to time under which the liberation of heat on crystallization will more than balance, for the requisite short time, the loss of heat due to Parkinson himself seems to hold that rhythmic interrupcooling. tion of crystallization was produced mechanically by the accumulation of water-vapour set free simultaneously with the felspar. As already mentioned, there can have been no liberation of vapour in the small spherulites at St. Catherine's (fig. 7), and even where clear quartz occurs between the felspar arcs the sharp onset and gradual ending of crystallization are more suggestive of a physico-chemical mechanism on the lines suggested by Wilhelm Ostwald. The exact nature of such a mechanism must next be considered.

Simple spherulites might well grow rapidly from a highly supercooled lava without reference to the direction in which heat was being lost. Rhythmic growth, on the other hand, implies a continual oscillation at the growing front between the stable and labile states; so that loss of heat will closely control the process.

The rhythmic growth of a complex spherulite symmetrically about a plane would at first sight appear to demand the conduction of heat from both sides towards the plane and its removal or annihilation on arriving there. This is impossible, and we must therefore look for a subsidiary controlling influence. The dominance of the direction of the thermal gradient will be mitigated by slow cooling. Assuming that cooling was very slow, let us consider the history of a fairly large mass of lava of sensibly uniform temperature at the supersolubility limit for felspar.

Owing to some slight chemical peculiarity of a particular flowplane (a purely physical peculiarity would hardly be sufficiently persistent) spherulitic crystallization begins almost simultaneously from a number of points in it. Crystallization will continue until the concentration of felspar in the immediately surrounding liquid is reduced to the normal solubility value. This crystallization will have liberated a certain amount of latent heat, and before our mass of lava can again crystallize spontaneously this heat must be conducted away. Once interrupted, there now seems to be no reason why crystallization should continue about the former centres. We should rather expect it to start from points distributed almost at random throughout the mass; and this is probably what does happen in many cases.

Accepting current theories of rhythmic crystallization, we can hardly avoid the conclusion that symmetrical rhythmic spherulitic growth can only take place in the body of a lava, as we see it to have done, if each pulse of crystallization gives rise to an agent which promotes nucleus formation in the succeeding zone. If we search for such an agent we immediately note one peculiarity in the zone surrounding the spherulites already formed. Water, which is a highly mobile mineralizing agent, has been liberated, and will undoubtedly diffuse outwards. It may now be assumed that, while the rest of the mass can remain in the labile state with some chance of immunity from crystallization, the presence of water promotes nucleus formation in the zone surrounding the spherulites. If this be granted we have the requisite conditions for rhythmic growth.

The simultaneous production of major growth-lines in adjacent spherulites (text-fig. 2) agrees well with the almost perfect uniformity of temperature at a given time, assumed above. On the other hand, the conditions assumed clearly represent an ideal case, and if they could hold at all it is almost certain that the lava remote from crystals and liberated water sometimes fell not merely slightly but considerably below the supersaturation point without crystallizing. It is also probable that, even near the growing spherulite, nuclei sometimes failed to arise immediately the same point was reached. By adopting this further hypothesis, which allows the existence of slight local temperature gradients, we can account for most of the irregularities observed in actual rocks, such as the interpolation of a quartz band along part only of the growing front.

The asymmetrical growth shown in text-fig. 2 is perhaps due to the migration of water, or water and silica, along a temperature gradient, or perhaps to bands originally of different composition. It is not so easy to account for the solid masses of felspar along the junctions of the separate systems. In most cases of the meeting of systems of rhythmic crystallization from two centres the lines of junction are marked by clear spaces as demanded by the simple theory. It is possible that in the rhyolite the mobile silica solution segregated somewhat, after the felspar crystals were formed. This particular fact regarding the distribution is more readily explained by Parkinson's theory (p. 235).

In spite of these points the theory here put forward is, of those yet suggested, the one to which the objections have the least appearance of being fundamental.

I should like to express my thanks for valuable help and criticism to Prof. H. L. Bowman, Mr. R. Brooks, Dr. A. Harker, Mr. W. E. Mourant, Prof. W. J. Sollas, Mr. R. C. Spiller, Dr. H. H. Thomas, and Dr. A. K. Wells, also to Dr. J. Parkinson for the loan of thin sections.

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## EXPLANATION OF PLATE XI.

Thin sections of spherulitic rhyolites from Jersey.

Fig. 3. The meeting of two simple spherulites in a spherulitic band at Les Hurêts. Oblique lighting.  $\times 23$ .

FIG. 4. The same field as fig. 3. Crossed nicols.  $\times 26$ .

FIG. 5. Crystallization in the spaces between the felspar arcs of a rhythmic spherulite from the sea-cliff north of Les Hurêts. Ordinary light.  $\times 23$ .

FIG. 6. Poikilitic structure in non-porphyritic rhyolite, Le Quesne's Quarry, La Crête, St. Martin. Crossed nicols.  $\times 69$ .

FIG. 7. Small rhythmic spherulite in perlitic rhyolite, St. Catherine's Bay. Ordinary light.  $\times 23$ .

F10. 8. Small spherulite, St. Catherine's Bay (same slide as fig. 7). Crossed nicols.  $\times 67.5$ .

Frg. 9. Complex rhythmic spherulite, St. Catherine's Bay. The grey areas represent felspar with some quartz and iron oxide. The white areas are quartz. Ordinary light.  $\times 23$ .



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