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The origin of myrmekitic intergrowths and a comparison with rod-eutectics in metals

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SUMMARY. A review is given of the metallurgical literature relating to the formation of metal rod-eutectics, which are texturally similar to myrmekite. Since no satisfactory explanation for rod-eutectics has been advanced, a comparison is made with myrmekite. It is proposed that this intergrowth develops as a result of the constriction of quartz during its recrystallization and inclusion in an expanding blastic growth of plagioclase, and the forces involved lead to the prolongation along the growth directions of quartz in the form of rods with a relatively small surface area (i.e. with circular cross-sections). Possible forces that could produce the same geometry in metal intergrowths are those that result from the relative contractions of the two components during freezing; in all cases for which quantitative data are known, the metallic rods have a greater contraction on freezing than the host substance. Examples of myrmekite and myrmekite-like intergrowths in the Constant Gneiss are described in relation to their particular origin. Two factors, the lack of proportionality of quartz to feldspar and the intimate association of myrmekite with myrmekite-like intergrowths, support the proposed mechanism of constriction of pre-existing quartz.

ONE feature of myrmekite that requires explanation is the vermicular or rod-like form of the quartz. A possible mechanism is the constriction of pre-existing quartz during its recrystallization and inclusion in an expanding blastic growth of plagioclase (Shelley, 1964). This hypothesis for myrmekite formation does not require any particular volume relationship between the quartz and plagioclase, and the hypothesis has also been extended to intergrowths of other minerals (Shelley, 1967). An earlier hypothesis (Schwantke, 1909), which proposes that myrmekite is formed during the simultaneous exsolution of plagioclase and quartz from alkali-feldspar, has been revived recently by Hubbard (1966) and Phillips and Ransom (1968). These workers do not offer an explanation of the vermicular form of the intergrowth and require a particular volume relationship between plagioclase and quartz, a relationship that has been disputed (Shelley, 1969). There is no doubt that similarly textured intergrowths are produced during eutectic crystallization in some metal systems, and the possibility of forming rod eutectoids (such as suggested by Schwantke) in the same way as rod eutectics in metals is worthy of examination.

Rod eutectics in metals

Lamellar eutectics are considered to be the normal structural type, but under certain conditions the lamellar material crystallizes as rods instead. The systems that have been discussed in recent metallurgical literature are listed in table I. Metallic colony structures, which are very similar to the typical plugs of myrmekite and in which rods are commonly formed, are produced under conditions of unidirectional growth (the term 'unidirectional' is in some ways misleading since the colony structures represent radiating growth directions on a small scale). Some of the more prevalent views on the origin of the metallic rods are summarized below.

TABLE I

Eutectic composition and texture			Contraction on freezing*	Eutectic composition and texture			Contraction on freezing*
Zn	host	70 % vol.	4·7	Cd	host	83 % vol.	4·0
Al	rods	30 % vol.	6·5	Zn	rods	17 % vol.	4·7
Sn	host	75 % vol.	2·3	Sn	host	91 % vol.	2·3
Cd	rods	25 % vol.	4·0	Zn	rods	9 % vol.	4·7
Al	host	45 % vol.	6·5	Sn	host	99·5 % wt.	2·3
Al_2Cu	rods	55 % vol.	? 92/8 Al/Cu = 8·7†	Al	rods	0·5 % wt.	6·5
Pb	host	81 % vol.	3·5	Al	host	92 % vol.	6·5
Cd	rods	19 % vol.	4·0	Al_3Ni	rods	8 % vol.	?

* 100 (vol. liquid—vol. solid)/vol. solid; data from Smithells (1967, p. 224).
† Data from Murphy (1954).

Chadwick (1963) considered impurities in the eutectic system to be essential to rod formation. Hunt (1966) showed, however, that the impurities have only an indirect effect on rod formation. Hunt and Chilton (1963), using very pure metals, showed that a lamellar intergrowth was changed into a rod intergrowth by diversifying the growth directions away from the plane of the stable lamellae.

It has been shown that a lamellar structure with a certain interlamellar spacing can reduce the interphase boundary area by forming rods with the same spacing, if the volume of the rods is less than approximately one-third of the total (Hunt and Chilton, 1963; Cooksey *et al.*, 1964). Considerations of the relative heat flow and solute distribution also favour the radial distribution of the rod eutectics (e.g. Cooksey *et al.*, 1967). If the lamellar plane itself has a particularly low energy then rods are not favoured unless their volume fraction is considerably less. Hunt and Chilton (1963) therefore suggest that even when the volume fraction is less than one-third, lamellae will be produced by growth on low energy planes, and the growth directions must be diversified in order to form rods. Although these factors are clearly important, they are not sufficient by themselves since, in the Al–Al_2Cu eutectic for example, rods are formed that make 55 % of the total volume.

The textures of the eutectics have been studied in connection with different freezing rates and temperature gradients. As a result of crystallization experiments, Cooksey *et al.* (1967), suggest that rod eutectics are characteristic of relatively quick freezing rates, while progressively slower rates produce lamellar eutectics that finally become degenerate or irregularly textured.

Jackson and Hunt (1965) and Hunt and Jackson (1966) relate types of eutectic structure to the relative entropies of melting of the two substances: when both entropies are low, lamellar or rod-like eutectics are formed; if one entropy is low and the other high then irregularly textured eutectics are formed; if both entropies are high the two substances grow separately without any intimate intergrowth.

From the above summary it is clear that a number of factors are important in determining the type of texture in an intergrowth, but the exact reasons for rod formation have never been completely elucidated. Therefore a common factor was looked for in both metal crystallization and subsolidus myrmekite growth. The essential principle behind the proposed mechanism of formation of quartz rods in myrmekite (Shelley, 1964) is that the constricted material takes on a form with a relatively small surface area. Quartz spheres would have the smallest surface area; however, during continuous growth of myrmekite, the quartz is prolonged as rods along the growth directions, and the relatively small surface area is reflected in their circular cross-sections. A force that could have the same effect in metal eutectic crystallization is that of differential contraction on freezing of the two components. An analogy may be drawn here with the surface tension effects that cause a bubble or drop of liquid to be spherical; this is a result of relative contraction as may readily be seen when a bubble bursts. The known values for percentage contraction on freezing in the well-known simple or nearly simple eutectic systems are listed in table I, and it is remarkable that the six pairs for which contraction values are known all have a greater contraction in the rods. It is therefore proposed that during active simultaneous growth of two substances in a colony structure (cf. Shelley, 1967, fig. 6*a*), the substance with the greater contraction on freezing will form a continuous rod with a circular cross-section.

It is admittedly difficult to visualize how the amount of contraction on freezing takes its effect, but since it is simply a relative thing, it must be due to mutual interference during the earliest stages of crystallization. It is interesting, therefore, that Jackson and Hunt (1965) show that rod intergrowths occur in substances with low entropies of melting, and that these substances have a diffuse boundary between the liquid and solid over which the molecules still have a relatively random orientation. Presumably it is in these initial phases of crystallization, where solid phases are not clearly established, that mutual interference effects can take place. This mutual interference will be most marked at relatively high growth rates since at low growth rates the forces involved would be dissipated before they could be effective in shaping the texture of the eutectic.

Myrmekite and myrmekite-like intergrowths from the Constant Gneiss

The Constant Gneiss outcrops near Westport on the west coast of the South Island of New Zealand. The structure and geology of the Gneiss is not well known; until recently it was thought to be pre-Cambrian (Bowen, 1964) but as a result of recent isotopic work (Aronson, 1968) it is now thought to be in part Devonian, in part Cretaceous. The following examples of myrmekite are taken from outcrops of Gneiss around Charleston, south-west of Westport. The structure of this particular area of

Gneiss is described in a separate paper (Shelley, 1970); in that paper, examples are given of myrmekite showing more than one stage of development, and the relevance of these stages to the general geology is discussed. The examples that are described here illustrate characteristics of the intergrowths that are particularly relevant to their origin.

Quartz: plagioclase ratio in myrmekite. According to Schwantke's hypothesis (1909), there should be a definite ratio of quartz to plagioclase depending on the composition of the plagioclase. Quantitative data supporting this hypothesis has only recently been produced by Hubbard (1966) and Phillips and Ransom (1968). This data was discussed subsequently (Shelley, 1969; Phillips and Ransom, 1969) in the light of examples of myrmekites from the Constant Gneiss showing ratios of quartz to plagioclase that do not substantiate the hypothesis (another example is given in Shelley, 1970). There is little purpose in labouring this point further here except to summarize: the problems involved in measuring the quartz/feldspar ratio in typical fine myrmekites are very great; there are frequent examples in which obvious inconsistencies in the ratio occur, but Phillips and Ransom do not share the opinion that these are significant. One particular example of myrmekite is, however, significant in understanding the mechanism of its growth (fig. 1). The typical plug of plagioclase, convex towards a perthitic K-feldspar, contains vermicules of quartz that halt at an inclusion of muscovite. If the quartz and plagioclase were both derived by exsolution from the K-feldspar then myrmekite would be expected on either side of the muscovite; in the experiments of Hunt and Chilton (1963), the simultaneous growth of the two metals continued quite regularly after passing an inserted barrier. It is suggested that the more probable explanation is that intergranular quartz was incorporated in the growing plagioclase but could not be carried past the muscovite barrier (fig. 2).

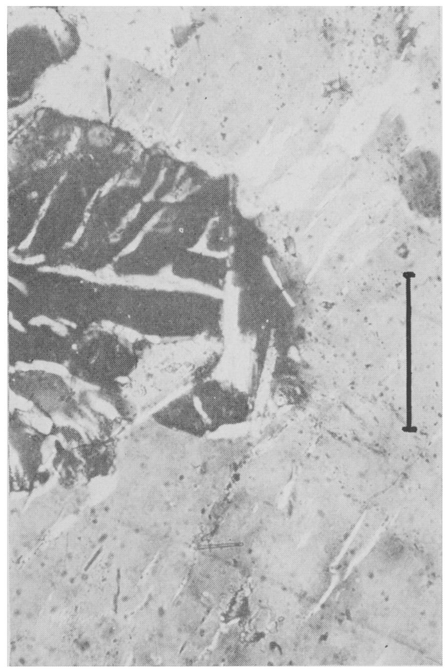


FIG. 1. Explanation in text. Scale mark represents 0·1 mm.

Association of myrmekite with muscovite–vermicular-quartz intergrowths. The fact that myrmekite is frequently associated with myrmekite-like intergrowths (i.e. intergrowths of similar texture to myrmekite but made up of different mineral pairs) has not received much attention in the literature. Sederholm (1916) described several mineral pairs in intergrowth; myrmekite and associated similar intergrowths have been described from the Lewisian Gneiss of Scotland (Shelley, 1967).

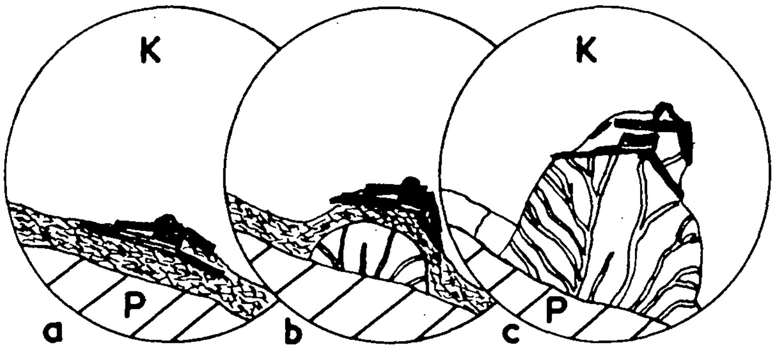


FIG. 2. Diagrammatic representation of growth of myrmekite shown in fig. 1. K — K-feldspar. P – plagioclase seed. (*a*) illustrates a strained (not necessarily cataclastic) groundmass of quartz with mica between K-feldspar and plagioclase. (*b*) plagioclase exsolved from K-feldspar grows on older plagioclase crystal and includes recrystallizing quartz as rods. (*c*) expanded intergranular is filled. Quartz was not carried past the muscovite barrier. The plug form minimizes the boundary area between K-feldspar and plagioclase.

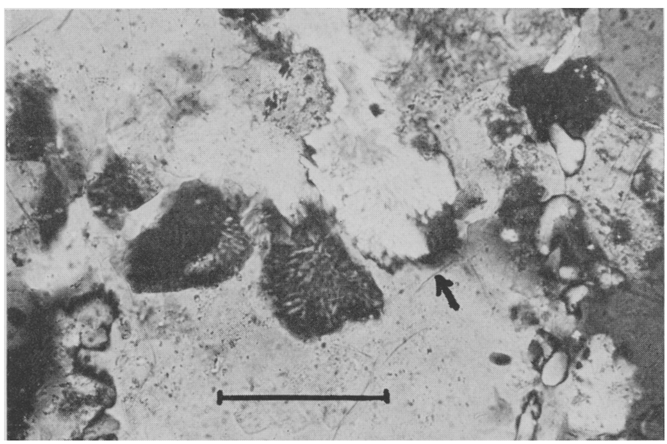


FIG. 3. The arrow indicates a muscovite-vermicular-quartz intergrowth. The associated similar intergrowths are myrmekites. Scale mark represents 0·1 mm.

Fig. 3 illustrates a muscovite–vermicular-quartz intergrowth associated with ordinary myrmekite from the Constant Gneiss. The two types of intergrowth are very similar, and it was at first thought that the muscovite had replaced the plagioclase of myrmekite. However, no general evidence has been found for this replacement and, further, it has been established that the geometries of the two intergrowths in three dimensions are quite different. The quartz vermicules in the myrmekite have shapes that vary in three dimensions (essentially a colony-type structure) whereas those in the muscovite vary in direction only two-dimensionally in the basal cleavage. This was established with the universal stage for the example in fig. 3; a typical side view of a similar intergrowth is shown in fig. 4*a* and a near perfect basal section in fig. 4*b*.

The very close association of similar vermicular intergrowths with differing mineral pairs, such as is illustrated in fig. 3 and was described previously from the Lewisian Gneiss, indicates their similar origin and the action of similar growth mechanisms. Schwantke's hypothesis requires a very particular origin for myrmekite that cannot be generalized to include the myrmekite-like intergrowths; further, no similar eutectoid crystallization systems are known that can be invoked to explain them. It is, however, as feasible to include a recrystallizing ground-mass of quartz by blastic

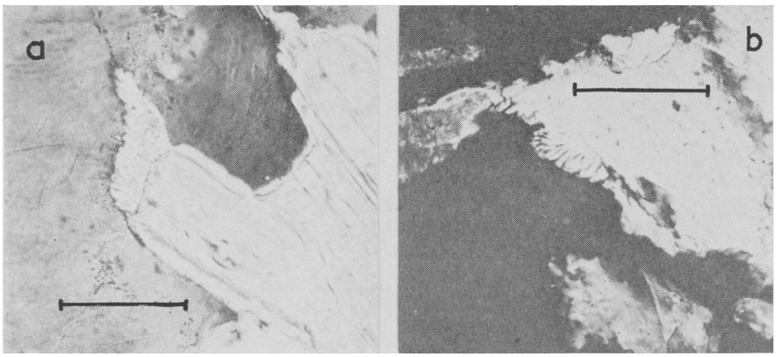
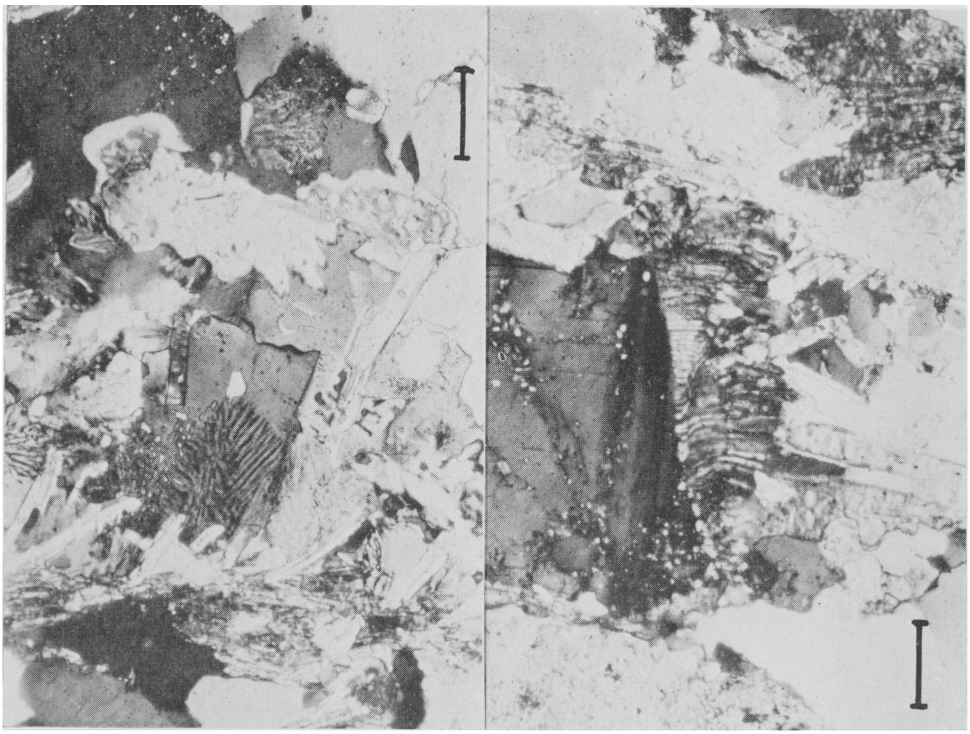


FIG. 4. Muscovite–vermicular-quartz intergrowths. Scale mark represents 0·1 mm.

muscovite as to include it by plagioclase. A common and convenient source of the plagioclase is from K-feldspar by exsolution, but this is not necessary to the basic mechanism (plagioclase possibly not derived by exsolution is described below). The vermicular intergrowths of the Constant and Lewisian Gneisses are conceived to be essentially similar to the poikiloblastic textures of metamorphic rocks, except that when growth is relatively fast, and the included mineral grains are simultaneously recrystallizing, these latter will be constricted to take on forms controlled by the growth of the host.

Myrmekites not associated with K-feldspar. Most hypotheses of myrmekite formation associate the plagioclase in some way with K-feldspar. It is an essential part of Schwantke's hypothesis that the plagioclase is exsolved from K-feldspar. Certainly, myrmekites are usually associated with K-feldspar, and Hubbard (1966), for example, has presented good textural evidence that some myrmekites are directly connected with exsolved perthitic plagioclase. In the Constant Gneiss, however, there are areas of plagioclase–quartz–biotite–muscovite gneiss that contain myrmekites with no associated K-feldspar. The Gneiss has suffered a complex history of plutonism and flattening and it is conceivable that K-feldspar was present but has been removed in some way. The myrmekites, however, do not have all the characteristics of the typical plug-like myrmekites that are elsewhere associated with K-feldspar. Although regular colony-type structures do occur (fig. 5), the vermicules in these myrmekites tend to be either more irregularly arranged and in apparently random patterns, or extremely regularly arranged in closely packed, long, and narrow vermicules (fig. 6; note how the area of unzoned myrmekitic plagioclase truncates well-developed oscillatory zones in the non-myrmekitic plagioclase). Individual plagioclase grains frequently have patchily developed concentrations of quartz vermicules. The myrmekites are typically clustered together in irregular mosaics with muscovite grains. Some plagioclase grains



FIGS. 5 and 6. Fig. 5 (left). Myrmekites not associated with K-feldspar. Note the colony structure and the irregular distribution of quartz in the plagioclase grains. The myrmekites are associated with muscovite flakes. Scale mark represents 0·1 mm. Fig. 6 (right). Myrmekite not associated with K-feldspar. Scale mark represents 0·1 mm.

in the clusters apparently contain no quartz at all. In others it can be seen that the plagioclase crystallization continued while there was an abrupt cessation of quartz crystallization (fig. 5). Fig. 5 shows in general a highly irregular distribution of quartz in plagioclase. The plagioclase varies slightly in composition over the border of albite and sodic oligoclase (the irregularities in the quartz distribution do not correlate with changes in the calcium content of the plagioclase). Muscovite–vermicular-quartz intergrowths are associated with these myrmekites.

Schwantke's hypothesis appears inapplicable to these examples, and it is not easy to explain the irregular distribution of quartz and albite in terms of a simple eutectic crystallization of the two minerals.

In summary, although it is probable that the blastic plagioclase of myrmekite frequently originates in K-feldspars, this does not seem to be always the case, and it is not essential to the more general hypothesis of myrmekite formation in which it is proposed that growing porphyroblasts of plagioclase include a recrystallizing groundmass of strained quartz. The hypothesis can be readily extended to other vermicular intergrowths.

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