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Muscovite breakdown and corundum growth at anomalously low f_{H_2O}: a study of contact metamorphism and convective fluid movement around the Omey granite, Connemara, Ireland

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ABSTRACT. In the aureole of the late Caledonian Omey granite corundum develops in the Dalradian country rocks in a zone up to 250 m from the granite contact. The distribution of andalusite and K-feldspar in pelites and calcite + wollastonite + grossularite in marbles is consistent with inner-aureole metamorphic conditions of 615 ± 25 °C at 2.5 ± 0.25 kbar, and $X^{\text{fluid}}_{H_2O} \simeq 0.85$. Corundum develops from the reaction muscovite → corundum + K-feldspar + H_2O and first appears over 100 m further from the granite than the assemblage wollastonite + grossularite + anorthite. Experimentally determined equilibria can be satisfied only if $X^{\text{fluid}}_{H_2O}$ for the corundum-producing reaction was less than 0.6 and perhaps as low as 0.4. Corundum always grows within large muscovite crystals; f_{H_2O} within the crystal lattice is unrelated to that in the grain-boundary fluid of the surrounding rock.

Although whole-rock oxidation ratios are irregularly distributed within the aureole they are uniformly low in corundum-bearing rocks. Reducing conditions probably resulted from localized flow of H_2O-rich fluid away from the granite in a diffuse channelway that contains most of the corundum localities and also a distinctive skarn. Although corundum growth within muscovite is sealed off from the external water vapour conditions, it is suggested that movement of H_2O down a thermal gradient (and hence down an f_{H_2O} gradient at constant pressure) promotes the escape of $(OH)^-$ from the muscovite lattice and so allows the corundum reaction to proceed.

KEYWORDS: muscovite, corundum, contact metamorphism, Omey granite, Connemara, Ireland.

IN pelitic rocks, the growth of corundum and K-feldspar from the breakdown of muscovite is characteristic of very high-temperature contact metamorphism. In the Dalradian of Connemara, for example, corundum occurs in desilicated hornfelses adjacent to (or forming xenoliths within) basic and ultrabasic rocks intruded during the regional metamorphism (Leake and Skirrow, 1960; Evans, 1964; Evans and Leake, 1970). In this paper we discuss the origin of corundum in the thermal aureole of the Omey granite, a small high-level pluton intruded into Dalradian metasediments some 100 Ma after the regional metamorphism. The distribution of contact metamorphic minerals in pelites and interfolded calc-silicates suggests that corundum must have formed in a micro-environment in which f_{H_2O} was substantially less than in the surrounding rocks. The estimated temperature, 600-620 °C, is also substantially less than that normally anticipated for corundum growth from muscovite breakdown at around 2.5 kbar.

The Omey granite (fig. 1), a plug-like body about 6.5 km in diameter at the present level of erosion, is probably a satellite pluton associated with the Galway Batholith, both intrusions being dated around 415 Ma (Leake, 1978). It is intruded into Dalradian rocks, mostly pelitic to psammitic schists, that had been previously metamorphosed to staurolite-sillimanite grade during the Grampian orogeny (*c*.500 Ma). The metamorphic aureole was described briefly by Townend (1966) and Cobbing (1969). A fuller account of the aureole mineralogy, and the first description of corundum, was given by Ferguson and Harvey (1979) who also outlined the stratigraphy, structure, and regional metamorphism

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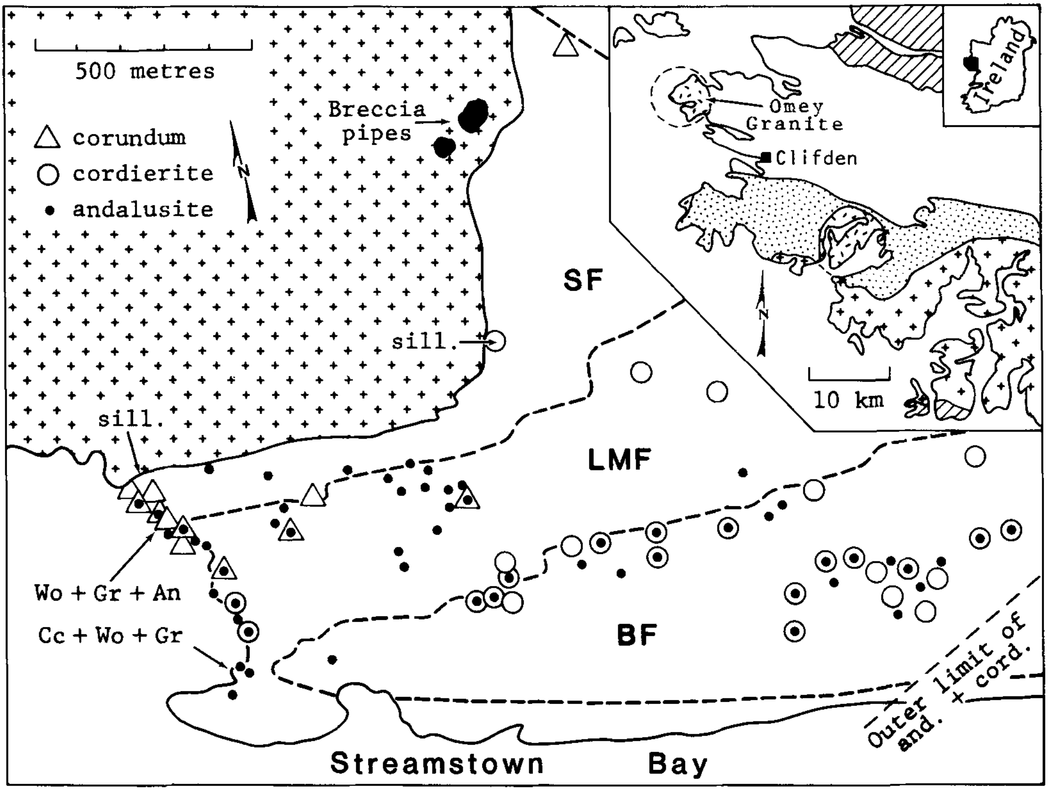

FIG. 1. Sketch map showing distribution of Streamstown Formation (SF), Lakes Marble Formation (LMF), and Ballynakill Formation (BF) within the best exposed part of the Omey granite aureole, and the distribution of contact metamorphic minerals (sill = sillimanite). Calc-silicate localities shown are discussed in text. Inset map shows regional geology: Connemara schists, plain; migmatites and basic intrusions, dots; Galway Batholith, crosses; Ordovician and Silurian sediments, oblique lines. (Delaney Dome window (Leake *et al.*, 1983) not shown.)

(see also Tanner and Shackleton, 1979; Leake *et al.*, 1981).

Three formations crop out in the aureole (fig. 1). The Streamstown Formation consists mostly of semipelitic and psammitic schists with subordinate pelite. The typical regional assemblage is quartz-plagioclase (An_{30})-biotite-muscovite; 'index' minerals are conspicuously deficient even in the pelites although sparse garnet and fibrolite occur. In the succeeding Lakes Marble Formation clastics again predominate, with psammites being abundant towards the base and pelites more prevalent towards the top. As before, the typical assemblage is quartz-plagioclase-biotite-muscovite although garnet is better developed and the biotite : muscovite ratio is usually greater. Discontinuous impure marbles are found near the contact with the Streamstown formation and are repeated by folding along the well-exposed coastal section running south from the granite contact. Calcareous schists and granoblastites are also developed in the upper part of the formation. The Ballynakill Formation mostly comprises coarse-grained pelites. The typical regional assemblage is quartz-plagioclase-biotite-muscovite-garnet-staurolite-fibrolite ± kyanite; garnet and staurolite are often very abundant.

Contact metamorphic effects. Andalusite and cordierite are found up to 1.4 km from the granite contact with sparse thermal biotite beyond that for a further 500 m or so. Within the andalusite-cordierite zone over 95% of Ballynakill Formation samples studied in detail contain one or both minerals. Andalusite replaces regional staurolite, kyanite, fibrolite, and garnet and also occurs as idioblasts in the matrix. Cordierite replaces garnet and fibrolite but more often occurs as large poikiloblasts, now mostly pinitized. In the Lakes Marble Formation andalusite and/or cordierite occur in only 40% of the samples studied and in the Streamstown Formation this proportion drops to less than a quarter although, locally, andalusite grows profusely. In both formations andalusite occurs as large poikiloblasts accompanied by K-feldspar and, as the latter is all but restricted to these formations, there is little doubt that much of the andalusite is a product of the reaction

$$\text{muscovite} + \text{quartz} \rightarrow \text{K-feldspar} + \text{andalusite} + H_2O. \quad (1)$$

However, some pelite bands are very rich in andalusite and there is clearly insufficient K-feldspar to satisfy the molar proportions of reaction (1). We have not identified the reaction responsible for the 'excess' andalusite.

Average major-element analyses for the three formations within the aureole are given in Table I. The Ballynakill Formation is appreciably richer in Al_2O_3 and FeO, and poorer in SiO_2 and K_2O; this presumably reflects a higher clay content in the original sediment.

Corundum occurs in the Streamstown and Lakes Marble formations up to 250 m from the granite contact, and forms part of the assemblage muscovite + quartz + K-feldspar + corundum ± plagioclase ± biotite. The corundum-bearing pelites are relatively aluminous with an average Al_2O_3 content of 23.5% (mean of 7 analyses) compared with an overall average for the two formations of less than 20%. The mean silica content of the 7 analysed samples is 54.0%. This is somewhat lower than the overall average for the two formations (Table I) but the rocks can hardly be described as desilicated, especially as we have analysed a corundum-bearing psammite (no. 19576) with over 75% SiO_2! Corundum invariably nucleates within muscovite and only occurs in K-feldspar-bearing rocks. The reaction is

$$\text{muscovite} \rightarrow \text{K-feldspar} + \text{corundum} + H_2O. \quad (2)$$

In many samples the quartz in the rock is enclosed within aggregates of K-feldspar, presumably a product of reaction (1), and it seems that these were effective in isolating free silica from the system relevant to reaction (2).

The spatial distribution of corundum and andalusite is shown on fig. 1. Note that andalusite accompanies corundum throughout, thermal sillimanite developing only in isolated samples within a metre or so from the granite contact. This suggests that the reactions (1), (2), and the andalusite-sillimanite equilibrium can be satisfied only at pressures little more than 1 kbar, and with a maximum temperature at the contact of around 680 °C (fig. 2). There are several reasons to suppose that this pressure estimate is far too low. First, only a very H_2O-undersaturated magma could rise this high into the crust (Brown and Fyfe, 1970); such a magma would draw much water in from the (cool) country rocks, thus leading to a narrow aureole. Instead we find an aureole extending up to 2 km from the steep granite contact. The granite bears biotite, and locally hornblende, and the late

TABLE I. Whole-rock analyses of pelites/semipelites from the inner aureole

	Streamstown,15		Lakes Mb. (P),15		Lakes Mb. (C),18		Ballynakill,28	
	$\bar{x}$	$\sigma n^{-1/2}$	$\bar{x}$	$\sigma n^{-1/2}$	$\bar{x}$	$\sigma n^{-1/2}$	$\bar{x}$	$\sigma n^{-1/2}$
SiO_2	57.87	1.56	58.44	1.93	57.64	1.76	55.63	0.99
Al_2O_3	19.14	0.91	19.92	1.18	19.89	0.87	21.51	0.72
TiO_2	0.92	0.04	0.85	0.04	0.85	0.07	0.98	0.05
Fe_2O_3	2.32	0.51	1.57	0.33	1.23	0.19	2.21	0.28
FeO	4.24	0.40	4.68	0.21	4.96	0.36	6.15	0.24
MgO	2.59	0.11	2.20	0.21	2.50	0.14	2.48	0.08
CaO	1.67	0.29	1.60	0.28	2.45	0.33	1.71	0.21
Na_2O	2.93	0.16	2.23	0.24	2.61	0.17	2.27	0.15
K_2O	4.88	0.53	5.32	0.48	4.30	0.35	3.27	0.19
MnO	0.14	0.02	0.12	0.02	0.16	0.02	0.21	0.02
P_2O_5	0.20	0.01	0.19	0.01	0.16	0.19	0.21	0.04
H_2O^+	1.93	0.17	2.15	0.22	2.35	0.18	2.71	0.18
OR%	31.83	6.28	21.39	5.52	17.75	2.36	23.19	1.85
M/FM	0.53	0.03	0.44	0.02	0.47	0.02	0.42	0.01

Lakes Marble Formation divided into lower psammite-rich member (P) and upper calcschist-rich member (C) but analyses are pelites / semipelites free of calcsilicate phases other than An in plagioclase or Gr in garnet. Numbers after formation names are number of analyses, n; $\sigma n^{-1/2}$ is standard error. OR = oxidation ratio ; M/FM = mol. Mgo/(Feo + Mgo).

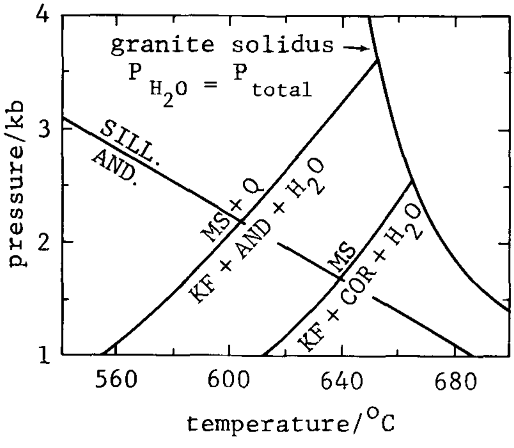

FIG. 2. Muscovite breakdown curves (reactions (1) and (2)) at $P_{H_2O} = P_{total}$, from Chatterjee and Johannes (1974); andalusite–sillimanite equilibrium from Holdaway (1971); granite solidus from Boettcher and Wyllie (1968).

magmatic stages include biotite and hornblende lamprophyres and the occurrence of marginal breccia pipes. None of these is consistent with a very H_2O-deficient magma. Secondly the Q–Or–Ab proportions in the granite indicate crystallization at water vapour pressures between 2 and 4 kbar with a best estimate (based on three rocks with $Q+Or+A > 90\%$) of between 2 and 3 kbar (fig. 3).

In the next section a *P–T* diagram for the inner aureole of the Omey granite is developed in which P_{H_2O} is not assumed equal to P_{total}, and which also shows the effect of overstepping on the andalusite–sillimanite equilibrium. Combined with information on the calc-silicate assemblages in the Lakes Marble Formation, the diagram is then used to show how the spatial relations of the aureole minerals can be satisfied.

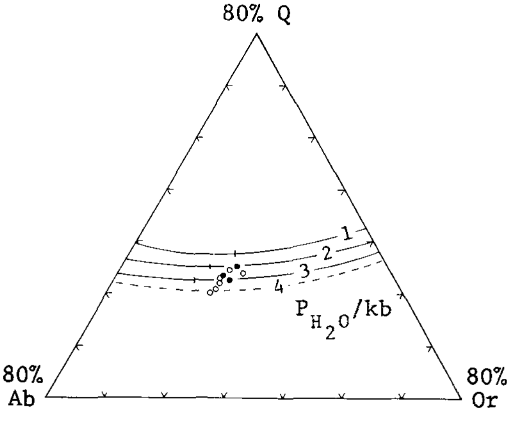

FIG. 3. Normative Q-Ab-Or plot of nine samples from the Omey granite (taken from Townend 1966, table 7) with $Q+Ab+Or > 80\%$ (or $> 90\%$, solid symbols). Vapour pressure curves and isobaric minima (short vertical lines) based on the experimental system $NaAlSi_3O_8$–$KAlSi_3O_8$–SiO_2–H_2O from Luth *et al.* (1964).

P, T relations for the inner aureole. The pure end-member reactions corresponding to (1) and (2), viz.

$$KAl_2Si_3AlO_{10}(OH)_2 + SiO_2 \rightarrow KAlSi_3O_8 + Al_2SiO_5 + H_2O \quad (3)$$

and

$$KAl_2Si_3AlO_{10}(OH)_2 \rightarrow KAlSi_3O_8 + Al_2O_3 + H_2O \quad (4)$$

are well bracketed between 1 and 5 kbar with $P_{H_2O} = P_{total}$ (Chatterjee and Johannes, 1974). Evaluating ΔG^0 for each experimental bracket,

$$\Delta G^0 = -RT\ln K - \int_1^P \Delta V_s dP, \quad (5)$$

linear regression analyses yield the equations shown on fig. 4, from which we obtain $\Delta H^0 = 19.92$ kcal, $\Delta S^0 = 0.0365$ kcal K^{-1} for reaction (3) and $\Delta H^0 = 23.59$ kcal, $\Delta S^0 = 0.0393$ kcal K^{-1} for reaction (4). The ΔH^0 and ΔS^0 values are then used to calculate the *P*, *T* equilibria for $P_{H_2O} < P_{total}$ as shown in fig. 5. In evaluating equation (5) we assumed $K = f_{H_2O}$ and also that the difference in standard molar volumes is independent of *P* and *T* so that the last term can be calculated as $\Delta V_s^0 (P-1)$. Water vapour fugacities were taken from Burnham *et al.* (1969) and molar volumes from Chatterjee and Johannes (1974, table 6).

For natural muscovites and K-feldspars with Na in solid solution the equilibrium constant should be calculated as

$$K = f_{H_2O}(\gamma_k^{kf} x_k^{kf})/(\gamma_k^{ms} x_k^{ms}).$$

Assuming that γ_k^{ms} and γ_k^{kf} are both very close to unity (Cheney and Guidotti, 1979; Waldbaum and Thompson, 1969) analysis of coexisting muscovites and K-feldspars in two inner aureole samples (19610 and 19681) yields $K \simeq 0.9 f_{H_2O}$. Using these *K* values displaces the curves on fig. 5 to lower temperatures by about 10 °C, but we prefer to present curves for pure reactants and products because our muscovite analyses yielded low totals and are probably unreliable. It should also be noted that the rocks bear coexisting plagioclase although Evans and Guidotti (1966) have shown that the partitioning of Na between K-feldspar and plagioclase is expected to have a negligible effect on the reactions.

The andalusite–sillimanite curve shown on fig. 5 is that of Holdaway (1971). This experimental

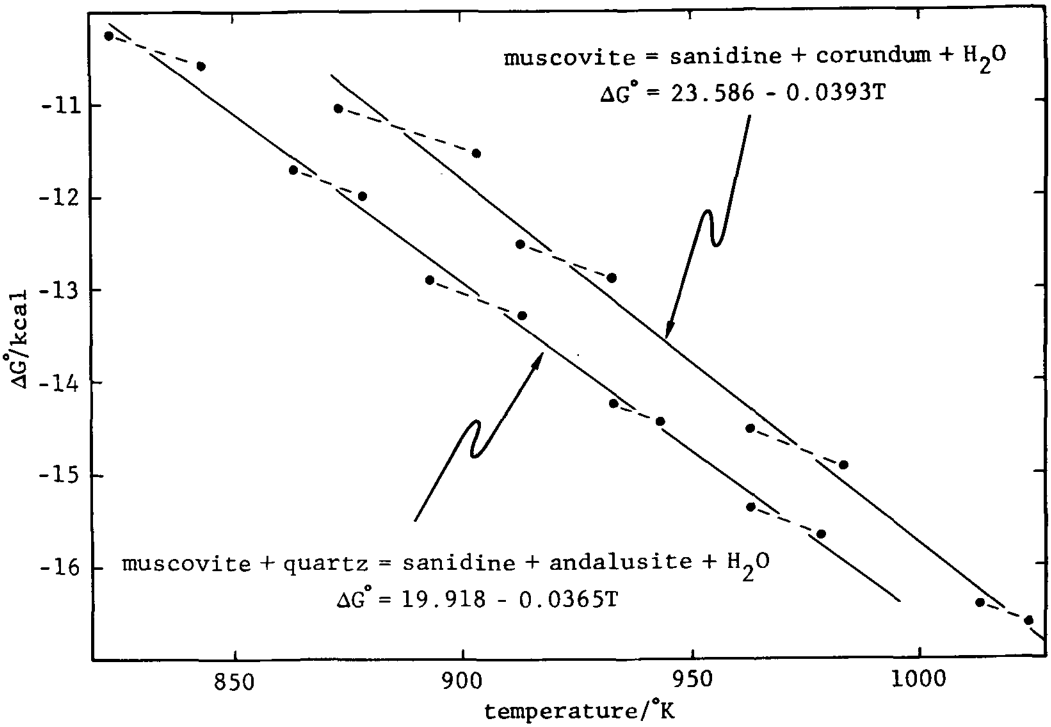

FIG. 4. Standard free energy change (ΔG°) for reactions (3) and (4) versus temperature, based on experimental brackets of Chatterjee and Johannes (1974). Linear regression equations as shown.

equilibrium is preferred over others because it is consistent with natural near-equilibrium kyanite–andalusite–sillimanite parageneses in New Mexico that are independently constrained by several other geothermometers and barometers (Grambling, 1981). All three polymorphs in this area have small and nearly equal Fe^{3+} contents so that partitioning of Fe^{3+} can also be ruled out as an important factor. When the Holdaway andalusite–sillimanite equilibrium is overstepped by a small amount the curve is displaced as shown in fig. 5. The overstep curves were calculated simply by adding the amount shown on the curves to the change in enthalpy of formation term in the equation

$$\Delta(\Delta H) - \Delta ST + \Delta VP = 0$$

and recalculating the temperature. We have used values for the equilibrium changes in ΔH, S, and V which accord with the Holdaway (1971) curve, although these differ somewhat from the determinations of Robie and Waldbaum (1968).

Also shown on fig. 5 are curves for the first appearance of wollastonite + grossularite (Wo + Gr) in the presence of calcite at $X_{H_2O}^{fluid}$ equal to 0.85 and 0.9, and the first appearance of wollastonite + grossularite + anorthite (Wo + Gr + An). These curves, based on the work of Storre are taken from Winkler (1978).

Interpretation. The assemblage Wo + Gr + Cc is common in the Lakes Marble Formation and is found up to 470 m from the granite contact. This locality (shown on fig. 1) may not be the 'first appearance' because suitable lithologies are not found further out, but it is probably close because wollastonite is very sparsely developed relative to locations nearer to the contact. The assemblage muscovite + quartz + K-feldspar + andalusite + plagioclase is found at the same locality and, as K-feldspar is found replacing the Ab-rich cores of regional plagioclase, the probable reaction is

$$\text{ms} + \text{qtz} + \text{plag} \rightarrow \text{Kfsp} + \text{and} + H_2O. \quad (6)$$

Thus the theoretically univariant reaction (1) is at least divariant here, although Evans and Guidotti (1966) show that Na partitioning has very little effect on the equilibrium. Although andalusite is abundant at greater distances from the granite (in the Ballynakill Formation) it does not occur with K-feldspar. We consider that the first appearance

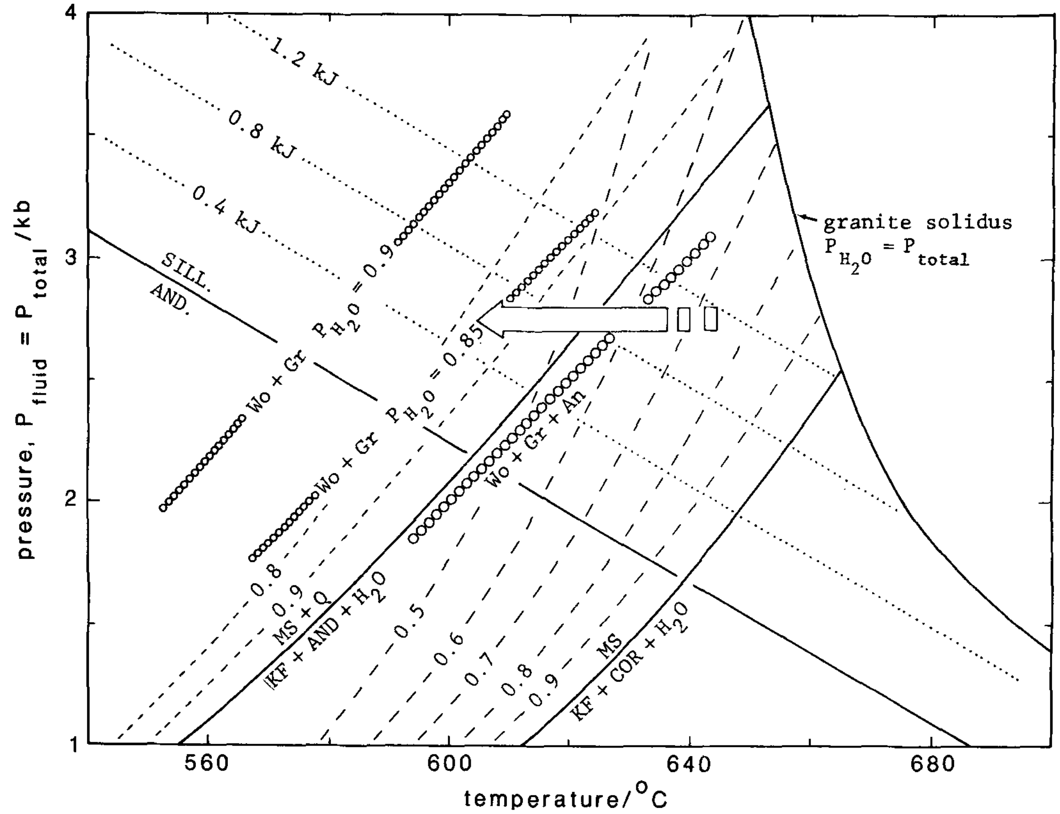

FIG. 5. P–T grid for the inner aureole. Solid curves as in fig. 2; curves for $P_{H_2O} < P_{total}$ derived from fig. 4 regressions; displaced andalusite–sillimanite curves result from overstepping the reaction by amount shown on curves (see text). Calc-silicate curves from Winkler (1978). Bold arrow shows preferred range of metamorphic conditions but is towards the upper limit of pressure consistent with the observations.

of andalusite *as a product of reactions (1) or (6)* is also at about 470 m from the contact. The temperature at which the assemblage Wo + Gr + Cc first stabilizes increases with decrease in $X_{H_2O}^{fluid}$ while that of ms + qtz + Kfsp + and decreases. Assuming $X_{H_2O} + X_{CO_2} = 1$, both reactions occur at about the same P, T conditions (600–615 °C, 2.5–3.0 kbar) when $X_{H_2O} \simeq 0.85$ (fig. 5). This vapour pressure estimate is similar to that derived independently from the granite composition (fig. 3). It is known that the reaction kinetics of Cc + Qtz → Wo + CO_2 are rapid, which suggests that the reaction is an effective fluid phase buffer (Tanner *et al.*, 1983). The calc-silicate assemblages are found closely interfolded with the pelites and, as convective circulation of fluid probably occurred in the aureole (see later), the fluid composition may well have been similar in both lithologies. At the very least it is unlikely that X_{H_2O} could be less than about 0.85 in the pelitic rocks.

The assemblage Wo + Gr + An is found 140 m from the granite contact in a large marble/calc-silicate lens exposed on the coastal section (fig. 1). Again, this may not be a 'first appearance' because the assemblage is restricted to a very narrow range of rock compositions. More to the point, the assemblage requires a *minimum* X_{H_2O} of 0.8 at 2 kbar (and a minimum only slightly smaller at 3 kbar) but, given this minimum, the equilibrium (fig. 5) does not shift with increasing X_{H_2O} (Winkler, 1978).

Corundum is found up to 250 m from the contact; that is, over 100 m further out than the Wo + Gr + An locality and presumably at somewhat lower temperatures. The inescapable conclusion is that reaction (2) must have equilibrated with X_{H_2O} *no greater than* 0.5 at 2.5 kbar (or 0.6 at 3 kbar), and at a temperature around 620 °C. If the pressure was as low as 2 kbar the equilibrium would require $X_{H_2O} \simeq 0.4$ and a temperature less than 600 °C.

Andalusite persists as the Al_2SiO_5 phase in the aureole right up to the contact although rare prismatic sillimanite is found within a metre or so of the contact. The equilibrium relationships (fig. 5) can only be satisfied above 2 kbar if the andalusite-sillimanite equilibrium is overstepped by up to 1 kJ or so, depending on the pressure. Probably, andalusite nucleated easily and grew rapidly during the initial heating of the envelope, and continued to grow into the stability field of sillimanite. This would be consistent with the observed profuse growth of large inclusion-ridden andalusites in some pelitic layers, and with the experimental observation that the quartz-muscovite dehydration reaction is sluggish due to the slow growth rate of sillimanite (Evans, 1965). In fact prismatic sillimanite is found at the same locality treated as the outer limit of Cc + Wo + Gr (fig. 1). The sillimanite occurs as radiating sprays of prismatic crystals within a quartz-feldspar vein. We have interpreted this occurrence as regional in origin because some sprays of regional fibrolite coarsen distally to take on a prismatic appearance. Nevertheless, this vein occurrence is morphologically distinct and may be thermal in origin. If so it suggests that, given a suitable nucleation and growth environment, sillimanite first enters at 470 m from the contact. A 'best' pressure estimate would then be about 2.3 kbar.

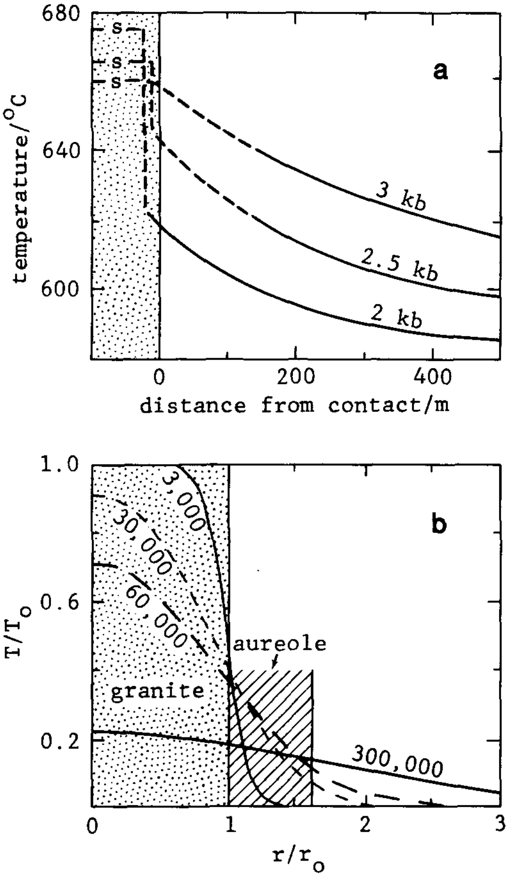

FIG. 6. (*a*) Thermal structure of inner aureole based on distributions of contact metamorphic minerals and the *P–T* grid (fig. 5); S denotes granite solidus at various pressures. Best estimate of pressure is 2.5 ± 0.25 kbar. (*b*) Simple model of conductive cooling (Carslaw and Jaeger, 1959, ch. II) around a granite intruded at temperature T_g into country rocks at temperature T_c. $T_o = T_g - T_c$ is the initial temperature difference. The granite is modelled as a vertical cylinder of radius r_o = 3.25 km. Curves show dimensionless temperature variation with radial distance at the times indicated (in years). Aureole shown out to 2 km from granite contact.

Discussion

The graphical relationships presented in fig. 5 suggest a thermal structure for the Omey granite aureole as shown in fig. 6*a*. In spite of the uncertainty in pressure estimate there is no doubt that the maximum temperature reached was a large fraction of the granite solidus temperature, with the temperature falling off at about 80 °C per kilometre. An idealized model of purely conductive heat loss around a granite body, modelled as a vertical cylinder of radius 3.25 km, is shown in fig. 6*b* (see Carslaw and Jaeger, 1959, ch. II). T_o is the difference in initial temperature between the cylinder and the country rocks (assuming instantaneous emplacement); the curves represent the dimensionless temperature variation with radial distance at the times shown (in years) on the curves. Thermal diffusivity is taken as 1.1×10^{-6} m^2/s for both granite and country rocks. It is clear that for any reasonable choice of initial temperatures for the intrusion and its envelope, shallow thermal gradients can occur only after a substantial time, by which time the temperatures would be far too low to satisfy the mineralogy. In short the thermal structure implied by fig. 6*a* is totally incompatible with purely conductive heating and cooling of the envelope. The thermal structure associated with *convective* heat transport is much more difficult to model and is likely to be especially complex near the top of a vertical cylindrical intrusion. Numerous blocks of Streamstown and Lakes Marble lithologies found in the breccia pipes shown on fig. 1 suggest that the intrusion top was, in fact, not much above the present level of erosion. In spite of the likely complexities the models of Norton and Knight (1977) show clearly that heat flux due to convection can far exceed that due to conduction, and that temperatures close to the intrusion

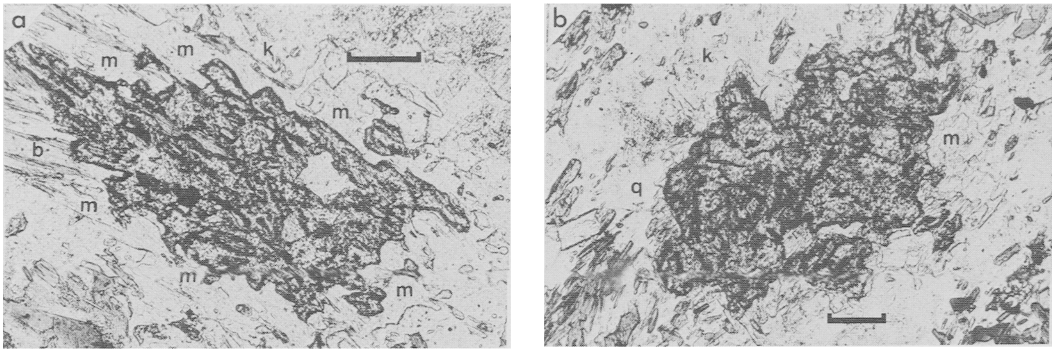

FIG. 7. (*a*) Corundum grown entirely within single muscovite crystal (m); sample 19569, (*b*) Large corundum breaching margin of host muscovite, fine-grained muscovite developed between corundum and quartz (q) in matrix; sample 19040. k = K-feldspar, b = biotite. Scale bars 0.1 mm.

temperature can persist for considerable distances. There seems little doubt that the thermal structure of the Omey granite aureole mostly reflects convective heat transport. This in turn supports our earlier suggestion that the fluid composition would be largely homogenized between pelitic and marble/calc-silicate layers.

The paradox remains that the spatial distribution of corundum in the aureole can only be explained if the H_2O fugacity controlling its growth was much less than that controlling the other reactions. For P_f somewhere between, say, 2.3 and 2.8 kbar, X_{H_2O} for reaction (2) must have been less than 0.6 (and perhaps as low as 0·4) whereas the calc-silicate assemblages and reaction (1) together require $X_{H_2O} \simeq 0.85$. We think it important that corundum in the Omey aureole always grows within (and usually entirely within) large muscovite crystals (fig. 7*a*). This suggests to us that f_{H_2O} *within* a muscovite crystal may not be closely related to f_{H_2O} of the grain-boundary fluid phase in the surrounding rocks. Indeed, where large corundum grains breach the margin of their host muscovite crystals, they are invariably mantled by fine-grained muscovite (fig. 7*b*). This suggests that once the corundum comes into contact with the grain-boundary fluid (with $X_{H_2O} \simeq 0.85$) the muscovite breakdown reaction is immediately reversed to yield a mantle of fine muscovite, thus isolating the corundum once more.

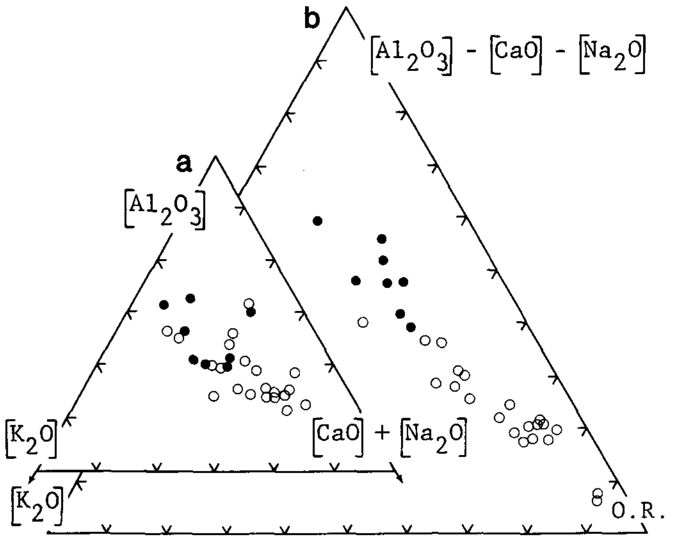

FIG. 8. Triangular diagrams showing relative mole proportions as indicated for analysed rocks in the inner aureole. Corundum bearing rocks shown as solid symbols. O.R. = Oxidation Ratio = $2Fe_2O_3/(2Fe_2O_3 + FeO)$.

This concept is not entirely novel because it is well-established that porphyroblasts can act as a seal preventing reactions which would otherwise have eliminated minerals included within them; interesting examples of this phenomenon are described by Zen (1963), Evans and Guidotti (1966), and Rosenfeld (1970). Our work suggests that large muscovites can act as a seal in the same way, and that the breakdown reaction yielding corundum is controlled by an internal effective H_2O fugacity (however that may be defined) different from that in the surrounding rock. Consequently reaction (2) can run at temperatures substantially lower than otherwise expected; corundum in pelitic rocks need imply neither desilication nor especially high temperatures.

None the less, it is clear that the growth of corundum *within* muscovite crystals must set up a flux of $(OH)^-$ and K^+ (or some complex thereof) out of the host. The corundum may be 'sealed off' but the progress of the reactions may still be controlled by the external system. Granitic magma must be somewhat H_2O-unsaturated to rise in the crust so that, initially at least, water will be drawn in from the envelope rocks. Flow of H_2O up a thermal gradient (towards the intrusion) will promote oxidation and the increase in f_{H_2O} with increasing temperature (at constant pressure) may inhibit flux of $(OH)^-$ out of muscovite crystals, thus retarding the reaction. Conversely, once a convective circulation pattern is established, flow of H_2O away from the intrusion will promote reduction and the decrease in f_{H_2O} may assist the progress of the muscovite breakdown reaction. It is particularly interesting, then, that although corundum-bearing rocks in the aureole are all rather aluminous (relative to $CaO + Na_2O$), there is no clean separation of rocks with and without corundum on an Al_2O_3-K_2O-$(CaO + Na_2O)$ plot (fig. 8*a*). There is, however, a much more distinct separation with respect to oxidation ratio (fig. 8*b*). The irregular distribution of corundum probably reflects an inhomogeneous pattern of convective fluid transport, promoted perhaps by fluid having to move across the strike of strongly foliated rocks. An irregular flow pattern is also suggested by the erratic spatial distribution of whole-rock oxidation ratio (and green biotite) within the aureole (fig. 9) although it is not known how much of this variation is inherited from the regional metamorphism. Note, however, that of the seventeen analysed rocks within 250 m of the granite contact, those with the seven lowest oxidation ratios are all corundum-bearing.

It should also be noted that the prevalence of corundum along a coastal strip is not wholly due to good exposure for other parts of the inner aureole have also been sampled densely. The Wo + Gr + An assemblage found in the same coastal strip (fig. 1) is part of a marble lens in which spectacular grossularite, vesuvianite, wollastonite, diopside, epidote, and sphene assemblages develop. Some bands up to 20 cm thick are almost pure grossularite-vesuvianite. This lens has long been interpreted as a skarn (Townend, 1966; Cobbing, 1969), presumably involving the introduction of Fe, Al, Si, etc., derived from the granite. Its chemistry is certainly distinct from marbles at the same horizon elsewhere in the area which, while somewhat sandy, are otherwise fairly pure calcite rocks. It seems likely that the coastal strip represents a diffuse channelway along which fluid was carried from the granite, thus producing the skarn and inducing the reducing conditions favourable to corundum growth.

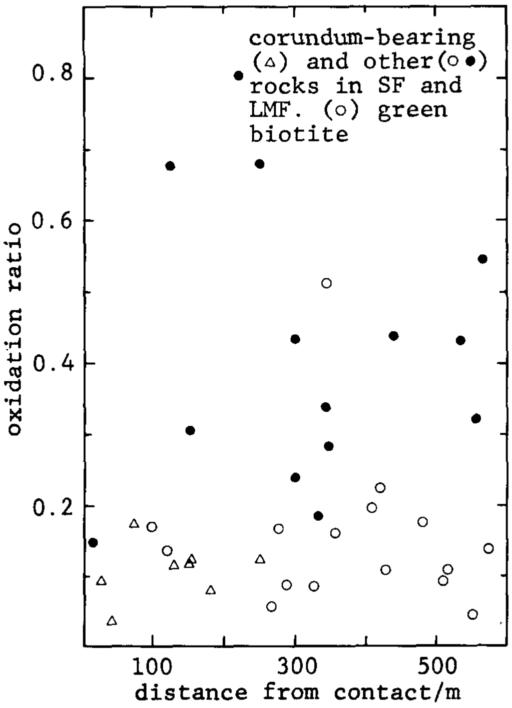

FIG. 9. Oxidation Ratio of Streamstown and Lakes Marble Formation rocks in inner aureole plotted against distance from granite contact.

Our main conclusion—that corundum must have grown in a microsystem with f_{H_2O} quite different from that pertaining to the rock as a whole—has obvious and important implications for the interpretation of natural parageneses. It also raises theoretical questions about the meaning of vapour fugacity within a crystal lattice. Unfortunately the micromechanics of nucleation and growth of one crystal within another are poorly understood. Detailed studies are long overdue for the problem has been attracting attention for over 300 years—'. . . both the number and length of the sides are variously changed without change in the angles, and in the very midst of the Chrystal there are left various cavities, & formed various plates' (Steno, 1671).

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