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Myrmekites of exsolution and replacement origins—a discussion

SIR,—In his study of myrmekite Dr Ashworth (1972) has presented an excellent account of his ideas on the genesis of this intergrowth, and his paper will undoubtedly rank as one of the best on this topic. An important part of his work covers material similar to that described by Phillips, Ransom & Vernon (1972) dealing with an association be-tween myrmekite, muscovite and relict potash feldspar in retrograde metamorphic rocks from Broken Hill, New South Wales. Because both papers were published at the same time, without any cross reference, I would like to extend our discussion here to include some of Ashworth's original presentation. In addition, I would like to take this oppor-tunity to comment on some other aspects of Ashworth's paper.
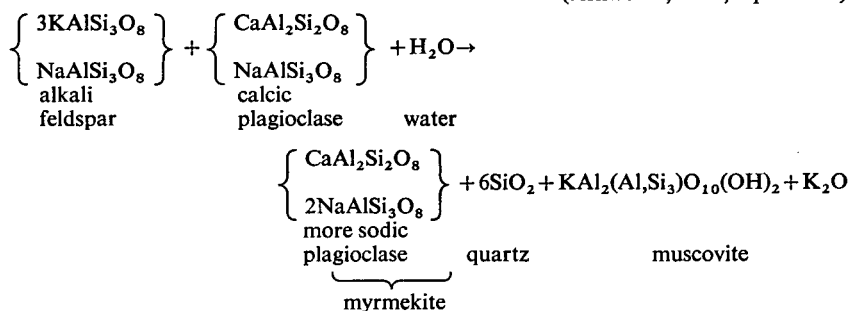
Ashworth said that myrmekite in the migmatites of the Huntly–Portsoy area had two distinct modes of occurrence (his types I and II). The type I (or 'normal') myrme-kites are attributable to exsolution-dominated processes and formed ' . . . as tempera-tures declined from their climactic values during regional metamorphism of the rocks'. The type II myrmekites do not result from simple exsolution but depend also on an essential metasomatic contribution, and ' . . . reaction occurred early in the cooling history of the rock'. He appeared to suggest that both forms develop under retrograde metamorphic conditions and he seemed to find no significant time difference between the formation of each of his myrmekite types. Work on the Broken Hill gneisses has suggested that perhaps three generations of myrmekite exist: one developed by metaso-matic replacement processes taking place during the approach of the culmination of prograde metamorphism (Phillips & Ransom, 1970), another possibly formed by exsolution effective shortly after the main prograde metamorphism (Ashworth's type I?), and the third (in which muscovite is associated with the myrmekite, Phillips *et al.*, 1972) related to a later retrograde metamorphic event (Ashworth's type II?). Shelley (1970, p. 677) also mentioned ' . . . myrmekite showing more than one stage of development . . .' so it is possible that these concepts may be applicable to the rocks studied by Ashworth.

Ashworth believed that his type II myrmekite, associated with a muscovite–quartz intergrowth, completely pseudomorphs potash feldspar. Phillips *et al.* (1972, Figs 3 & 4) noted similar occurrences, but we do not find any quartz intergrown with the muscovite. Further, after another examination of our slides, I find it difficult to define exactly the limits of any pseudomorph (as Ashworth does when he places a large muscovite grain across a pseudomorph boundary, his Fig. 1*b*), and I cannot be sure, because of the limits of the two-dimensional cut of a thin-section, that any given pseudomorph represents a completely replaced potash feldspar.
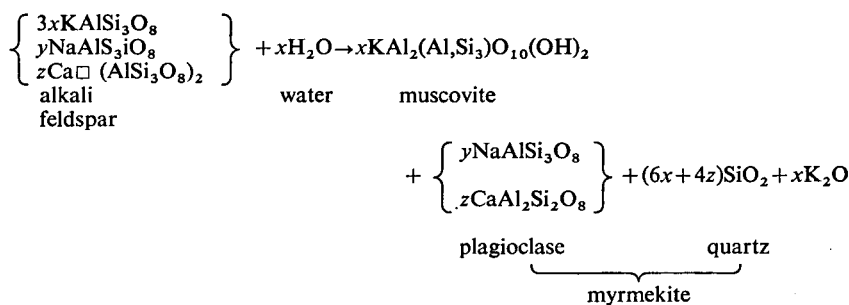
Considerable similarities exist between:

Potash feldspar + sillimanite + fluid →

muscovite + quartz + plagioclase
(Ashworth, 1972, equation 6)

$$\left\{\begin{matrix} 3KAlSi_3O_8 \\ NaAlSi_3O_8 \end{matrix}\right\} + \left\{\begin{matrix} CaAl_2Si_2O_8 \\ NaAlSi_3O_8 \end{matrix}\right\} + H_2O \rightarrow$$

alkali   calcic
feldspar   plagioclase   water

$$\left\{\begin{matrix} CaAl_2Si_2O_8 \\ 2NaAlSi_3O_8 \end{matrix}\right\} + 6SiO_2 + KAl_2(Al,Si_3)O_{10}(OH)_2 + K_2O$$

more sodic
plagioclase   quartz   muscovite

myrmekite

(Phillips *et al.*, 1972, equation 2)

and

$$\left\{\begin{matrix} 3xKAlSi_3O_8 \\ yNaAlS_3iO_8 \\ zCa\square(AlSi_3O_8)_2 \end{matrix}\right\} + xH_2O \rightarrow xKAl_2(Al,Si_3)O_{10}(OH)_2$$

alkali   water   muscovite
feldspar

$$+ \left\{\begin{matrix} yNaAlSi_3O_8 \\ zCaAl_2Si_2O_8 \end{matrix}\right\} + (6x+4z)SiO_2 + xK_2O$$

plagioclase   quartz

myrmekite

(Phillips *et al.*, 1972, equation 4).

All three equations lead essentially to an assemblage of muscovite, quartz and plagioclase. Apparently Ashworth believed that some of the quartz produced by such reactions becomes intergrown with muscovite as a muscovite–quartz symplectite, whereas we suggested that it supplements the quartz produced by the proportionality reaction (*e.g.* Barker, 1970; Becke, 1908; Schwantke, 1909; Phillips & Ransom, 1968), giving rise to myrmekite with 'excess' quartz. We have not proved that a disproportionate amount of quartz is present because of the fineness of the intergrowths and our lack of sophisticated equipment. However, I have examined many myrmekites and it appears to me that the quartz is unusually abundant. If Dr Ashworth tentatively accepts this possibility we may find some grounds for a compromise. The three equations listed above do not indicate where the quartz derived from the reactions is actually situated and I can envisage that it may go into three distinct positions within a pseudomorph: (i) discrete quartz grains, (ii) quartz intergrown with muscovite as is suggested by Ashworth, and (iii) 'excess' or supplementary quartz associated with plagioclase in a myrmekitic inter-growth as suggested by Phillips *et al.* (1972). The reasons for the quartz occurring at any one of these positions, rather than another, are not apparent to me at present, but it appears that the overall reaction is promoted by retrograde metamorphism, and that ultimately a fine-grained aggregate of quartz, muscovite and feldspar is produced. (Such a proposition may not be acceptable to Sharma (1969, p. 216) who believed that the presence of quartz vermicules in muscovite indicates that contiguous myrmekite forms by the replacement of plagioclase by potash feldspar, *cf.* Bhattacharyya, 1971.)

As a concluding thought on this particular matter I believe that some caution should

be exercised before proposing that potash feldspar alone is replaced by a muscovite–quartz–myrmekite pseudomorph. There may have been some plagioclase associated with the potash feldspar and equation 2 of Phillips *et al.* (quoted above) goes some of the the way in explaining the presence of more sodic plagioclase in the type II myrmekite of a pseudomorph, as compared with a more calcic 'primary' plagioclase found in the rest of the rock, *cf.* Ashworth, Table 1, rock/slide number 105785/3.

The thought-provoking equations devised by Ashworth show that at least four models may lead to a proportionality relationship between the plagioclase and the quartz of myrmekite. If these are taken as chemical alternatives, then it is difficult to give preference to any one, and as Ashworth himself said (in his interpretation of the origin of type I myrmekite), 'The data are insufficient to decide between Schwantke and non-Schwantke mechanisms'. Thus he did not reject the exsolution–vacancy hypothesis but believed that emphasis placed on it in the past may have obscured the other models. I have been an advocate of the Schwantke mechanism in 'granitic' rocks for the following reasons:

   (i) Rim albite, intergranular albite, and albite in perthitic arrangement may form by simple exsolution *without any metasomatic process.* As I have stressed elsewhere (Phillips, 1964, 1972) many of the common forms of myrmekite in granites are very similar in their spatial relationships to such albite and it seems logical to suppose that myrmekite has a similar origin to secondary albite. (I would also include here the quartz-bearing plagioclase found in some perthites.) If the formation of secondary albite need not require metasomatism, then why should such a process be invoked for myrmekite which is so closely involved with the albite?
  (ii) My support for the Schwantke model is also the result of a dissatisfaction with the hypothesis which proposes that vaguely defined sodic (or calcic) 'emanations' or solutions are able to penetrate to all feldspar grain boundaries in a rock mass and form albite rims *etc.*, yet cannot replace a sizeable part of any one feldspar crystal. Fortunately Tuttle's (1952) paper offers a more plausible explanation for the local origin of this albite.
 (iii) Further, there is always the question of whether the dominant K^+ and Na^+ ions in a high-temperature alkali feldspar would allow the very small amount of Ca^{++} to disturb the $AlSi_3O_8$ lattice and enter into a diadochic arrangement within the alumino-silicate framework.

Thus I came to favour the Schwantke mechanism which has no need for 'potential metasomatic contributions', 'a co-existing fluid as a sink for anorthite components', nor 'cationic metasomatism'. Perhaps the exsolution–vacancy hypothesis is an over-simplification but it has been supported elsewhere (with the added suggestions that sillimanite and nepheline may be exsolution products), *e.g.* Barth (1969, pp. v, 14, 28); Carstens (1967); Hubbard (1966, 1967a, 1967b, 1969); Kennan (1972); Perry (1968, p. 216); Sturt (1970) and Widenfalk (1969, 1972). Rejection of the need for metasomatism in myrmekite genesis reduces the four choices devised by Ashworth to one—the Schwantke hypothesis. Such a notion does not, however, rule out the possibility that myrmekite is polygenetic and Ashworth has lucidly proposed four possible models that demand serious consideration. This applies especially to metamorphic rocks in which local metasomatism over short distances may occur. I believe that no matter what the final outcome of this debate may be, the concept of the formation of myrmekite essentially by exsolution, for so long given scant mention in the literature before about 1964, has undoubtedly become the most viable hypothesis for the genesis of myrmekite.

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SIR,—I am very grateful for Dr Phillips' thoughtful attention to my paper (Ashworth, 1972). Comparison with the parallel work of Phillips *et al.* (1972) is indeed illuminating. There seem to be large areas of agreement, notably that

(1) myrmekite is polygenetic in that several processes producing the characteristic intergrowth can be distinguished, and

(2) in all cases under discussion, space now occupied by myrmekite was previously occupied by potash feldspar: the latter mineral, having formed at relatively high grade, became unstable with respect to either exsolution or other reactions at lower temperatures.

Phillips suggests that some parts of myrmekite-bearing pseudomorphs may have replaced plagioclase. It is not necessary to propose this in order to explain the trend to relatively sodic compositions, which is a natural consequence of formation under retrograde conditions by any of the proposed reactions (*cf.* Ashworth, 1972, pp. 58, 59). Where the plagioclase of a Type II myrmekite is sodic relative to its environment (*e.g.* 105785/3 of Ashworth, 1972, Table 1, as noted by Phillips), I would interpret this in terms of a potash-feldspar replacement reaction that was relatively late (low-temperature) as well as locally metasomatic (see below). Primary plagioclase survives in intimate contact with the pseudomorphs (*e.g.* Ashworth, 1972, Fig. 1*b*) and I have seen no evidence for its replacement by myrmekite. Bulk-rock reaction involving wholesale change of plagioclase composition (Phillips *et al.*, 1972, p. 574) evidently complicates matters in the Broken Hill area.

If one accepts that myrmekite forms from potash feldspar, it remains reasonable to propose that the characteristic features of myrmekite arise from some feature of the pre-existing mineral, such as its content of Schwantke component (Phillips) or its Al:Si ratio (Ashworth). It was the existence of myrmekites which cannot reasonably be attributed to exsolution that led me to propose a fundamental control which could operate in most or all of the polygenetic cases. I maintain that the obvious proposition is difficulty of bulk Al, Si concentration adjustment.

I regarded it as important to make the model as general as possible, catering in particular for hypothetical deviations from 'normal' quartz proportionality (Ashworth, 1972, p. 45), and it is now clear that my treatment is too narrow. While noting the possibility of quartz proportionalities less than the ideal, I did not admit the possibility that values *greater than* ideal could theoretically arise by similar reactions. Phillips *et al.* (1972) clearly correct this omission and tentatively record an actual occurrence. It now seems that alternative mechanisms could not be distinguished by deviant quartz proportionality as I had suggested (Ashworth, 1972, p. 46). Although the data remain sparse, it still seems reasonable to treat non-ideal proportionalities as special cases which can arise owing to the action of factors additional to the proposed fundamental control.

In my view, Phillips' most important points concern

(1) the nature of non-exsolution reactions contributing to myrmekite genesis, and
(2) the plausibility or otherwise of the 'non-Schwantke exsolution hypothesis'.

I shall deal with these in turn. Under (1), there is no conflict. It is clear that Ashworth (1972) observed a different reaction from that described by Phillips *et al.* (1972). The following points distinguish the latter case from the former:

(i) different time relations;
(ii) lack of quartz intergrown with the muscovite;
(iii) absence of Al_2SiO_5.

Phillips suggests that the concept of two or more distinct times of myrmekite formation (established beyond doubt by Phillips *et al.* in the Broken Hill area) might apply to the Huntly-Portsoy rocks, but I stand by the inference (Ashworth, 1972) that the two types there are coeval. There is no late metamorphic event to which Type II could be attributed even if the petrography suggested a time difference, which it does not. There are no systematic differences in morphology or composition: my Types I and II are modes of occurrence, not varieties of myrmekite. They need not be entirely synchronous, but the periods of formation must have at least overlapped substantially.

Whether the muscovite-myrmekite reaction of either area ever reached completion seems to me to be a minor point. Phillips *et al.* (1972, p. 574) write of 'almost total' replacement in advanced cases; this is true also in my rocks, with 'complete' replacement of potash feldspar in some of them. Despite obvious sampling limitations, it is

usual to estimate the amount of a mineral in a rock as being zero if thin sections of the rock do not contain any of the mineral.

I do not claim to be able to define the limits of any pseudomorph exactly. On the contrary, they can only be interpolated approximately, as Fig. 1*b* of Ashworth (1972) should serve to show. This does not invalidate the explanatory comment, concerning the approximate position of a boundary, in the caption of Fig. 1*b*.

The 'late' time-relations, and association with cataclastic deformation and sericitization (Phillips *et al.*, 1972, p. 576), suggest that the reaction of Broken Hill occurred at lower grade than that described by Ashworth (1972). Furthermore the primary assemblages are different. It is not surprising that the products are not identical. That myrmekite is formed in both cases supports the idea of some fundamental control. At least in the case studied by Ashworth, it seems that the amounts of quartz produced at the different locations mentioned by Phillips was controlled so that the myrmekite proportionality law was approximately obeyed. I am certainly willing to compromise by allowing roles for other factors. Even though excess silica apparently remained in the myrmekite volume when generated by the non-exsolution reactions whose postulation is necessitated by muscovite participation in the rocks described by Phillips *et al.* (1972), the question why myrmekite formed at all would still seem amenable to treatment in terms of constraints essentially corresponding to those invoked by Ashworth.

Phillips is justified in clarifying the point that Schwantke exsolution is the only hypothesis that is not metasomatic on the scale of a single potash feldspar grain (exsolution of pure albite is, of course, the end-member case $x^{M}_{An} = 0$ that is also non-metasomatic on the other model, and may theoretically occur after either hypothetical myrmekite exsolution process, or independently of both under different kinetic constraints). It is important that the size of the system, with reference to which metasomatism is to be discussed, be clearly stated (Ashworth, 1972, p. 53). Even Schwantke exsolution is obviously metasomatic on a slightly smaller scale, while the replacement-reaction processes need not demand transport distances more than one order of magnitude larger. At the risk of seeming pedantic, may I stress that the non-exsolution reactions contributing to myrmekite genesis in the rocks described by Phillips *et al.* (1972) and Ashworth (1972) *must* be metasomatic on the scale under discussion. The late reactions proposed at Broken Hill are explicitly so, and the fate of released K_2O is not certain (Phillips *et al.*, 1972, p. 576). The reactions are indeed similar to that proposed in the Huntly-Portsoy rocks, where, however, it is possible to discuss alkali movements more fully by reference to the probable role of nearby sillimanite (Ashworth, 1972, p. 59). 'Rejection of the need for metasomatism in myrmekite genesis' (Phillips) is, then, impossible in general. Let me reassure Phillips that I am not about to lapse into 'emanationism': vagueness of mechanistic discussion is deplorable, but inevitable at the present stage of research.

Having re-acquainted ourselves with small-scale metasomatism, we may proceed to point (2) mentioned above. The non-Schwantke exsolution hypothesis of Ashworth (1972) is undoubtedly the weakest and most poorly argued of the propositions under discussion. Phillips is right to clarify the point that the process is not a simple case of exsolution as normally understood, because it demands that some material be lost from the observed solid system. Perhaps it would be better described as a hypothetical process driven by the thermodynamic conditions that normally produce exsolution, but under kinetic constraints demanding metasomatism. The basic idea is that myrmekite may form because, as temperature falls, potash feldspar containing non-Schwantke plagioclase solute departs from equilibrium with a coexisting fluid: under the kinetic controls sketched by Ashworth (1972) for myrmekite formation in general, exsolution might be inhibited until the degree of disequilibrium was such that Ca, Na-poor potash feldspar + myrmekite + a fluid that has exchanged material in the required manner is a more stable configuration than Ca, Na-rich potash feldspar + fluid of a composition that was in equilibrium therewith at high grade. This end-member case with the fluid as a 'passive sink' seemed inherently unlikely, so I attempted to derive a more general interactive hypothesis. Necessity of small-scale metasomatism in the production of replace-

ment myrmekites renders it likely that allied processes would interact with exsolution, particularly where the two myrmekite types formed essentially simultaneously (Huntly-Portsoy area).

Perhaps the most serious weakness of a non-Schwantke exsolution-driven hypothesis arises from the fact that the pore-fluid in a metamorphic rock constitutes a very small proportion of the bulk system. The amount of interacting fluid required might be such that a large body of rock would be involved, and metasomatism could no longer be maintained to be 'small-scale'.

However, it is possible that a more bulky fluid may sometimes participate. I fear that my use of the word 'fluid' has caused confusion; I meant it as a neutral term to avoid specific nomination of either an aqueous fluid or a silicate melt. The possible presence of a relatively large amount of the latter during early myrmekite formation must be taken seriously in granitic and migmatitic rocks. It is likely that *in some cases* this fluid was available for an interactive reaction of the type discussed in Section 7 of Ashworth (1972).

In summary, a non-Schwantke exsolution-dominated hypothesis for myrmekite formation remains tenable, but requires a bulky and/or mobile fluid medium, which should be better defined before the hypothesis can be discussed adequately. The existence of myrmekites formed by several retrograde reactions under different conditions, in addition to those formed by exsolution or exsolution-dominated processes, reinforces the need to consider controls that could lead to the characteristic features of the intergrowth and are more generally applicable than the possible existence of Schwantke solid solution.

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Ordovician stratigraphy of Abereiddy Bay, Pembrokeshire

SIR,—The recent debate (Waltham, 1971; Black *et al.*, 1972) on the Ordovician stratigraphy of Abereiddy Bay, Pembrokeshire, with its relevance to the work of the late Professor A. H. Cox, lends interest to some of the modifications of his earlier views that Professor Cox intended to publish in the later years of his lifetime of research into the geology of Pembrokeshire.

Following periods of field work in the late nineteen forties Professor Cox wrote for the Geological Survey, in about 1950–51, the outline of a 'memoir' on the geology of the St David's area and, on pp. 142–5 of our copy of his Ms, he discussed the Ordovician sequence on the north side of Abereiddy Bay. Referring to the section in the old tramway cutting that enters the south side of Porth Gain Quarry Professor Cox wrote:

'The apparent sequence exposed along this cutting from north to south is: Llandeilo Flags with many trilobites, and black graptolitic shales; Castell Limestone—dark grey-blue limestone with partings of graptolitic black shale with trilobites and corals; Black shales—*Dicranograptus* Beds.

'Observed dips are consistently north at 50°–60°. When this section was first observed it was believed, in view of the persistently northern dip, that the sequence in the limestone was a normal sequence and that the Llandeilo Flags were introduced by a fault. More recently Dr Robertson, in the course of a visit, expressed his belief that the whole sequence was inverted, basing his opinion on the relations between bedding and cleavage, the cleavage dip being at a much smaller angle than the bedding dip. This conception of an inverted dip helped to a more harmonious reading of the sequence.